AGENDA ITEM 2.5

Developments in isotope enrichment techniques
and trends in costs for enrichment services

Innovations dans les méthodes de séparation des isotopes
et évolution des coûts de l'enrichissement

Разработка методов изотопного обогащения и тенденции
изменения стоимости обогащения

Progresos de las técnicas de enriquecimiento isotópico
y evolución del costo de los servicios de enriquecimiento

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THE SEPARATION NOZZLE PROCESS FOR ENRICHMENT OF URANIUM-235

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Abstract–Résumé–Аннотация–Resumen

THE SEPARATION NOZZLE PROCESS FOR ENRICHMENT OF URANIUM-235.

The separation nozzle process is based on pressure diffusion in a UF_6/He or UF_6/H_2 jet flowing along a curved wall. At Karlsruhe (Federal Republic of Germany), a ten-stage pilot plant has been working successfully since November 1967. A full-scale separating stage with a capacity of 1700 kg SWU/year was built and is in permanent use for testing different types of element separation. The construction of this stage is described.

Calculations based on preliminary tenders from industry indicate that the specific investment costs of separation nozzle plants will be considerably less than those of diffusion plants of equal capacity. In the present state of development, the specific energy consumption is still higher by a factor of approximately 1.6 as compared with the diffusion process. However, current theoretical and experimental studies of the separation nozzle effect show that a considerable reduction of the specific energy consumption is within the realm of possibility. The general scope of these investigations is presented.

In March 1970, the Gesellschaft für Kernforschung, Karlsruhe, and STEAG, Essen, signed a contract on co-operation in the separation nozzle process. The purpose of this co-operation is to provide further development, industrialization and practical application of the process. All prerequisites for starting the construction of a plant that would work economically with a net output of 500 000 - 1 000 000 SWU/year are expected to be fulfilled in 1974 at the latest. The main features of such a plant are discussed.

LE PROCEDE DE SEPARATION PAR TUYERES POUR L'ENRICHISSEMENT DE L'URANIUM-235.

Le procédé de séparation par tuyères utilise la diffusion dans le gradient de pression d'un jet d'UF_6/He ou d'UF_6/H_2 obtenu par détente du mélange gazeux le long d'une paroi courbe. Depuis le mois d'octobre 1967 une installation pilote à 10 étages fonctionne de manière satisfaisante à Karlsruhe (République fédérale d'Allemagne). Un étage de séparation avec une capacité de 1700 kg de travail de séparation par an a été construit et mis en service. Cet étage est utilisé actuellement pour essayer différents types d'éléments de séparation. La construction de cet étage est décrite.

Les calculs basés sur les offres préliminaires de l'industrie montrent que les coûts d'investissement des installations utilisant le procédé de séparation par tuyères seront nettement inférieurs aux coûts d'investissement des installations de diffusion gazeuse de même capacité. Au stade actuel du développement la consommation spécifique d'énergie dépasse encore d'un facteur d'environ 1,6 celle du procédé de diffusion gazeuse. Cependant, les études théoriques et expérimentales en cours sur l'effet de séparation par tuyères permettent d'envisager une réduction considérable de la consommation spécifique d'énergie. Les principaux résultats de ces études sont communiqués.

En mars 1970 la Gesellschaft für Kernforschung, Karlsruhe, et la STEAG, Essen, ont conclu un contrat de coopération dans le domaine du procédé de séparation par tuyères. Cette coopération a pour but le développement futur, l'industrialisation et l'application industrielle de ce procédé. Il est prévu que l'ensemble des conditions nécessaires pour la construction d'une installation d'une capacité de 500 000 à 1 000 000 kg de travail de séparation par an devrait être établi en 1974 au plus tard. Le mémoire décrit les principales caractéristiques d'une installation de ce genre.

ОБОГАЩЕНИЕ УРАНА-235 С ПОМОЩЬЮ ГАЗОВОГО СОПЛА.

Процесс разделения изотопов с помощью газового сопла основывается на принципе пьезодиффузии в струе UF_6/He или UF_6/H_2, проходящей вдоль выгнутой стенки. С ноября 1967 года в Карлсруэ (Федеративная Республика Германии) успешно работает десятикаскадная опытная установка. Построен в натуральную величину разделительный каскад производительностью 1700 кг единиц разделительной работы в год, который постоянно используется для испытания различных методов разделения элементов. Описывается процесс создания этого каскада.

Расчеты, основанные на предварительных заявках, поступивших от представителей промышленности, показывают, что удельные капиталовложения в сопловые разделительные установки будут гораздо ниже, чем те же вложения в диффузионные установки равной мощности. На современной стадии разработки удельное потребление энергии все еще выше, примерно в 1,6 по сравнению с процессом диффузии. Однако текущие теоретические и экспериментальные исследования производительности при разделении методом газовой струи показывают, что значительное сокращение удельного расхода энергии вполне возможно. В общих чертах изложен характер этих исследований.

В марте 1970 года Общество по ядерным исследованиям, Карлсруэ, и "СТЕАГ", Эссен, подписали контракт о совместной работе по разделению с помощью газового сопла. Целью этого сотрудничества является обеспечение дальнейшей разработки, внедрение в производство и практическое применение процесса. Все предварительные работы для начала сооружения установки, которая работала бы экономично с чистой производительностью 500 000 – 1 000 000 единиц разделительной работы в год, как ожидают, закончатся самое позднее в 1974 году. Обсуждаются основные характеристики такой установки.

PROCESO DE SEPARACION POR TOBERA PARA EL ENRIQUECIMIENTO DEL URANIO-235.

El proceso de separación por tobera se basa en la difusión a presión de un chorro de UF_6/He o UF_6/H_2 a lo largo de una pared curva. En Karlsruhe (República Federal de Alemania) este proceso se viene desarrollando con éxito desde noviembre de 1967 en una planta piloto de diez etapas. Se preparó una etapa de separación a escala industrial con una capacidad de 1700 kg SWU/año, que está en uso permanente para ensayar diferentes tipos de séparación de elementos. La construcción de esta etapa se describe en la memoria.

Los cálculos basados en ofertas preliminares de la industria indican que los costos de inversión específicos de las plantas de separación por toberas serán considerablemente menores que los costos de las plantas de difusión de igual capacidad. En el estado actual de desarrollo, el consumo específico de energía es todavía 1,6 veces mayor aproximadamente que en el proceso de difusión. No obstante, los actuales estudios teóricos y experimentales del efecto de separación por tobera demuestran que entra en lo posible una reducción considerable del consumo de energía específica. Se indica el alcance general de estas investigaciones.

En marzo de 1970, la Gesellschaft für Kernforschung de Karlsruhe y la STEAG de Essen firmaron un contrato de cooperación para la separación por toberas. Esta cooperación persigue un mayor desarrollo, una mejor industrialización y más posibilidades prácticas del proceso. Se espera que en 1974 a más tardar se den todas las condiciones previas para iniciar la construcción de una planta que funcione económicamente con una producción neta de 500 000 a 1 000 000 SWU/año. Se examinan las características principales de dicha planta.

1. PHYSICAL BASIS

At the Karlsruhe Nuclear Research Centre, the separation nozzle process for uranium enrichment was developed on a semi-industrial scale [1 - 5]. Figure 1 shows the cross-section of the typical slit-shaped nozzle system for the process.

A mixture of about 5 mol.-% UF_6 and 95 mol.-% helium or hydrogen is forced to flow along a curved wall. Subsequently, a knife edge is employed to divide the gas stream into two fractions which are pumped off separately. The deflection of the jet by the curved wall results in a partial spatial separation of the components, the gas moving close to the deflecting wall becoming enriched in the heavy isotope, while the light iso-isotope is deflected away from the wall.

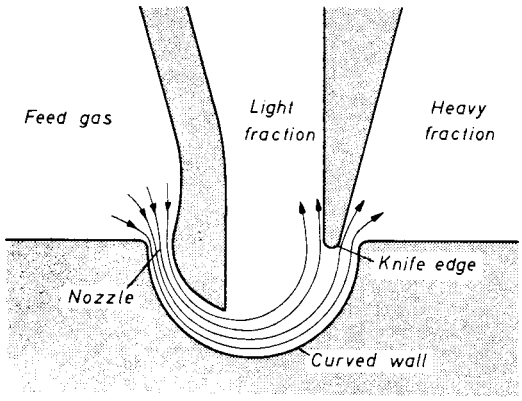

FIG. 1. Cross-section of the slit-shaped separation nozzle system with a schematic representation of the stream-lines.

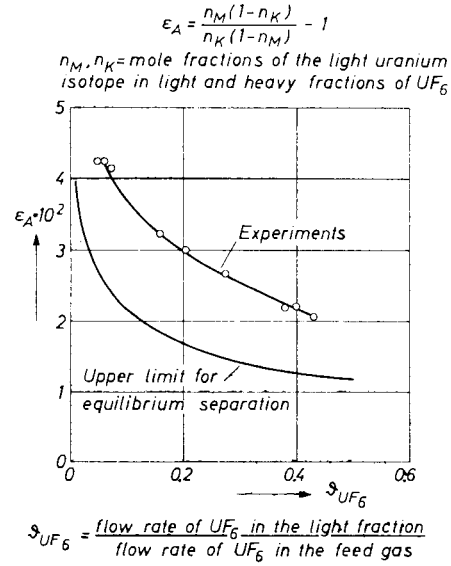

FIG. 2. The upper curve indicates the experimentally determined relationship between the elementary effect ϵ_A of the separation of uranium isotopes and the UF_6-cut ϑ_{UF_6} (mixture of UF_6/H_2 with 1.6 mol.-% UF_6; expansion ratio 8). The lower curve gives the upper limit for equilibrium separation calculated for centrifugal flow with an infinite velocity.

Figure 2 shows the experimentally determined relationship between the elementary effect ϵ_A of the separation of uranium isotopes and the UF_6 cut, ϑ_{UF_6}. For these experiments, a mixture consisting of 1.6 mol.-% UF_6 and 98.4 mol.-% H_2 was used at an expansion ratio of 8. With the lowest cut investigated, the elementary effect is 4.2%, i.e. about ten times larger than for the diffusion process. Under conditions of practical application, this factor is reduced to 3 or 4.

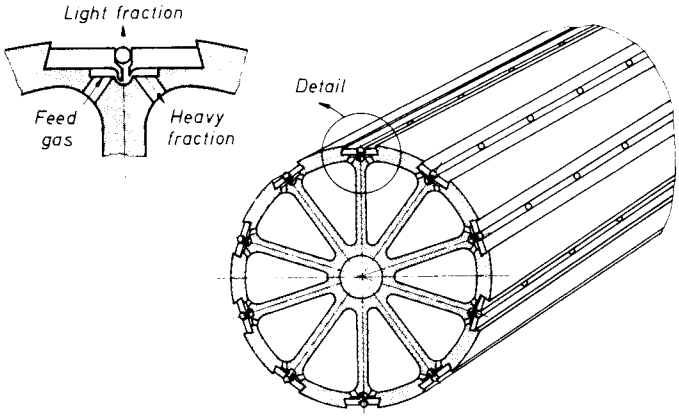

FIG. 3. Tubular separation element with 10 separating slits.

The mass-dependent centrifugal force generated in the curved flow may be regarded as a main cause of separation. The main effect of the added light gas was expected to be the increased flow velocity reached by a gas of lower average molecular weight. Figure 2, however, indicates that the experimentally determined separation effects are considerably in excess of those calculated for equilibrium separation for centrifugal flow with an infinite velocity. In accordance with present calculations, the additional separation effect can be explained mainly by the added gas causing a delay in establishing the hypsometric density distribution of the UF_6 as a whole. This delay results in a transistory reduction of the mixing diffusion of the uranium isotopes.

2. SEPARATION ELEMENT

In the separation nozzle process, the optimum pressure of the feed gas is inversely proportional to the characteristic dimensions of the slit-shaped separation nozzle systems [1]. Since, for economic reasons, high inlet pressures are desirable, separation nozzle systems with particularly small characteristic dimensions should be used. Figure 3 shows how these nozzles are arranged as tubular separation elements. Ten slit-shaped separation nozzle systems are arranged on the surface of an aluminium tube with an outside diameter of about 10 cm. The inside of the tube is divided into ten channels by means of partitions. Five of these channels are used as inlets for the feed gas and five for removing the heavy fraction. The light fraction flows radially outward into a tank encasing the separation elements.

Several methods for inexpensive mass production of separation nozzle systems with optimum pressures between 400 and 600 mm Hg have been developed in co-operation with industry.

In the separation element shown in Fig.3, the separation nozzle systems are made of aluminium strips pressed into dovetails within the tube.[1] The separation tubes supplied by industry produce, within the

[1] Manufactured by Messerschmitt-Bölkow-Blohm GmbH, Munich.

FIG. 4. Metal foil with separation nozzle structures applied by means of photo-etching.

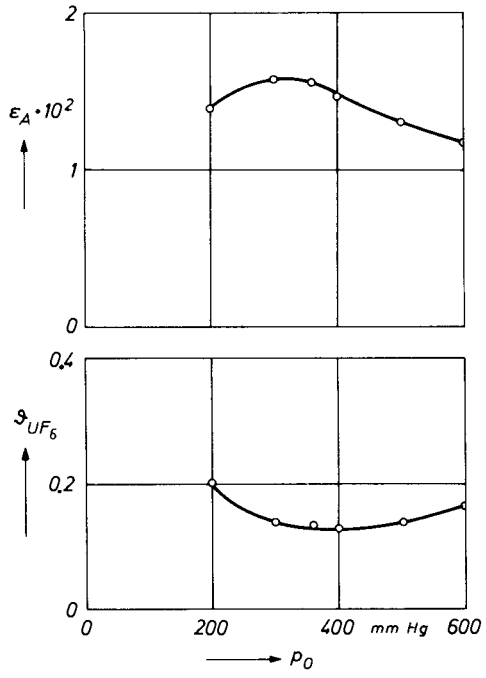

FIG. 5. Elementary effect of uranium isotope separation ϵ_A and UF_6-cut ϑ_{UF_6} as a function of inlet pressure p_0 measured with the stack of foils shown in Fig. 4. (5 mol. -% UF_6, 95 mol. -% He, expansion ratio 4).

limits of error, the separation effects generated oy laboratory-scale elements. The price of a mass-produced element, as determined by the supplier on the basis of two years of experience with this type of element, indicates that a specific investment for the separation elements of less than US $16/kg of separative work per annum can be expected.

In addition to mechanical means, various methods developed in the field of micro-electronics may be employed for the fabrication of separation elements. Figure 4 shows a metal foil with separation nozzle structures produced by means of photo-etching [2]. Stacking such foils will produce separation nozzle systems of an extremely high separative capacity per unit volume. Figure 5 gives the measurements, in such a

[2] Manufactured by Siemens AG.

stack of foils, of the elementary effect of uranium isotope separation and the uranium cut as a function of inlet pressure. In this case also, the results agree within the limits of error with those obtained on laboratory-scale elements. The output to be expected from stacks of foils applied to tubular separation elements is between 20 and 40 kg of separative work per annum and metre of tube length.

3. SEPARATION STAGE

In the technical application of the process, a large number of separation element tubes are installed in a common separation element tank. This tank, together with a compressor and a cooler, constitutes the separation stage. At Karlsruhe, an industrial scale separation nozzle stage has been set up in collaboration with industry. It will have a separation capacity of about 2000 kg of seperative work per annum at full power, i.e. roughly one third of the separation capacity of the largest American diffusion stages.

Figure 6 shows the cross-section of the stage designed to hold 81 tubular separation elements. The stage comprises the tank for the separation elements, a crosspiece for gas distribution, a combination of intermediate and final gas coolers, a two-stage radial compressor, and a motor rigidly coupled to the compressor. The overall height of the unit is about 7 m.

As the stage has a very high throughput, it can be tested in short-circuit operation only. This means that the gas sucked in by the compressor from the tank passes through a loop and is fed into the crosspiece at the point where this stage would accept its feed gas from another stage when working within a separation cascade. The feed gas is transferred via the crosspiece to the separation tubes, where it is divided into the light and heavy fractions. The light fraction is collected within the tank, while the heavy fraction arrives at the cover. In the cascade system, the heavy fraction would be transferred to another stage via the tube located in an eccentric position in the cover, while the central tube would allow the inflow of the respective fraction from another stage. In short-circuit operation, the heavy fraction is recycled to the stage via the upper loop. The light and the heavy fractions are combined in the mixing diffusor installed between the supporting plates of the separation elements. This means that feed gas is again available in the suction pipe of the compressor.

The lower loop is equipped with a Venturi tube for gas flow measurements, a throttle valve, and the connection for the vacuum system. To it is connected the separation element test loop used for testing separation elements individually. In the upper loop, an additional throttle valve is installed, which allows adjustment of the UF_6 cut of the stage.

The two-stage radial compressor with a capacity of 100 000 m^3/h at a compression rate of 4 was developed and built by Hispano-Suiza (France). A suitable 300-cycle motor producing 150 rev./s was provided by Siemens.

The process gas heated at the first and second stages of the radial compressor is cooled to a suction temperature of 40°C in the intermediate and final coolers. These consist of finned tube bundles arranged concentrically around the suction pipe and housed in a common cylindrical

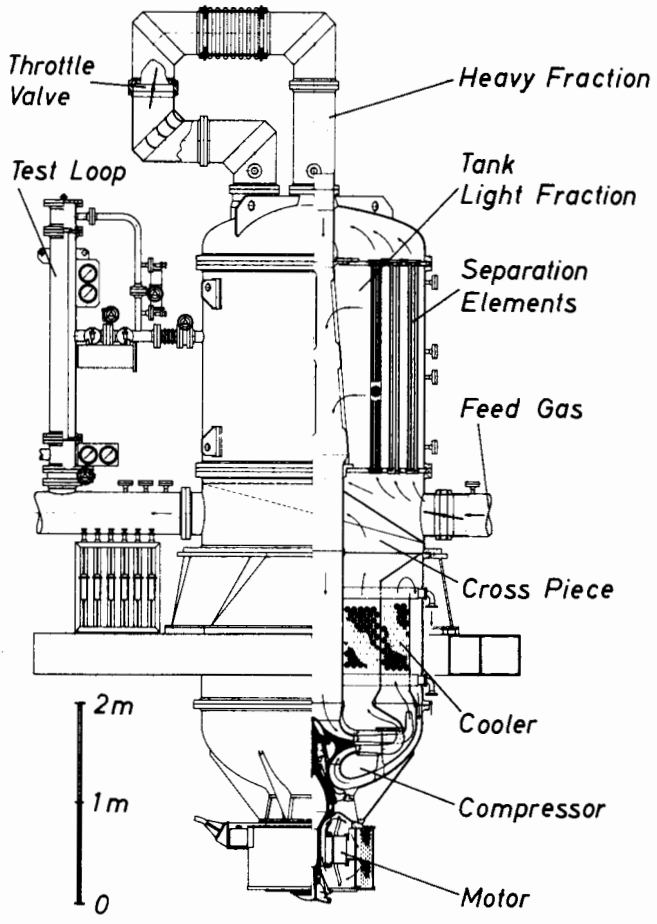

FIG. 6. Cross-section of the separation nozzle stage with a rated separative work capacity of about 2000 kg per annum.

casing of the separation stage. Two different coolers, provided by SNECMA and GEA, are at hand. The casing and piping of this stage were delivered by Leybold-Heraeus.

Figure 7 shows the upper part of the separation stage, including the tank for the separation elements, the separation element test loop and the two short-circuit loops for the feed gas and the heavy fraction, respectively. Figure 8 shows the lower part during assembly of the compressor. The two-stage cooler can be recognized in the upper part of the picture.

In July 1970, the lower part of the stage, i.e. the combination of the compressor and the cooler, started operation with a UF_6 -He mixture. It passed a four-week endurance test without significant failures. At present, the stage is continuously used for testing individual separation element tubes delivered by industry. Starting in mid-1972, the stage is to be operated continuously with a complete separation element charge.

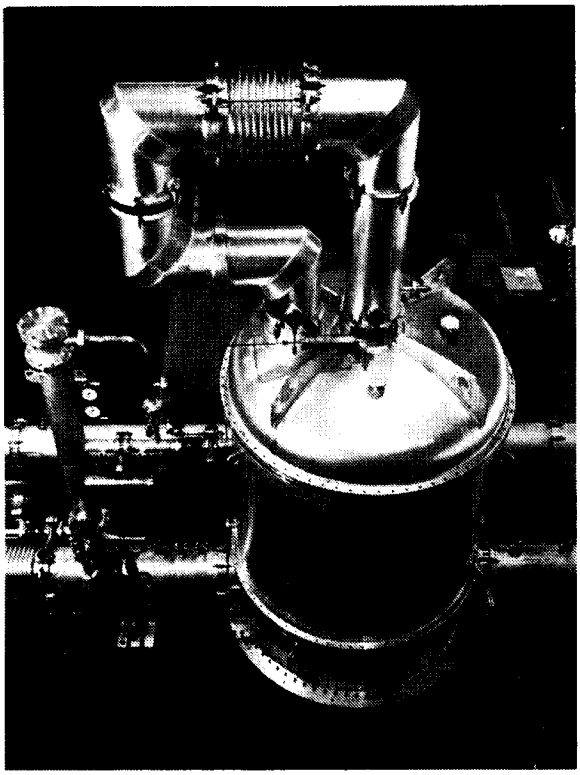

FIG. 7. Upper part of the separation nozzle stage, including the tank for the separation elements, the separation element test loop and the two short-circuit loops for the feed gas and the heavy fraction, respectively.

4. FURTHER DEVELOPMENT

In March 1970, the Gesellschaft für Kernforschung, Karlsruhe, and STEAG, Essen, signed a contract to co-operate in the separation nozzle process. It is the purpose of this co-operation to provide for the further development and practical application of the process. To evaluate the economics of the separation nozzle method, it is planned to submit the design documents for a separation nozzle demonstration plant with a separation capacity of 500 000 - 1 000 000 kg of separative work per annum in 1974 at the latest.

The block diagram used for the conceptual design study is shown in Fig.9. The base sections of the enrichment and stripping parts of the plant are made up of a total of 270 separation stages of the type described above (Figs 6 - 8). The head sections contain a total of 182 stages with a

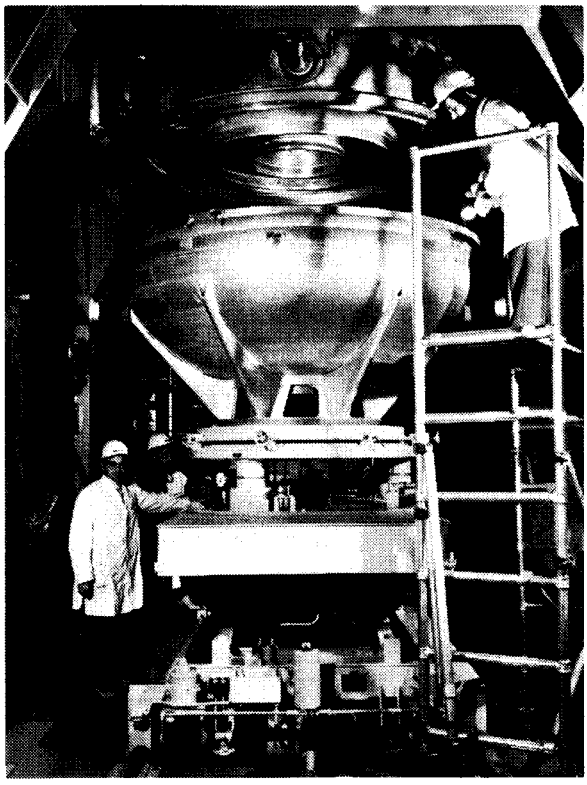

FIG. 8. Lower part of the stage during assembly of the compressor; the two-stage cooler can be recognized in the upper part of the picture.

throughput three times as small. Special separation nozzle stages, combined with a low temperature precipitation, will be used for the separation of UF_6 from the light added gas at the head and the shoulder of the enrichment section.

At 100% operating time and a waste concentration of 0.26%, the plant should produce 160 000 kg of uranium per year with 3% ^{235}U content. This corresponds to an output of about 610 000 kg of separative work per annum, which would be sufficient to fuel some 6000 MW(e) of light-water reactors continuously. In these figures it has been assumed that the performance data derived from laboratory scale tests as outlined in Table I can be employed.

With the present state of the art, the plant will have an electricity requirement of about 400 MW and cost about US $100 000 000. Its production capacity, and hence its power consumption, could be regulated

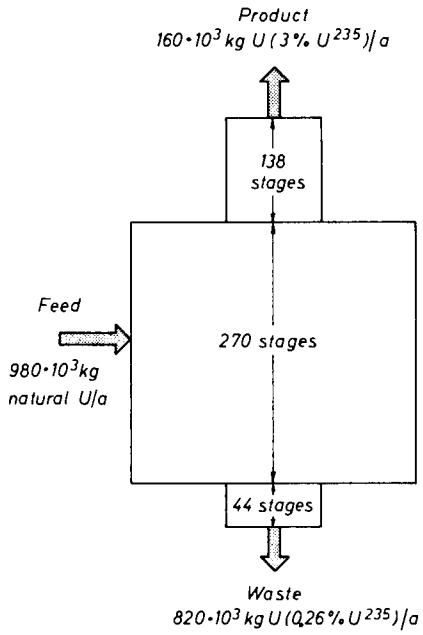

FIG. 9. Block diagram used for the conceptual design study of a separation nozzle demonstration plant.

TABLE I. OPERATING CONDITIONS AND PERFORMANCE DATA ASSUMED FOR THE CONCEPTUAL DESIGN OF THE DEMONSTRATION PLANT

UF_6/H_2 mixture with 5 mol.-% UF_6	
Initial nozzle pressure P_0	600 mm Hg
Suction pressure of compressor	150 mm Hg
Suction and final temperatures of compressor/coolers	40°C
UF_6 cut, ϑ_{UF_6}	1/3
Elementary separation effect of the uranium isotopes ϵ_A	0.015

within a range of roughly + 20 and - 30% through changes of pressure and temperature without major economic disadvantages. This and the extremely short adjustment time make the plant particularly suitable as an electricity buffer in a large interconnected electrical grid.

On the basis of the financing terms proposed by the Euratom Commission for uranium enrichment plants, the separative work costs would be 30 - 40% above the present United States standard. When judging these results, one should bear in mind that the demonstration plant considered

is smaller by a factor of ten as compared with an American diffusion plant. In addition, current basic investigations indicate the possibility of major improvements of the separation effect. Hence, the separation nozzle method should be regarded as a true alternative to the other processes of uranium isotope separation.

REFERENCES

[1] BECKER, E.W., BIER, K., BIER, W., SCHÜTTE, R., SEIDEL, D., Separation of the isotopes of uranium by the separation nozzle process, Angew. Chem. int. Edn. (in English) 6 (1967) 507 (this publication includes a summary of the previous work in this field).

[2] BECKER, E.W., FREY, G., SCHÜTTE, R., SEIDEL, D., Entmischung der Uranisotope in einer zehnstufigen Trenndüsen-Versuchsanlage, Atomwirtschaft 13 (1968) 359; see also Operation of a 10-stage separation nozzle pilot-plant, Symp. on Problems Bearing on the Isotope Separation of Uranium, Turin, October 1/2, 1968; KFK-Bericht 854.

[3] BECKER, E.W., BIER, W., SCHÜTTE, R., "Principles and economic aspects of the separation nozzle process", Symp. on Problems Bearing on the Isotope Separation of Uranium, Turin, October 1/2, 1968; KFK-Bericht 853.

[4] BECKER, E.W., BIER, W., FREY, G., SCHÜTTE, R., Trenndüsen-Demonstrationsanlage für die Urananreicherung, Atomwirtschaft/Atomtechnik 14 (1969) 249.

[5] BECKER, E.W., The separation nozzle process for uranium enrichment, Atoomenergie en haar toepassingen 11 (1969) 272.

ACQUIS FRANÇAIS EN MATIERE DE SEPARATION ISOTOPIQUE

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Abstract–Résumé–Аннотация–Resumen

FRENCH EXPERIENCE WITH ISOTOPE SEPARATION TECHNIQUES.

French experience in the field of isotope separation covers a wide range. The first laboratory investigations date back to 1954, while the first experience with gaseous diffusion was acquired on prototype plants in 1961. True industrial experience was first gained with the 'low-enrichment' plant in 1964 and with the whole complex working at design output in spring 1967. Since that time the excellent performance of the Pierrelatte plant has contributed still further to French know-how in this field. Moreover, intensive research on the equipment necessary for a gaseous diffusion plant of multinational scale is beginning to give a solid base to what was merely hypothesis just a few years ago. Other means of enrichment have also been investigated, in particular ultra-centrifugation: the mechanical problems of this technique have been resolved and those associated with the operation of centrifuges in a large plant have been tackled.

During a period of more than 15 years, France has accumulated a considerable amount of experience widely applicable to the various enrichment processes utilizing UF_6 (the handling of fluorine and its aggressive derivatives, and problems relating to surface treatment, passivation, corrosion, sealing; special devices for input and delivery, extraction and drawing-off, storage and purification; chemical and isotopic methods of analysis) or involviug duplicated units (problems of design, assembly and interchangeability; collection and real time processing of data necessary for the control and optimum utilization of the cascades together with the sophisticated mathematical methods required). To this may be added experience specific to gaseous diffusion, covering barriers, the choice and maintenance of their performance and their mass production at minimum cost; compressors of diverse types (piston, centrifugal or axial, operating at widely differing pressure levels); long-lived compressor sealing gaskets with a very low rate of leakage; electrical power supply, provision for partial or total load-shedding, and restarting times.

All this places France in a leading position, enabling her to propose precise construction estimates and enrichment cost schedules for a gaseous diffusion plant producing from 6 to 10 million SWU/yr of slightly-enriched uranium.

ACQUIS FRANÇAIS EN MATIERE DE SEPARATION ISOTOPIQUE.

L'acquis français en matière de séparation isotopique couvre de multiples aspects. Les premières études de laboratoire remontent à 1954, alors qu'une expérience en diffusion gazeuse débutait sur les matériels prototypes des usines en 1961. L'expérience vraie en usine basse a été obtenue dès 1964, et au printemps 1967 pour l'ensemble des usines à leur régime nominal de production. Depuis, l'excellent fonctionnement de Pierrelatte a fortement contribué à l'accroissement des connaissances. En outre, les études poursuivies activement sur les matériels nécessaires dans une usine de diffusion gazeuse de taille multi-nationale commencent à asseoir sérieusement ce qui n'était qu'hypothèse il y a seulement quelques années. D'autres voies d'enrichissement sont également examinées, en particulier l'ultra-centrifugation, où les problèmes mécaniques ont été résolus et ceux associés à la mise en œuvre de centrifugeuses dans une usine de grosse capacité abordés.

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** Centre d'études nucléaires de Saclay.
† Centre de Pierrelatte.

En fait, la France a accumulé depuis plus de 15 ans une expérience considérable, applicable dans une large mesure aux divers procédés d'enrichissement utilisant l'UF_6 (manipulation du fluor et de ses dérivés aggressifs, et problèmes de traitement de surface, de passivation, de corrosion, d'étanchéité; dispositifs particuliers d'introduction et d'alimentation, d'extraction et de soutirage, de stockage, de purification; méthodes d'analyse chimique et isotopique) ou mettant en œuvre des installations répétitives (problèmes de construction, de montage, d'interchangeabilité; collecte et traitement en temps réel des informations nécessaires à la régulation et à l'exploitation optimale des cascades avec la méthodologie mathématique sophistiquée que cela requiert). A ceci s'ajoute l'acquis spécifique à la diffusion gazeuse. Il porte sur: les barrières, le choix et le maintien de leurs performances, leur fabrication en série au moindre coût; des compresseurs très divers (à piston, centrifuge, axial, fonctionnant à des niveaux de pression très différents); l'endurance très importante du joint d'étanchéité des compresseurs malgré une fuite très faible; l'alimentation en énergie électrique, les possibilités de délestage partiel ou total et les temps de redémarrage.

Tout ce savoir donne à la France une position prépondérante et lui permet d'avancer un devis de construction et un coût d'enrichissement précis pour une usine de diffusion gazeuse produisant de 6 à 10 millions d'UTS par an d'uranium faiblement enrichi.

ОПЫТ ФРАНЦИИ В ОБЛАСТИ РАЗДЕЛЕНИЯ ИЗОТОПОВ.

Опыт Франции в области разделения изотопов имеет многочисленные аспекты. Первые лабораторные исследования относятся к 1954 году, тогда как метод газовой диффузии был впервые применен на прототипах заводских установок в 1961 году. В 1964 году был получен практический опыт эксплуатации завода, работавшего на неполной мощности, а весной 1967 года – группы заводов, работавших на проектной мощности. Превосходная работа завода в Пьерлате в последующее время в значительной степени способствовала накоплению знаний в данной области. Кроме того, активно проводившиеся разработки оборудования, необходимого для многонационального газодиффузионного завода, начинают подводить солидную основу под то, что еще несколько лет тому назад было лишь простой гипотезой. Изучаются и другие способы обогащения урана, в частности, процесс ультрацентрифугирования, для которого были решены технические проблемы и решаются проблемы, связанные с пуском в действие центрифуг на заводе большой мощности.

Более чем за 15 лет Франция накопила значительный опыт, вполне применимый к различным процессам обогащения с использованием UF_6 (обращение с фтором и его агрессивными производными, а также проблемы обработки поверхности, пассивации, коррозии и герметичности; отдельные устройства для ввода и подачи, экстракции и разделения, хранения и очистки; методы химического и изотопного анализа или, с вводом в действие репетитивных установок, проблемы конструирования, монтажа, взаимозаменяемости; сбор и обработка в нужное время информации, необходимой для регулирования и оптимальной эксплуатации каскадов с помощью требуемой для этого сложной математической методологии). Следует добавить к этому и знания, приобретенные в области газовой диффузии. Они касаются: различных перегородок, выбора и сохранения их характеристик, их серийного изготовления с наименьшими затратами; разнообразных компрессоров (поршневых, центробежных, осевых, работающих при самых различных уровнях давления); длительных сроков службы герметических соединений компрессоров с исключительно малой утечкой; энергоснабжения, возможностей частичной или полной выгрузки баласта и времени повторного пуска.

Благодаря этому опыту Франция занимает передовые позиции и может составить смету строительства газодиффузионного завода производительностью от 6 до 10 млн. единиц сепаративной работы в год для производства малообогащенного урана и определить точную стоимость обогащения.

REALIZACIONES FRANCESAS EN MATERIA DE SEPARACION ISOTOPICA.

Las realizaciones francesas en el campo de la separación isotópica abarcan múltiples aspectos. Los primeros estudios de laboratorio datan de 1954, y en 1961 se inició un experimento de difusión gaseosa con los materiales prototipo de las plantas. La verdadera experiencia comenzó a adquirirse en 1964, en planta de bajo enriquecimiento, y en la primavera de 1967, en lo que respecta al conjunto de las plantas a su régimen nominal de producción. Desde entonces, el excelente funcionamiento de Pierrelatte ha contribuido en gran manera a la expansión de nuestros conocimientos. Además, los estudios desarrollados activamente sobre los materiales necesarios para una planta de difusión gaseosa de dimensiones multinacionales comienzan a dar una base sólida a lo que hasta hace solamente algunos años no era más que una hipótesis. Se estudian también otras vías de enriquecimiento, especialmente la ultracentrifugación, cuyos problemas mecánicos han sido resueltos, y abordados los inherentes a la puesta en servicio de centrífugas en una planta de gran capacidad.

Así, pues, Francia ha acumulado en más de 15 años una experiencia considerable, aplicable en gran medida a los diversos procedimientos de enriquecimiento en que se utiliza el UF_6 (manipulación del flúor y

de sus derivados químicamente activos, y problemas de tratamiento de superficies, de pasivación, de corrosión y de estanqueidad; dispositivos especiales de introducción y alimentación, de extracción y trasiego, de almacenamiento y de purificación; métodos de análisis químico e isotópico) o en que intervienen instalaciones repetitivas (problemas de construcción, de montaje, de intercambiabilidad; acopio y tratamiento en tiempo real de las informaciones necesarias para la regulación y explotación óptima de las cascadas con la refinada metodología matemática requerida). A esto se añaden las realizaciones específicas de la difusión gaseosa, que se refieren a: las barreras, la elección y la conservación de sus características de funcionamiento, su fabricación en serie al costo mínimo; compresores muy diversos (de pistón, centrífugos, axiales, que funcionan a niveles de presión muy diferentes); la resistencia – muy importante – de la junta de estanqueidad de los compresores pese a una fuga muy pequeña; la alimentación de energía eléctrica, las posibilidades de vaciado parcial o total y el tiempo de nueva puesta en marcha.

Toda esta experiencia pone a Francia en una posición preponderante y le permite presentar un presupuesto de construcción y un coste preciso de enriquecimiento para una planta de difusión gaseosa capaz de producir de 6 a 10 millones de UTS/año de uranio ligeramente enriquecido.

L'USINE DE PIERRELATTE; MATERIELS NOUVEAUX ET PROTOTYPES

La construction d'une usine d'enrichissement de l'uranium représente une somme d'études très considérable.

En France, les premières études de laboratoire remontent à 1954, et dès 1958, dans le cadre de cette même conférence de Genève, nous avons fait état des résultats notables que nous avions obtenus en un court laps de temps. La construction de l'usine n'était encore qu'à l'état d'avant-projet, mais la définition des matériels progressait rapidement, puisque l'année 1961 voyait débuter les essais sur prototypes des composants fondamentaux de l'usine, d'abord les compresseurs, puis les barrières, tant dans la région parisienne que dans la zone d'expérimentation de Pierrelatte.

Ensuite, après une expérimentation menée rondement, les choix ont été précisés rapidement et la construction, sous la houlette de la Société USSI, l'architecte industriel, progressait rapidement puisque l'usine basse, la première des quatre, démarrait à l'UF_6 au cours du deuxième trimestre de 1964 et que la totalité de l'usine était construite et atteignait son régime nominal de production à une concentration de plus de 90% au début du printemps 1967. Depuis cette date, l'usine, dont le fonctionnement est resté irréprochable – taux de disponibilité supérieur à 98% –, a vu sa puissance de séparation accrue très sensiblement grâce à l'augmentation des niveaux de pressions et à la mise en place de barrières améliorées dans l'usine basse.

Ainsi, depuis plus de sept ans pour les étages les plus anciens et plus de quatre ans pour la totalité de l'usine, Pierrelatte nous apporte des éléments indiscutables sur l'exploitation et confirme, avec une sûreté toujours accrue, nos connaissances sur la durée de vie des divers composants.

En plus des connaissances acquises en exploitation industrielle, nous avons entrepris dès 1966, à partir des travaux effectués pour Pierrelatte, des études d'installations industrielles de grande capacité à faible enrichissement; pour cela ont été maintenus en place dans les Sociétés d'études, en particulier chez USSI (l'architecte industriel) et chez les industriels intéressés, les noyaux des équipes auxquelles Pierrelatte doit son excellent fonctionnement.

En se basant sur les connaissances acquises lors de l'exploitation, les nouvelles études poursuivies par ceux-là mêmes qui avaient présidé à la conception puis à la construction ont eu pour premier objectif une critique positive de Pierrelatte visant à dégager l'essentiel afin d'arriver à la meilleure économie globale. Ainsi, cette même usine pourrait être construite aujourd'hui à un coût nettement inférieur sans sacrifier aucunement ses performances. L'analyse de valeur sur les divers composants et dispositifs a été poussée à l'extrême, afin que chacun soit sensibilisé aux gains réalisables grâce à la connaissance précise du comportement de l'usine, des conditions d'exploitation et de la fiabilité des matériels et des circuits.

En parallèle, et avec le souci de profiter des résultats précédents, a été lancée la réalisation d'appareillages prototypes qui pourraient être utilisés dans la construction d'une puissante usine d'enrichissement. Dans une première période, on s'est intéressé aux matériels nouveaux nécessaires pour une usine répondant uniquement aux besoins nationaux, de l'ordre d'un million d'UTS par an. La plupart des matériels ont été effectivement construits et montés à Pierrelatte sur des circuits d'essais en UF_6 ; ils donnent toute satisfaction, répondent aux spécifications imposées et sont utilisables dans les étages de tête d'une installation de grande capacité.

Toutefois, de tels étages, d'une puissance d'environ 800 kW, sont encore relativement petits, et, de ce fait, ne peuvent prétendre, utilisés seuls, à la compétitivité économique.

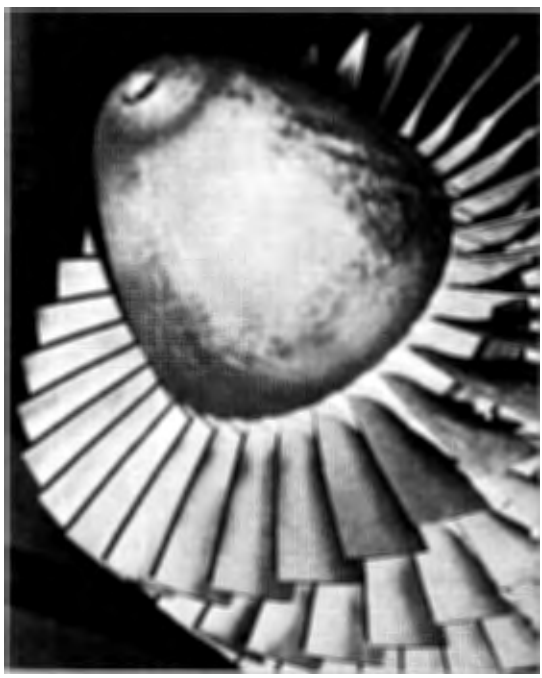


FIG. 1. Premiers aubages du compresseur axial de 800 kW.

Une deuxième étape a donc été entreprise qui a conduit à la réalisation des appareils correspondant aux plus gros étages d'une usine de 6 à 10 millions d'UTS par an. La plupart de ces appareils, en particulier les compresseurs à haute performance conçus par le GERCOS (Groupement d'étude et de réalisation de compresseurs spéciaux), ont été réalisés et font actuellement l'objet d'essais en UF_6 (fig. 1).

L'ensemble de ces travaux doit permettre, d'une part, de vérifier que les techniques de construction mises au point pour l'usine actuelle de Pierrelatte sont transposables à une usine de séparation isotopique à vocation européenne, d'autre part, de donner aux études économiques, grâce aux résultats d'appels d'offres véridiques, des bases saines et réalistes permettant de s'assurer de l'exactitude de prix de revient avancés.

Parallèlement, des études sont poursuivies sur l'ultracentrifugation, tant au niveau des composants, les centrifugeuses en particulier, qu'à celui de l'usine les mettant en œuvre. Certaines techniques propres aux grands ensembles de matériels répétitifs et qui ont été mises au point pour la diffusion – détermination de la fiabilité des composants, optimisation de la maintenance, qu'elle soit préventive, circonstancielle ou mixte, topologie des circuits, groupement en unités d'entretien pour une fiabilité maximale de l'ensemble – sont aisément transposables à ce procédé. De plus, les techniques particulières propres aux unités d'introduction, d'extraction, de soutirage et d'alimentation sont directement utilisables. Enfin, dans le domaine de la conduite d'installations répétitives, les dispositifs mis en œuvre à Pierrelatte sont certainement en grande partie reconductibles à tous les procédés de séparation.

EXPERIENCE D'ENSEMBLE CONCERNANT L'UF_6

Nous avons accumulé une expérience importante depuis 15 années afin d'être parfaitement maîtres des conditions de fonctionnement de l'usine. Les larges domaines de conditions opératoires que nous avons balayés dans nos études nous permettent de répondre pratiquement à tous les problèmes rencontrés dans les procédés d'enrichissement mettant en œuvre l'UF_6 :

– Nous avons acquis une connaissance étendue de la technologie de base, tant au laboratoire qu'à l'échelle industrielle, de la manipulation des composés fluorés dans un large domaine de températures et de pressions, et des contraintes imposées par ces composés sur la conception des circuits et des appareillages (matériaux, méthodes complexes de traitement de surface et de passivation, joints nécessaires à l'obtention de normes d'étanchéité draconiennes: 10^{-2} lusec/m^3).

– La gamme très étendue des dimensions des appareillages réalisés à ce jour assure aux industriels français une maîtrise complète des problèmes de construction et de montage: si nous prenons pour exemple les vannes de circuits UF_6, les plus petites ont un diamètre de passage de 4 mm (analyse) et les plus grandes réalisées, 700 mm (fig. 2). Des vannes de 1200 mm sont en cours de fabrication pour les collecteurs des gros étages.

– En outre, la présence d'un nombre important d'unités identiques, installées dans les mêmes conditions, nous a permis de résoudre, tant

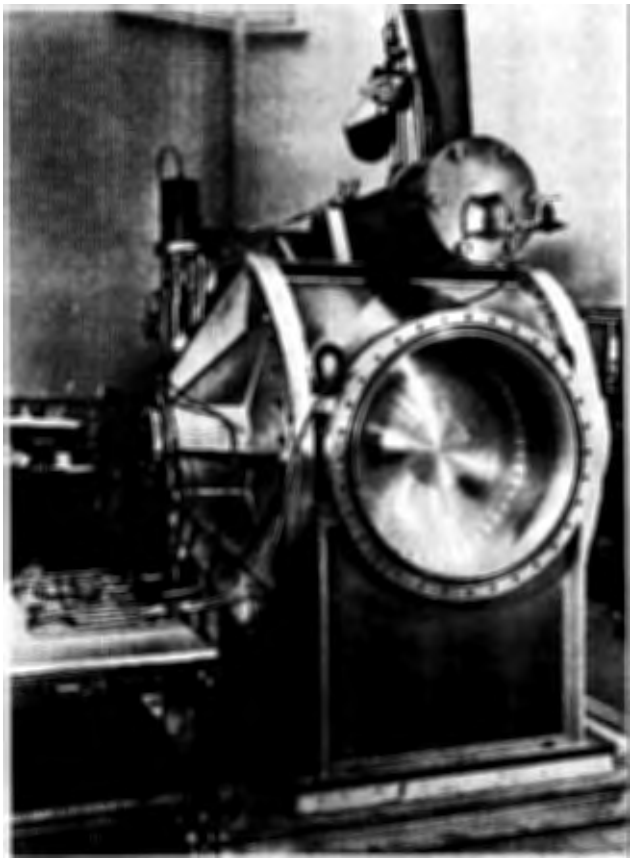


FIG. 2. Vanne Matra (diamètre 700 mm) pour UF_6.

pour le montage que pour la maintenance, le problème de l'interchangeabilité soit des unités soit des appareils constitutifs, et les solutions trouvées se sont montrées tout à fait sûres. Les dispositions prises permettraient, par exemple à Pierrelatte, de permuter n'importe quel ensemble de deux éléments de circuit homologues au sein d'une usine déterminée. Pour un supplément modique de coût de fabrication, que le faible entretien nécessaire ne justifierait pas, on réalise une économie très substantielle sur les frais de premier montage.

LES ANNEXES

Le fonctionnement d'une installation industrielle mettant en jeu un produit aussi particulier que l'UF_6 suppose l'existence d'un certain nombre d'unités spéciales assurant avec une grande fiabilité ce que l'on pourrait appeler les relations du procédé d'enrichissement avec l'extérieur.

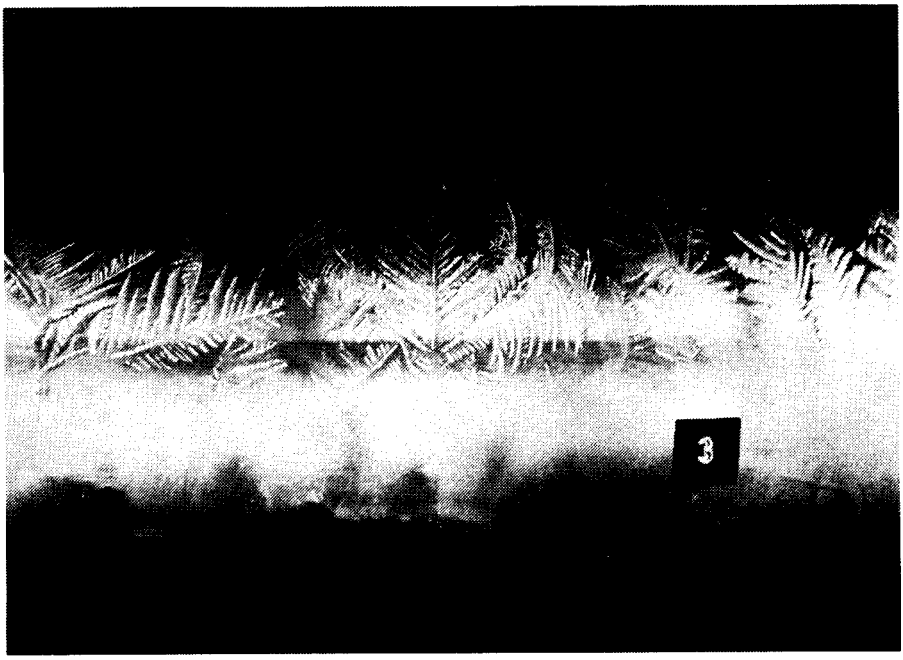


FIG. 3. Visualisation de la cristallisation de l'UF_6.

Il faut en effet:
- introduire le fluide de procédé dans les unités avant leur mise en service,
- extraire ce fluide en cas de besoin (fig. 3),
- assurer la purification du fluide de procédé,
- assurer les fonctions d'alimentation permanente en UF_6 naturel et de soutirage continu d'UF_6 appauvri et enrichi,
- stocker ou traiter l'UF_6 appauvri non repris par les clients de l'usine.

Les annexes de l'usine, bien que constituant une charge importante d'études du fait de leur diversité et unicité, ne représentent qu'un faible pourcentage du devis total; mais leur conception judicieuse et leur bonne exploitation sont une condition essentielle du fonctionnement correct de l'ensemble. La connaissance des problèmes correspondants est un atout important dans la réussite d'un projet.

PURGE DES LEGERS

Une usine, si étanche soit-elle, ne peut prétendre à l'étanchéité absolue. Par ailleurs, les garnitures de compresseurs, même très sophistiquées, qui sont en service à Pierrelatte, laissent passer un faible débit de gaz léger. Enfin, l'UF_6 naturel d'alimentation peut contenir un faible pourcentage de HF.

Tous ces gaz vont s'élever dans la cascade, leur concentration molaire va croître dans les usines supérieures, et il est nécessaire de les éliminer avant que leur concentration ne devienne gênante.

De nombreux procédés sont envisageables et plusieurs d'entre eux ont été exploités industriellement à Pierrelatte. La bonne conservation de l'étanchéité à la construction permet des réductions de frais de fonctionnement, grâce à la suppression des purges intermédiaires et par le choix, pour la tête d'usine, du procédé le plus fiable et le moins coûteux.

ANALYSES

Les analyses physico-chimiques sont un élément fondamental de la maîtrise de tout procédé de séparation isotopique. Deux techniques essentielles sont mises en œuvre: l'analyse par spectrométrie de masse et la chromatographie en phase gazeuse.

Les spectromètres de masse sont des analyseurs à sources fonctionnant en double collection, dont les résultats sont fournis par rapport à des étalons connus avec une grande exactitude et élaborés à Pierrelatte même. Ils donnent des concentrations en ^{235}U au millième et des enrichissements relatifs au dix-millième. Environ 200 000 analyses isotopiques ont été réalisées depuis le démarrage.

Les chromatographes, ainsi que certains types de spectromètres de masse spéciaux, analysent le gaz de procédé, donnant en permanence les teneurs en impuretés (air, azote, fluorures volatils, HF). Environ 900 000 h de scrutation ont été réalisées à ce jour.

Partant d'équipements initialement conçus pour le laboratoire, Pierrelatte a développé de véritables capteurs industriels satisfaisant aux critères suivants: précision, fiabilité, cadences de scrutation élevées (en ligne), points de scrutation très nombreux, coûts d'exploitation minimaux.

TRAITEMENT DES INFORMATIONS

Le manque de connaissance précise du fonctionnement de l'usine nous avait conduits à une conception de conduite largement manuelle, mais faisant appel à des automatismes simples. Toutefois, dès le démarrage, les données de fonctionnement ont été recueillies afin d'être présentées sous une forme synthétique de plus en plus précise jusqu'à préfigurer un dispositif complet d'acquisition et de traitement automatique des informations «procédé».

L'équipement des usines était effectué entre 1968 et 1970 à partir d'une connaissance excellente du comportement de l'usine qui, seule, a permis de dégager les paramètres fondamentaux.

Dans chacune des quatre usines, un ordinateur scrute en permanence les informations utiles analogiques ou tout-ou-rien: au total, environ 12 000 données. Les restitutions ont lieu sur des ensembles de visualisation électroniques au niveau de la salle de conduite d'usine et, grâce à un ordinateur central puissant, en salle de conduite centrale (fig. 4).

Le système assure une surveillance constante des matériels en fonctionnement et des paramètres de contrôle de procédé; il informe

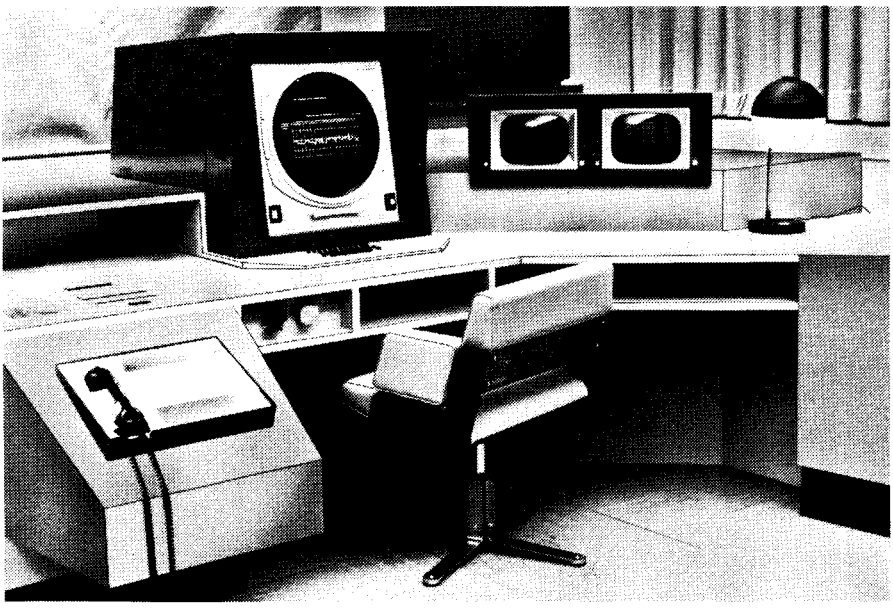


FIG. 4. Conduite centralisée: console de visualisation et de contrôle.

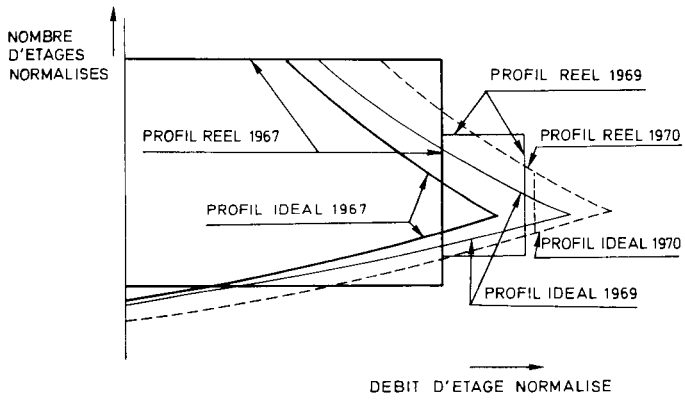


FIG. 5. Profils successifs de l'usine basse et comparaison avec le profil idéal.

automatiquement les opérateurs de toute anomalie. Cette surveillance centralisée, et à distance, est venue se superposer aux automatismes réflexes câblés et aux régulations simples déjà en place.

Le «software», entièrement mis au point par nos spécialistes, associe en temps réel des traitements simples (comparaison de valeurs à des fourchettes ou des seuils) et des traitements complexes. Il délivre aux opérateurs une information élaborée leur permettant de prendre des décisions rapides et sûres.

La disponibilité du système est de l'ordre de 98%.

La mise en service de ce nouvel outil de conduite apporte:

– Une connaissance parfaite et permanente du procédé. Toute perturbation aérodynamique susceptible d'entraîner un remélange isotopique, donc une perte de travail de séparation, est immédiatement détectée et contrée. Les températures et pressions peuvent être tenues respectivement à mieux que ±1 °C et ±15 mb; les fluctuations de la masse d'uranium contenue dans les différents tronçons de la cascade sont inférieures à 1‰.

– La possibilité d'ajuster le profil réel de l'usine très près du profil idéal. Le profil «carré» initial a été transformé en un profil approprié, ce qui permet de faire varier fictivement la «taille» de chaque groupe d'étage en fonction de son rang dans la cascade et ainsi de gagner en efficacité (fig. 5).

– Une réduction très importante du personnel direct de conduite.

Financièrement parlant, ce système sera totalement amorti au bout de quatre ans d'exploitation seulement.

MODELES MATHEMATIQUES

Grâce à l'exploitation systématique des résultats accumulés au fil des années, les modèles mathématiques théoriques ont pu être corrigés et raffinés et nous assurent désormais une connaissance très approfondie du procédé et de ses composants. Ils nécessitent de puissants moyens informatiques agissant, suivant les cas, en temps réel ou différé. Ils permettent une gestion très fine de la cascade, sans risque pour les matériels même au voisinage de leurs performances extrêmes.

Des modèles permettent, à partir d'un programme de production donné (soutirages et introductions simples ou multiples), d'optimiser la configuration et les paramètres de conduite de la cascade, de manière à obtenir le travail de séparation voulu au meilleur coût.

Enfin, une gestion d'ensemble a été mise au point, qui, tenant compte du programme des besoins, des espérances de vie de matériels, des niveaux de fonctionnement possibles, détermine les paramètres d'exploitation et la stratégie de maintenance qui minimise le coût du produit.

ACQUIS EN DIFFUSION GAZEUSE

Cet ensemble de connaissances, non spécifique à la diffusion gazeuse, est applicable à la plupart des procédés connus d'enrichissement de l'uranium. Mais il ne faut toutefois pas négliger l'acquis spécifique à ce procédé et qui porte en particulier sur:

La barrière

C'est l'un des éléments fondamentaux dans la conception d'une usine de diffusion gazeuse, et ses performances, son prix, sa durée de vie, pèsent lourdement sur l'optimisation. Toutefois, une expérience industrielle obtenue sur plusieurs types de barrières de natures très

différentes nous amène à les considérer comme des biens d'investissement. Par ailleurs, plusieurs années de pratique de séparation isotopique et de génie chimique expérimental nous ont amenés à conclure qu'il faut, pour arriver à des investissements raisonnables, travailler à des pressions amont nettement supérieures à l'atmosphère. Plusieurs boucles d'essais ont permis de vérifier qu'il est possible de le faire.

Ces conditions requièrent l'usage de barrières performantes, c'est-à-dire dont le rayon de pore est inférieur à 100 Å. Notre expérience nous a montré qu'il s'agissait là d'un problème complexe qui nécessite un effort important dans le domaine des études fondamentales.

L'analyse très poussée des pores de faible rayon nous a conduits à améliorer la précision de certaines mesures et à mettre au point des appareillages, tels que des porosimètres pouvant travailler avec une grande sensibilité, des appareils d'adsorption donnant automatiquement des isothermes avec une précision de l'ordre du pour-cent, etc.

La figure 6 représente un exemple de défauts observés avec le microscope électronique, défauts difficiles à détecter par les moyens classiques.

Les essais de séparation isotopique à l'UF_6 sont forcément limités, car ils mettent en œuvre des installations coûteuses: pilotes tels que PC 5, spectromètres de masse de haute précision, etc.

La séparation des isotopes de l'argon nous semble un modèle commode pour l'étude de la séparation isotopique de vapeurs; la comparaison de ces résultats avec ceux obtenus avec l'UF_6 montre une corrélation satisfaisante.

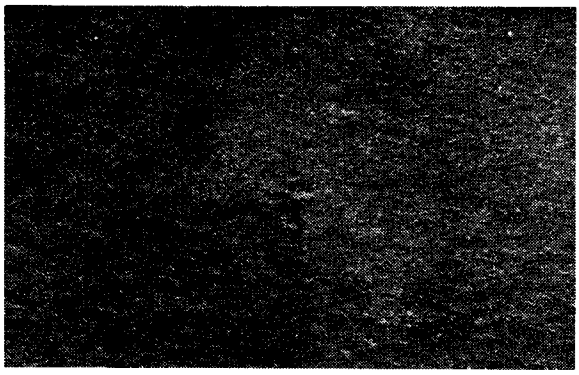
En outre, les problèmes d'industrialisation de millions de barrières, qui conditionnent fortement les prix de revient, n'ont pas été négligés. L'expérience de la fabrication des barrières de Pierrelatte a permis la conception d'une chaîne automatisée grâce à laquelle les nouvelles barrières de l'usine basse ont été fabriquées dès 1968. Depuis, des études industrielles ont été poursuivies sur divers types de barrières avec les industriels compétents, afin de cerner des prix de série réalistes.

Enfin, les différents modes d'assemblages de barrières entre elles à l'intérieur d'un diffuseur, en vue de la conservation dans le temps de bonnes performances aérodynamiques, alliés à un prix réduit sont étudiés sur pilotes industriels.

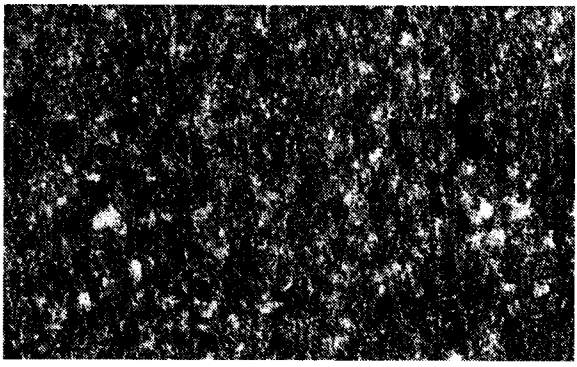
Le compresseur

Un autre élément important est le compresseur. L'ensemble des installations de Pierrelatte comporte une grande variété de compresseurs, installés tant dans les étages de diffusion eux-mêmes que dans les unités annexes.

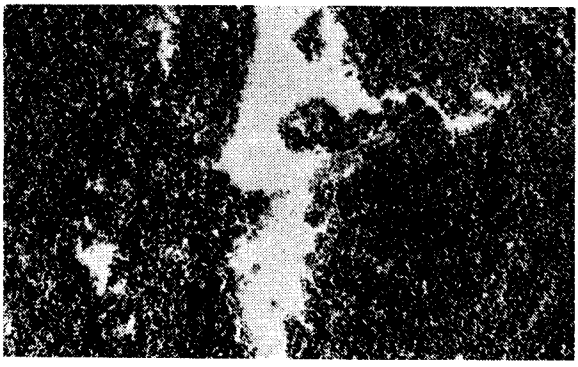
Nous mentionnerons bien entendu les machines utilisées dans les étages de diffusion: pour obtenir les performances très poussées nécessaires afin de maintenir la puissance consommée au niveau le plus bas, des études aérodynamiques ont été menées activement par le GERCOS, qui regroupe les industriels qui ont construit les compresseurs de Pierrelatte, et l'on peut dire que le rendement de compression obtenu est très proche du rendement maximal possible, malgré une conception économique de la machine (fig. 7). Le type centrifuge retenu pour cette première usine fait maintenant place aux techniques axiales. Diverses aérodynamiques,



A



B



C

FIG. 6. Examen en microscopie électronique de couches diffusantes: A. sans défauts; B. avec quelques défauts; C. présence de divers types de défauts.

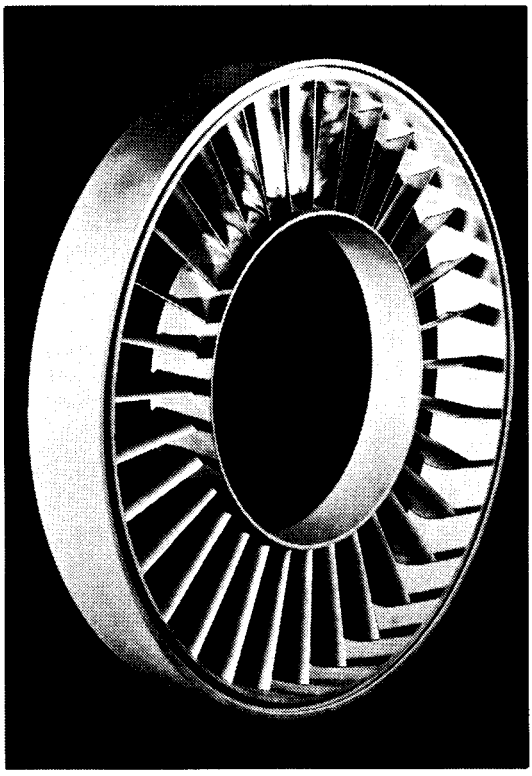


FIG. 7. Couronne d'aubages, redresseur de compresseur axial (acier inoxydable et alliage léger).

depuis le subsonique classique jusqu'au supersonique poussé, ont été évaluées tant en performance qu'en prix de revient, et le compresseur choisi résulte d'une sous-optimisation de ces paramètres. Les résultats obtenus sur les pilotes PP 201 et PP 202 confirment entièrement les prévisions de rendement, jugées un temps optimistes, des aérodynamiciens. Il était également nécessaire de prévoir, par exemple pour la purge du flux de procédé (élimination des incondensables) ou des soutirages, des compresseurs spéciaux pouvant fonctionner à des niveaux de pression variés, avec des gaz de compositions très diverses: des machines volumétriques ont été mises au point. Elles assurent un service sans défaillance.

Un des composants importants dans une machine tournante est la garniture d'étanchéité. Les conceptions qui ont conduit aux excellentes performances obtenues à Pierrelatte, tant du point de vue des fuites d'azote que de la contre-diffusion de l'UF_6, ont été reconduites sur les compresseurs axiaux. Cet élément, normalement soumis à usure, atteint des durées de vie dépassant 40 000 h. C'est presque un bien d'investissement.

Les matériels

Chacun des autres composants du circuit, bien que moins important que la barrière et le compresseur, est étudié dans de nombreuses variantes avec un soin tout particulier, qu'il s'agisse des vannes, des tuyauteries, des joints et autres méthodes de raccordement, des matériaux et de le leur revêtement, ou du génie civil, afin de toujours obtenir les performances et la fiabilité visées au meilleur coût.

SOURCE D'ENERGIE

Un élément déterminant du projet est l'alimentation en énergie.

Une usine de séparation en est une grosse consommatrice: environ 2000 MW pour 6 millions d'UTS par an. La quasi-totalité de la puissance électrique fournie à l'usine de diffusion est utilisée pour faire tourner les moteurs entraînant les compresseurs. En cas d'interruption, même très brève, de l'alimentation, ces derniers ralentissent et, au retour de la tension, leur réaccélération provoque un appel considérable d'intensité. Si le réseau de distribution ne peut pas y répondre, l'usine s'arrête. Il faut donc, compte tenu du moment d'inertie des machines tournantes et de la durée maximale prévisible des microcoupures, étudier la stabilité électrique du système.

L'expérience de Pierrelatte a permis de contrôler la validité des hypothèses faites lors de la construction; il a été ainsi possible d'extrapoler aux dimensions d'une usine de grosse capacité et de déterminer les caractéristiques fondamentales de son alimentation.

Par ailleurs, chacun sait l'intérêt qu'un producteur d'électricité porte aux possibilités d'élimination volontaire d'une partie de sa clientèle aux périodes de pointe de consommation. Ces clients peuvent donc bénéficier de tarifs préférentiels. Pour une usine de diffusion gazeuse, un tel avantage est très intéressant, puisque le coût de l'énergie entre pour près de la moitié dans le prix de revient du travail de séparation.

Le CEA et l'EDF ont entrepris des études communes visant à comparer le gain dû à une réduction des tarifs de fourniture d'énergie à la perte de production de l'usine consécutive aux temps d'arrêt et de remise en route de la cascade.

Des gains substantiels peuvent être envisagés par rapport aux tarifs industriels habituels, et l'expérience de Pierrelatte nous apporte sur l'exploitation des éléments concrets indiscutables.

CONCLUSION

Les éléments qui viennent d'être exposés ne représentent que quelques exemples particuliers de l'acquis de Pierrelatte, résultant des études fondamentales effectuées, de l'expérience gagnée lors de la construction de quatre usines différentes et de leurs annexes, et des résultats d'exploitation. Ils assurent à la France une connaissance d'ensemble approfondie de la technologie applicable aux nombreux procédés d'enrichissement basés sur l'UF_6 et tout particulièrement à la diffusion gazeuse.

Dans cette technique, nous poursuivons activement un programme détaillé d'études et d'expérimentation axé vers l'obtention du dossier complet d'une usine de grosse capacité, dans la gamme 6 à 10 millions d'UTS par an. Les prototypes des appareillages constitutifs des étages sont réalisés en plusieurs variantes et les performances acquises sur la plus petite taille, pleinement conformes aux spécifications imposées, ont été obtenues dans les conceptions économiques envisagées pour l'usine finale. Le test des prototypes des appareillages de la grosse taille est en cours et nous avons bon espoir d'atteindre ici encore les objectifs tant techniques qu'économiques visés. Les barrières méritent une mention particulière, car elles sont des composants sur lesquels des améliorations se font constamment, même lors de la période de construction. Les performances imposées ont d'ores et déjà été obtenues sur installation pilote et les études d'industrialisation en cours permettront leur reproduction en série au prix le plus bas, probablement même dans une version améliorée.

Tout ceci concourt donc à l'obtention d'ici à 1973 d'un ensemble complet et cohérent de données confirmées qui vont constituer le dossier de réalisation d'une usine d'enrichissement multinationale. Les coûts d'investissement seront connus très en détail, les performances des matériels auront été vérifiées et les frais d'exploitation explicités en fonction de l'endurance prévisible des matériels.

Le choix de la taille d'une usine et la détermination des conditions qui seront consenties pour le loyer de l'argent et le prix de l'énergie permettront alors de calculer avec précision le coût du travail de séparation. Toutefois, les objectifs techniques que nous nous sommes fixés doivent nous permettre d'atteindre la compétitivité pour une capacité comprise entre 6 et 10 millions d'UTS par an, tout en assurant une marge suffisante pour la rémunération du capital dans des conditions industrielles normales.

REMERCIEMENTS

Cette communication est le résultat du travail mené en commun par des équipes du CEA et de l'industrie privée. Il faut mentionner les contributions importantes apportées par MM. H. Bouzigues, J. Charpin, P. Delarousse, J. Ergalant, C. Fréjacques, M. Gelée, C. Lebrun, Y. Martin, M. Mézin, J. Ohmann, M. Perrault, J. P. Petit et leurs collaborateurs.

d'un programme établi en accord avec les services d'enrichissement de l'uranium depuis ses débuts, en janvier 1969. Il examine les modifications ou les considérations de politique générale liées à l'augmentation de la capacité de séparation des isotopes en tenant compte de leur incidence sur les futures centrales d'enrichissement à façon. Il donne également un aperçu de la politique suivie dans l'établissement d'un système de contrôle et d'une coopération, à l'échelon international, dans le domaine de l'enrichissement de l'uranium.

ИЗОТОПНОЕ ОБОГАЩЕНИЕ УРАНА.

Быстрый рост мощности атомных электростанций с реакторами, работающими на слабообогащенном урановом топливе, значительно повысил интерес к процессам и установкам для разделения изотопов урана. Хотя мощность существующих заводов по обогащению урана используется не полностью, оценка будущих потребностей показывает, что их мощность вскоре придется увеличивать. В докладе представлены результаты исследования, проведенного КАЭ США по долгосрочному планированию. Эти исследования включают оценку мощности существующих заводов и анализ необходимых изменений эксплуатационных характеристик с учетом будущих потребностей в обогащенном ядерном топливе. Они показывают влияние изменения различных факторов в оценках будущих потребностей в ядерном топливе и в оптимальных эксплуатационных характеристиках обогатительных заводов. Рассматриваются такие факторы, как повторное использование плутония в реакторах на тепловых нейтронах, изменение отношения к хвостовым продуктам обогатительных заводов, изменения стоимости электроэнергии или стоимости урановой руды, а также влияние совершенствования реакторной техники, включая реакторы-размножители. В докладе последовательно и подробно обсуждается программа и политика США, направленная на удовлетворение предполагаемых потребностей в обогащении урана со стороны потребителей внутри страны и за рубежом. Удовлетворение потребностей все увеличивающейся мощности атомных электростанций может быть достигнуто за счет следующих изменений: увеличение в снабжении электроэнергией газодиффузионных заводов, изменение отношения к хвостовым продуктам, установка оборудования, повышающего эффективность использования электроэнергии (в пределах существующего потребления энергии или с превышением этих лимитов) и, наконец, строительство новых заводов. Дается экономическая оценка всех этих изменений. Обсуждается опыт работы в системе обслуживания по обогащению урана по соглашениям, введенной в январе 1969 года. Возможные изменения политики или подхода в связи с предстоящими новыми крупными капиталовложениями в промышленность по изотопному обогащению урана обсуждаются во взаимосвязи с новыми налоговыми обязательствами. В докладе также обсуждается политика в отношении международного контроля и кооперации в области изотопного обогащения урана.

ENRIQUECIMIENTO DE ISOTOPOS DE URANIO.

El rápido incremento de la generación de electricidad nuclear basada en reactores que utilizan combustible de uranio ligeramente enriquecido atrae naturalmente creciente atención sobre los procedimientos y plantas para separación de isótopos de uranio. Aunque las plantas existentes de enriquecimiento de uranio están funcionando a mucho menos de su capacidad total, las previsiones de necesidades futuras indican que urge tomar importantes decisiones referentes a la expansión de tal capacidad. En esta memoria se describen recientes estudios de planificación a largo plazo realizados por la USAEC. Esos estudios se inician con la capacidad de las plantas actuales y analizan las futuras posibilidades de operación en relación con el cálculo de las futuras necesidades de combustibles nucleares enriquecidos. Esos estudios muestran los efectos de los cambios de varios factores sobre las estimaciones de las necesidades futuras de combustibles nucleares y sobre el modo óptimo de operación de las plantas de enriquecimiento. Entre los factores considerados se incluyen el reciclado del plutonio en los reactores térmicos, los cambios en los análisis químicos de los estériles de las plantas de enriquecimiento, los cambios en los costos de la producción energética o de los minerales de uranio, y los efectos de la tecnología de reactores avanzados, incluyendo los reactores reproductores. Los programas de los Estados Unidos y las directrices destinadas para satisfacer las necesidades previstas de enriquecimiento de uranio de los consumidores nacionales y extranjeros se tratan de manera gradual y extensa. Las posibilidades existentes para proporcionar combustibles nucleares para satisfacer la creciente demanda de electricidad de origen nuclear comprenden un aumento de la capacidad generadora de las plantas de difusión, los cambios en los análisis de estériles, la instalación de equipo para mejorar la eficiencia del empleo de la fuerza producida dentro o más allá de los límites nominales actuales, y finalmente el establecimiento de nuevas plantas. Se resumen los cálculos de las consecuencias económicas de tales posibilidades. Se revisa la experiencia adquirida con el programa de acuerdos sobre servicios de enriquecimiento de uranio, desde sus comienzos en enero de 1969. Los cambios en las directrices o las consideraciones resultantes de las necesidades previstas en relación con las inversiones en gran escala para el aumento de la capacidad de enriquecimiento de isótopos de uranio se consideran en sus incidencias sobre las obligaciones

derivadas de los nuevos acuerdos sobre tasas de enriquecimiento. Se resumen también en la presente memoria las directrices referentes a los controles y la cooperación internacionales en el campo del enriquecimiento de los isótopos del uranio.

INTRODUCTION

Nuclear power is now entering a 'rapid growth' phase as a major source of new electric power generating capacity in many parts of the world. Power reactors utilizing partially enriched uranium fuel are being built in ever increasing number. This directs attention toward the complex problem of how best to satisfy the world's expanding need for uranium isotope enrichment.

The United States of America has had more than twenty-five years of highly successful experience in enriching uranium using the gaseous diffusion process. We now have three major plants representing a total investment of over US $2300 million with an annual capacity of more than 17 million units of separative work when operated at their nominal full power of about 6100 megawatts of electricity. However, during the current year, these plants are being operated at only about 50% of their full capacity.

There is interest apparent in many other countries in the establishment of alternate sources of uranium enrichment services. In addition, effort is being directed in numerous countries toward the development of other uranium enrichment processes.

The extreme importance of having an adequate supply of enriching capacity available on a timely basis at reasonable cost, coupled with the tremendous cost of process development and enrichment plant construction, justifies the closest possible co-operation and communications among the users and suppliers of enrichment services.

We will attempt in this paper to present our views on these matters.

PROJECTED NUCLEAR POWER GROWTH

Enough uranium fuel will have to be enriched to about 3% ^{235}U over the next several decades to meet the demands of hundreds of reactors (currently light-water reactors) which are now or will soon be under construction. US nuclear generating capacity, for example, is projected to grow from 5000 MW(e) in 1970, to 150 000 MW(e)[1] in 1980, and to 300 000 MW(e) by 1985. It is estimated that in 1980, 29% of the electricity generated by utilities in the United States will be from nuclear power, and in the decade of the 1990's, that fraction may be about 50%.

Foreign nuclear capacity requiring enriching services will be in the range of 70 000 - 100 000 MW(e)[2] by 1980. Taking into account the fact that some enriching services will be furnished by foreign sources, the USAEC projects the foreign and domestic nuclear capacity using United States' enriching services to about 225 000 MW(e) in 1980.

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[1] Median value for the forecast range 130 000 to 170 000 MW(e).

[2] Excluding the United Kingdom capacity for which the United Kingdom plans to furnish enriching services.

SEPARATIVE WORK REQUIREMENTS

There are several uncertainties which affect the projected future demand for separative work. While projections of the growth rate of electric generating capacity have proven to be fairly reliable in the past, there are new factors, and in particular those associated with the rising world-wide concern for the environment which could modify them in the future. Projections of nuclear power growth will depend not only on the growth rate of the power industry as a whole, but also on the incidence of delays in reactor start-up, the changes in projected long-term competition between nuclear and fossil fuels, and nuclear plant performance experience. Each of these factors will probably have some influence on the rate at which utilities decide to build nuclear plants to meet their generating requirements.

In addition, there are at least three technological developments which can be projected over the next two decades, which, if successful, will tend to conserve enriched uranium and therefore reduce demands on enriching capacity. In the possible order of their occurrence, they are:

(a) The introduction of plutonium recycle which we project will occur in the time period 1974 - 1976.
(b) The possible large-scale introduction of advanced 'converter' reactors (HTGR) in the late 1970's or early 1980's.
(c) The large-scale introduction of the breeder reactor as a fully competitive power source in the middle or late 1980's.

The recycle of plutonium could reduce the total separative work requirements by about 5% in the 1974 - 1980 period and 10% in the 1981-1985 period. On an individual reactor basis, plutonium recycle is expected eventually to reduce separative work requirements by about 30%. Relative to the light-water reactors, the fuel cycle for the high-temperature gas reactors may lead to higher initial but lower annual reload requirements for separative work. The successful introduction of the breeder reactor will produce further changes in the requirements for separative work, eventually bringing about a decrease in quantities.

Annual separative work requirements from the US enrichment plants are being projected with plutonium recycle starting in 1974 and the introduction of breeders starting in the mid 1980's.

LONG-RANGE PLANNING

We in the United States recognize our responsibility to plan for the anticipated growth in separative work demands. Because of the several uncertainties previously mentioned, our plans must permit considerable flexibility in the operation and expansion of capacity in the United States. The plans also must recognize that other countries have or will develop the capability to supply enriching services. Our projections of separative work to be supplied from plants in the United States include assumptions that increasing fractions (5% in 1975 and 35% in 1985) of the market which we might supply will, in fact, be supplied by non-US plants.

One of the key variables in the planning process is the ^{235}U assay of the tails stream of the enriching plant. If power and operating costs (which

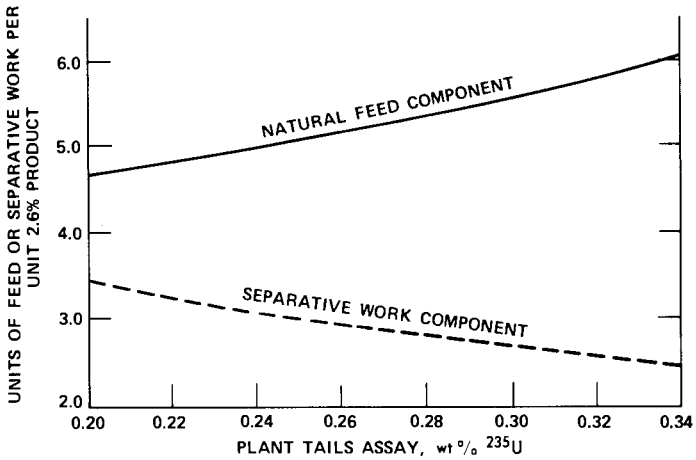

FIG. 1. Separative work and natural feed to produce one unit of 2.6% ^{235}U product versus plant tails assay.

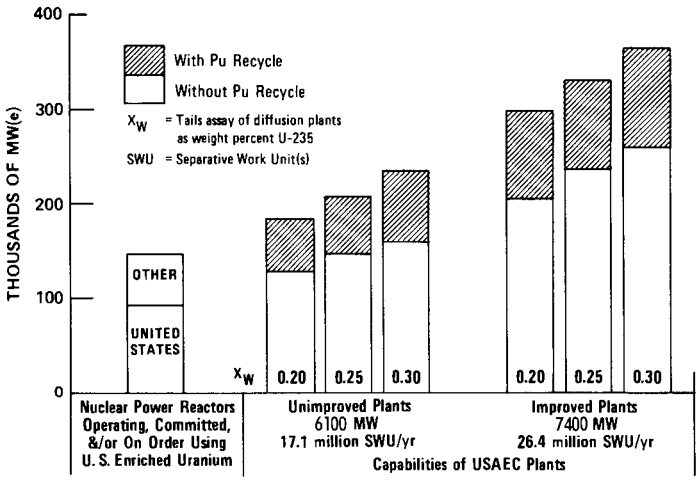

FIG. 2. MW(e) sustaining capacity of gaseous diffusion plants for light-water reactors.

directly affect the cost of separative work) increase, the economics suggest reducing the amount of separative work per unit of enriched product. This result is obtained by increasing the amount of normal uranium feed to the enriching plant which has the effect of increasing the assay of the waste or tails stream. Conversely, if the feed costs increase, then it becomes desirable to use less feed and more separative work and the tails assay should decrease. This relationship of feed and separative work components as a function of 'tails assay' is shown in Fig. 1. The relative effects of the price trends of these two components interact to determine an optimum combination of separative work and feed, thus resulting in an optimum tails assay for a given time period.

As one can see, optimizing the enrichment plant operations requires some ability to forecast reasonably well trends in electric power and operating costs as well as trends in feed costs. These latter are in turn related to sources of supply. The cost of electric power furnished to the gaseous diffusion plants has been on the increase and this was the primary cause of the recent upward adjustment in our charge for separative work (from US$28.70 to US$32 per unit).

Figure 2 shows the importance of varying the enrichment plant feed rate and tails assay, of utilizing plutonium recycle, and of improving existing plants in satisfying increasingly higher nuclear electric generating capacities. As this figure shows, there is considerable flexibility in how existing plants may be used to support growing nuclear power demands.

PRESENT PLANT CAPABILITIES

For the fiscal year ending June 30, 1972, our three gaseous diffusion plants will use an average of about 2700 megawatts of electricity which will result in the production of about 8.5 million units of separative work for the year. This represents about 44% of their present maximum potential power utilization of 6100 megawatts of electricity and about 50% of the present maximum separative work that can be produced from them. At these reduced operating levels we are still producing in excess of current requirements. Our operating plans call for continuing preproduction. The present preproduction inventory is adequate to satisfy nuclear power requirements for about the next two years.

Through carefully planned and co-ordinated equipment design, fabrication, operation and maintenance programs, a routine on-stream operating efficiency of greater than 99% has been achieved in the gaseous diffusion plants. The Atomic Energy Commission is planning for additional power procurement to return the existing three-plant complex to its full utilization of 6100 megawatts of electricity by 1978.

EXPANSION OF PRESENT PLANT CAPABILITIES

In addition to restoring electric power use to the design level, which will double the present output of separative work, there are plans to improve the large separating stages to incorporate advancements in technology made over the past decade which will improve the efficiency of power use. The average power utilization index (PUI) for the existing gaseous diffusion plants when fully powered at 6100 megawatts is 7.7 separative work units per megawatt day of electricity consumed. With advanced technology which can be incorporated in the present plants during the next several years, the PUI can be increased to 9.8 separative work units per megawatt day of electricity. The planned improvements will cost from US$500 - 600 million and will provide an additional 4.8 million units of separative work per year when the plants are powered at 6100 megawatts of electricity. One important benefit of this improvement program, which we have termed our Cascade Improvement Program (CIP), is that the increase in separative capacity can be achieved with no increase in the amount of electric power used and no increase in operating cost. It clearly provides increased capacity at the lowest possible unit cost.

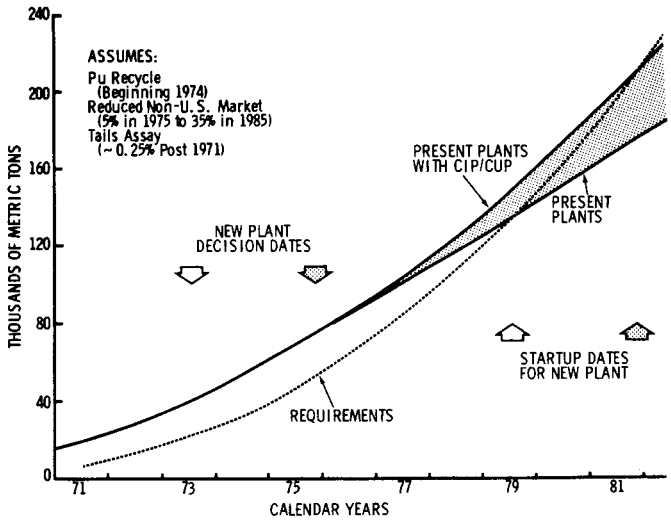

FIG. 3. Cumulative separative work availability versus requirements.

Either following or during the CIP, the plant equipment can be uprated to increase the power input. Increasing power to the extent that it can be efficiently used will permit an increase in the mass flow rate of process gas, thereby increasing the output of the improved plant with only very slight reduction in the average PUI. An increase in power level of 1300 megawatts of electricity to a total nominal level of 7400 megawatts of electricity will require equipment modifications costing from US$200-300 million and will result in additional separative work production of 4.5 million units per year. The CUP will involve additional operating costs for power purchases of about US$65 million per year.[3] While the attractiveness of this method of capacity expansion, which we have termed our Cascade Uprating Program (CUP), is sensitive to rising power costs, it continues to be economically desirable in comparison with the construction of new plant.

Programmed restoration of electric power to the present plants together with the improvement and uprating programs for them should meet foreign and domestic separative work requirements for about the next decade. The separative work requirements and availability for this period are compared in Fig. 3.

The President of the United States recently announced initiation of a program to increase the capacity of the three existing uranium enrichment plants in the United States. The President noted that: "There is currently no shortage of enriched uranium or enriching capacity. In fact, the Atomic Energy Commission has substantial stocks of enriched uranium which have already been produced for later use. However, plant expansions are required so that we can meet growing demands for nuclear fuel in the late 1970's - both in the United States and in other nations for which this country is now the principal supplier."

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[3] Based upon 5.55 mills/kWh.

NEW PLANT PLANNING

The needs for separative work which the Atomic Energy Commission projects may be requested of the United States in the early 1980's will exceed the capability of the present diffusion plant complex, even after the planned power restoration, process improvement, and power uprating programs have been implemented. Current assumptions regarding nuclear power growth, plutonium recycle, plant operating 'tails assay', and partial supply of markets outside the United States suggest, for example, that additional capacity amounting to about 20 million units of separative work per year will be required by 1985. The expected rate of increase in requirements is such that this new capacity could be added at a levellized annual rate of about 5 million units of separative work per year, beginning in about 1982. The date new plant construction planning and design should start is sensitive to the kinds of assumptions and variables illustrated in Fig. 3.

While the projected date of 1982 for new plant in the United States might appear to some to remove the urgency of the question of building new enriching capacity, any comfort taken will be short-lived. We believe that the commitment date for a new plant must, at a minimum, precede operation by about six years. Thus, the decision year for a new plant is not more than about four short years away. This time schedule is important to us in the United States both in terms of what we shall be doing domestically and how we shall look to strengthening our bonds of co-operation abroad.

While considerable work has been done in the United States and elsewhere on other processes, especially gas centrifugation, we believe the gaseous diffusion process is the only proven process at this time suitable for the large-scale enrichment of uranium. Therefore, present estimation of new plant requirements is based on the gaseous diffusion process.

In 1967, the Atomic Energy Commission initiated a series of scoping studies to define better the cost and probable characteristics of new plants. In addition, for a number of years, work has been in progress looking at advanced concepts to provide more efficient and less costly new stage systems. While most of the technologies developed for the CIP/CUP have direct application in new plants, especially barrier and compressor improvements, the design freedom associated with new stages offers possibilities for significant additional improvement.

New stage design, development, and testing programs are being planned and conducted in order to have the best design available when new capacity is needed. In addition to continued programs in barrier and compressor development, alternatives and innovations in areas such as energy recovery, process heat removal, compressor drive schemes, stage aerodynamics, and the design of other plant components are being evaluated. Emphasis is being placed on improved energy utilization and lower capital costs, while maintaining the high degree of equipment reliability that we have experienced in the existing gaseous diffusion plants. Technology for new plants on a time schedule consistent with projected new plant requirements is expected to yield an average PUI of 10.0 or more.

Whether by adding to existing plants or by entirely new plants, we believe new plant capacity in the United States can be counted on to meet separative work demands in the future, and at a reasonable unit charge for separative work, provided that needs are identified and firm commitments are made on a timely basis.

In his energy message, President Nixon noted that: "With the exception of uranium enrichment, the nuclear energy industry is now in private hands," and added, "I expect that private enterprise will eventually assume responsibility for uranium enrichment as well." It was explicitly in the context of this long-standing Presidential policy that our Commission recently announced a program for affording to a limited number of private industrial companies in the United States access to enriching process technologies which the Commission has developed over the past quarter of a century.

Our Commission stated that this program will afford American industry the opportunity, which has heretofore been denied it, to determine realistically the role it desires to assume in the enriching phase of the nuclear fuel cycle.

We are confident that the response by private industry to the Commission's initiative will be positive and that the private sector of our economy will make a major contribution to new enriching capacity on the time-scale that we now believe will be necessary.

From the standpoint of our customers in other countries, we should also stress that their contractual interests will be fully protected should enrichment functions eventually be transferred to private American enterprise. We will make certain that whoever is involved in the enrichment business in our country will comply with the fundamental obligations which have been undertaken by our Commission in this field.

CAPITAL COSTS

Attention has been called to the large capital investments required for an efficient gaseous diffusion plant and to the sensitivity of separative work costs to the cost of electric power for the plant. It is perhaps worthwhile to put the capital cost and electric power requirements of large gaseous diffusion plants in perspective in relation to the nuclear power plants which they serve. Enrichment capacity of one separative work unit per year will support from 8 kW[4] to nearly 15 kW[5] of nuclear power capacity in current large water reactors. At a PUI of 10 separative work units per megawatt day, and assuming an average load factor of 80% for the power supply, each kilowatt of power capacity supplying input to the gaseous diffusion plant would produce about 3 separative work units per year. Therefore, for each kilowatt of power capacity supplying the enrichment plants, its separative work production will sustain 24 to 45 kilowatts of nuclear power capacity depending upon the modes of operation of the enrichment plants and the power plants. The capital cost required for enrichment capacity, including associated power supply, would amount to something like 5 to 10% of the capital cost of the nuclear power plants being served.

URANIUM ENRICHMENT CONTRACTS

For many years the Atomic Energy Commission has been firmly committed to the principle of satisfying, within its capabilities, the long-term uranium enriching requirements of both domestic and foreign nuclear power plants. We have recognized from the earliest days that the growth

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[4] Tails assay of 0.2% ^{235}U, no plutonium recycle.

[5] Tails assay of 0.3% ^{235}U, with plutonium recycle.

of nuclear power both at home and abroad depends on the assured long-term availability of ^{235}U under relatively stable and attractive conditions. The Commission's program of toll enrichment, which began on January 1, 1969, permits contracts with customers in the United States and abroad under which the customer delivers feed material for enrichment in USAEC facilities. An appropriate charge is made for the enriching services. The quantity of feed to be provided in relationship to the quantity of enriched uranium desired and the related amount of separative work required are determined in accordance with AEC's standard table of enriching services.

Two types of enriching service contracts are available. One is called a Firm Quantity Contract and is normally used for short-term requirements. The more commonly used contract is a Requirements Contract which provides for the furnishing of all or a specified part of the customer's requirements for enriching services for a designated facility or facilities during the contract term which can run as long as the anticipated economic life of the reactor subject, in the foreign cases, to the terms of the applicable government Agreement for Co-operation. Both feed material and product are in the form of UF_6. The customer has an option to acquire tails or depleted uranium resulting from the performance of the enriching services. An enrichment contract may be terminated by the customer without charge by giving a specified period of notice. Provided certain conditions are met, which assure the customer of continuing supply under reasonable terms, the Atomic Energy Commission may terminate a contract in the event commercial (non-government) enriching services become available in the United States.

Our business of providing enriching services has been increasingly active and has reflected the rapid growth of the nuclear power industry. We had executed through April 1971 a total of 67 contracts including 35 with customers outside the United States for about 139 million units of separative work, 37 million of which are for distribution abroad.[6]

FUTURE TRENDS

The rapidly increasing requirements for uranium isotope enrichment for the growing nuclear power industry in many parts of the world naturally will lead to change in the pattern of availability of services. The capacity of the enrichment plants now existing in the United States, expanded in accordance with planned improvements, will be sufficient to meet projected requirements for about 10 years. Considering the lead time required to put new capacity into operation and the fact that orders for nuclear power plants depend upon assurance that the required enrichment capacity will be available when needed, it is natural that there has been mounting interest and activity both in the United States domestically and abroad in the isotope enrichment field. Since the United States has been the principal world supplier of enrichment services, it also is understandable that there should be great interest in future enrichment policies of the United States.

The availability of substantially more capacity than that needed in recent years has made it possible for the United States to make uranium enrichment services available to the electric power industry, both at home and abroad,

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[6] Based upon operating the separation plants at a 0.2 tails assay.

under agreements containing great flexibility for the customer, including extremely favourable termination provisions. As the available capacity becomes fully committed, and it becomes necessary for major capital investments to be made in new capacity and related power supply facilities, these investments will require the protection of a more adequate termination notice and associated payment provisions than we have used in the past.

It is the United States Atomic Energy Commission's objective that adequate uranium enriching capacity be available to support the growth in nuclear power including needs of domestic and foreign enriching customers.

In looking toward the future, we are also mindful of the interests of other countries in alternative sources for enrichment services. This question was addressed in President Nixon's second annual review of United States foreign policy. This review, issued February 25, 1971, includes the following statement:

"Having carefully weighed the national security and other factors involved, we have undertaken consultation with the Joint Committee on Atomic Energy of the Congress concerning ways in which the United States might assist our allies to construct a multi-nation uranium enrichment plant to help meet future world demands."

We are pleased to report that following these consultations with the Congress that the President mentioned, we undertook to notify a number of nations in Europe and the Pacific that had indicated an interest in exploring the possibility of a multi-national uranium enrichment project of our willingness to enter into exploratory discussions on the matter. These discussions may commence this fall and would serve to determine whether there would be sufficient interest to move into detailed negotiations. From our standpoint, we would be prepared to discuss possible arrangements for making available, for fair compensation, our advanced gaseous diffusion technology and know-how for the establishment, on a mutually agreeable schedule, of multi-national enrichment facilities of substantial capacity subject to appropriate controls against the unauthorized disclosure of sensitive process technology outside of those participants in the project having a need to know and use it. We would, of course, expect that the product of any multi-national project would be subject to the appropriate safeguards arrangements to assure its use for peaceful purposes only.

CONCLUSION

We are moving into a new and stimulating period in the translation of the atom's legendary promise into real energy at the service of mankind. Perhaps the largest single enterprise to which we will have to direct our attention in the coming decade is the one of assuring together, fairly and equitably, that the vast enriching facilities which this burgeoning industry will require are available on time, that they can furnish enriching services at assured low costs, and that they are so organized as to reassure all customers that this vital link in the nuclear fuel cycle is not the monopoly of any single interest.

In this new venture, the Government and industry of the United States will, we are certain, make their full contribution in the spirit of co-operation that has been the special character of nuclear affairs since the first Geneva Conference sixteen years ago.

DEVELOPPEMENTS PREVUS PAR LA FRANCE DANS LE DOMAINE DE LA SEPARATION ISOTOPIQUE DE L'URANIUM

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Abstract–Résumé–Аннотация–Resumen

FRENCH DEVELOPMENT PLANS FOR THE ISOTOPIC SEPARATION OF URANIUM.

In the fuel cycle the importance of separating uranium isotopes is self-evident. France is pursuing a major program in this field, designed to select and develop the process best suited to providing for her needs and those of other Western countries. First and foremost, French scientists are currently engaged in studying prospects for constructing a gaseous diffusion plant large enough to meet a substantial part of European requirements by the end of the present decade. The purpose of these studies, which are described in the paper, is the compilation of a feasibility report relating to construction of such a plant. It will be based partly on experimental findings, more particularly results obtained at the full-scale pilot plants built and put into operation at Pierrelatte, and partly on a detailed industrial project. The latter project will provide precise information on the cost of building and operating a plant of this kind, and will deal in addition with various problems relating to the economics and operation of the plant, including the supply of power, together with optimization of the plant/grid system, and the gradual start-up of facilities so as to keep pace with demand as effectively as possible. Indications will be given as to the economic outlook for such a project. Work in France, however, is not restricted to gaseous diffusion studies, although it is felt that this process would best fit in with the commissioning of a plant within the scheduled period; work is in progress on ultra-centrifuge methods in view of their potential for improvement on a long-term basis. Indications will be given on such important aspects of the matter as trends in the cost of machinery in relation to the size of production runs, the overall design of a plant, and the relationship between the total cost of the plant and that of the basic machinery. The economic outlook for the process as compared to other methods is touched on at the end of the paper, together with prospects for the next few years, as viewed within a European and world context.

DEVELOPPEMENTS PREVUS PAR LA FRANCE DANS LE DOMAINE DE LA SEPARATION ISOTOPIQUE DE L'URANIUM.

L'importance de la séparation des isotopes de l'uranium dans le cycle de combustible est évidente. La France, pour sa part, poursuit un effort important dont le but est de choisir et de mettre au point le meilleur procédé pour répondre au problème posé par son approvisionnement et par celui des autres pays occidentaux. La France étudie tout d'abord les perspectives de réalisation d'une usine de diffusion gazeuse dont la taille devrait répondre à une part importante des besoins européens à la fin de la décennie. Ces études, dont il est rendu compte dans le mémoire, visent à la constitution d'un dossier de construction de cette usine. Ce dossier sera basé d'une part sur les résultats expérimentaux et en particulier sur ceux des pilotes à l'échelle 1 qui sont réalisés et exploités à Pierrelatte, et d'autre part sur un projet industriel détaillé. Ce projet permettra d'avoir des indications très précises sur le coût de construction et d'exploitation d'une telle usine. De surcroît, différents problèmes liés à l'économie et à la mise en service d'une telle usine sont évoqués: tout d'abord, le problème de l'alimentation en énergie avec en particulier l'optimisation de l'ensemble usine-réseau, ensuite, les problèmes de démarrage progressif des installations pour pouvoir suivre au mieux l'évolution de la demande. Des indications sont données sur les perspectives économiques d'un tel projet. Mais la France ne se limite pas aux études de diffusion gazeuse, bien qu'elle pense que ce procédé soit mieux adapté au lancement d'une usine dans les délais envisagés; elle travaille en particulier l'ultra-centrifugation du fait du potentiel d'amélioration que ce procédé peut éventuellement présenter à long terme. Des indications sont fournies sur certains problèmes importants comme l'évolution des coûts des machines en fonction de la série, la conception générale d'une usine et le rapport entre le coût total de l'usine et celui des machines élémentaires. Le mémoire aborde enfin les perspectives économiques de ce procédé en comparaison avec celles des autres procédés, ainsi que les perspectives possibles au cours des prochaines années dans leur contexte européen et mondial.

ФРАНЦУЗСКАЯ ПРОГРАММА ПО РАЗДЕЛЕНИЮ ИЗОТОПОВ УРАНА.

Важность процесса разделения изотопов урана для топливного цикла не вызывает сомнений. Во Франции основная цель исследовательской программы связана с выбором и разработкой метода разделения, который лучше всего обеспечит потребности страны в снабжении ураном, а также потребности других европейских стран. Во Франции, в первую очередь, исследуются перспективы проектирования и строительства газодиффузионного завода по разделению изотопов урана, который обладал бы достаточно высокой производительностью и смог бы покрыть основную долю потребностей Европы в уране в ближайшем десятилетии. Эти исследования, результаты которых излагаются в докладе, предназначены для разработки проекта строительства такого завода. Проект будет основываться на экспериментальных данных, в основном на данных, полученных при эксплуатации полупромышленных установок, построенных и эксплуатируемых в Пьерлатте, а также на детальных экономических расчетах. Этот проект даст точные данные по стоимости строительства и эксплуатации заводов такого типа. Кроме того, будут рассмотрены различные проблемы, связанные с экономикой и эксплуатацией заводов такого типа: во-первых, проблема энергоснабжения и, в особенности, проблема оптимизации общей системы – завод/энергосистема; и проблема дальнейшего ввода в эксплуатацию последующих установок по разделению изотопов урана с целью покрытия потребностей в уране наиболее рациональным способом. Даются сведения по экономическим аспектам такого проекта. Научно-исследовательские работы, которые проводятся во Франции, не ограничиваются рассмотрением газодиффузионных заводов по разделению изотопов урана, хотя этот метод, по-видимому, является наиболее перспективным для проектирования и строительства завода в приемлемые сроки. Среди других методов исследуется, главным образом, метод с применением ультрацентрифуги в связи с большими возможностями его усовершенствования. Даются сведения по некоторым основным проблемам: например, зависимости стоимости оборудования от производительности завода, общей конструкции завода и отношению общей стоимости завода к стоимости отдельных видов оборудования. Наконец, в докладе делается сравнение экономических показателей этого метода разделения с другими методами, а также рассмотрены перспективы этих исследований на ближайшие несколько лет в европейском и мировом масштабе.

PERSPECTIVAS FRANCESAS DE DESARROLLO EN EL CAMPO DE LA SEPARACION DE LOS ISOTOPOS DE URANIO.

Es evidente la importancia de la separación de los isótopos del uranio en el ciclo del combustible. Francia, por su parte, mantiene un esfuerzo importante cuyo fin es seleccionar y poner a punto el procedimiento que responda mejor al problema planteado por su aprovisionamiento y por el de los otros países occidentales. En primer lugar, en Francia se estudian las perspectivas de realización de una fábrica por difusión gaseosa cuyo tamaño permitiría hacer frente a una parte importante de las necesidades europeas al final del decenio. Estos estudios, de los que se da cuenta en la memoria, van dirigidos a la preparación del conjunto de especificaciones para su construcción, basado, por un lado, en los resultados experimentales y, en particular, en los ensayos piloto a escala 1 que se realizan y aprovechan en Pierrelatte y, por otro lado, en un proyecto industrial detallado. Este proyecto permitirá obtener indicaciones muy precisas sobre el coste de construcción y de explotación de la fábrica. Además se pondrán de manifiesto diferentes problemas ligados a su economía y puesta en servicio; primeramente, el del suministro de energía, incluyendo, en particular, la optimización del conjunto fábrica-red; a continuación, los problemas de la entrada progresiva en servicio de las instalaciones para poder seguir de la mejor forma posible la evolución de la demanda. Se aportan datos sobre las perspectivas económicas de un proyecto de este tipo. Ahora bien, los estudios que se llevan a cabo en Francia no se limitan a la difusión gaseosa y, aunque se opina que este procedimiento es el más adecuado para el lanzamiento de una fábrica en los plazos previstos, se trabaja en particular, en la ultracentrifugación debido a los posibles perfeccionamientos que este procedimiento puede llegar a ofrecer a largo plazo. Se aportan indicaciones sobre ciertos problemas importantes como la evolución de los costes de las máquinas en función del tamaño de la serie, la concepción general de una fábrica y la relación entre el coste total de la misma y el de las máquinas elementales. Finalmente, se abordan las perspectivas económicas de este procedimiento en comparación con las de otros sistemas, así como las perspectivas posibles en el curso de los próximos años en su contexto europeo y mundial.

La situation générale dans le domaine de l'enrichissement isotopique de l'uranium à des fins civiles est bien connue. Nous nous contenterons donc d'en rappeler brièvement les grandes lignes dans la présente introduction.

La courbe des besoins en uranium enrichi d'ici 1985, date à laquelle les surrégénérateurs pourraient prendre place sur le marché de l'énergie, présente une allure rapidement croissante. Les points de passage de la demande occidentale exprimée en puissance de séparation annuelle sont proches de 18 M UTS en 1975, 36 M UTS en 1980, 72 à 75 M UTS en 1985, autrement dit, ils traduisent un doublement des besoins tous les cinq ans. En outre, ce marché est géographiquement dispersé. La consommation prévue en 1985 se répartit à raison de 40 M UTS aux Etats-Unis, 20 M UTS en Europe, 10 M UTS au Japon, 5 M UTS dans le tiers monde. Face à cette demande, l'USAEC, qui est actuellement le seul producteur occidental, dispose d'une capacité de séparation de 17 M UTS qui sera éventuellement portée à 25 M UTS si l'ensemble du projet CIP/CUP est adopté. Quelle que soit la décision prise, une insuffisance de production se dessine entre 1977 et 1979. A cette époque, la croissance du marché occidental sera de l'ordre de 5 M UTS/an. Compte tenu d'une durée de construction d'usine de 5 ans, il apparaît que la décision d'entreprendre une telle opération est maintenant très proche.

Quels sont les procédés en présence ? Essentiellement deux pour l'instant : la diffusion gazeuse et l'ultracentrifugation.

La diffusion gazeuse a fait ses preuves dans plusieurs pays et, de ce fait, ne présente guère de risques ni sur le plan technique, ni sur le plan économique. C'est un procédé bien adapté aux grandes capacités, souple, et très endurant.

En revanche, ce n'est pas nécessairement le plus attrayant sur le plan économique à long terme, en raison de sa forte consommation spécifique d'énergie qui entre pour près de 40 % dans le coût de l'unité de travail de séparation. Sous cet angle, l'ultracentrifugation, dont la consommation d'énergie n'intervient que pour 10 % dans le coût de l'unité de travail de séparation, est tres séduisante. Par contre, elle fait appel à un investissement plus important dont le montant dépend étroitement de l'aptitude aux très grandes séries des fabrications choisies et de l'efficacité des méthodes appliquées. En outre, sa mise en œuvre, qui nécessite un réseau aérodynamique complexe, doit être préalablement testée à échelle industrielle et cette étape, nous le savons, ne doit débuter qu'en 1972.

Il ressort, de ce rapide survol de la situation, deux considérations essentielles :

1) l'impératif de non-pénurie oblige à faire appel au procédé de diffusion gazeuse pour les usines qui devront entrer en production avant la fin de la présente décennie,
2) l'ultracentrifugation présente des perspectives intéressantes à long terme, et à ce titre, mérite attention et encouragements.

Le programme des activités françaises dans le domaine de l'enrichissement tient largement compte de cette situation. Il se place cependant dans une optique plus large et plus dynamique qui comprend les caractéristiques prévisibles du marché mondial, les possibilités technico-économiques qui sont offertes et les avantages qui peuvent découler d'une implantation géographique et d'associations internationales appropriées. Ces différents facteurs interviennent dans la poursuite d'une politique d'ordre essentiellement commercial.

Il apparaît en effet que face à la croissance du marché, et compte tenu des moyens d'enrichissement existants ou prévisibles, une usine de diffusion gazeuse de grande capacité devra être lancée d'ici 1973. Réalisée dans des conditions appropriées, cette usine apparaît non seulement comme un premier complément de capacité indispensable, mais également comme une entreprise qui peut être particulièrement compétitive par rapport aux capacités existantes.

Au-delà de cette première étape, la croissance du marché ainsi que l'uniformisation progressive du coût de l'énergie dans les pays développés favoriseront les procédés à faible consommation spécifique d'énergie tels que l'ultracentrifugation. Toutefois, il apparaît que l'échéance plus lointaine de cette deuxième étape laisse place à la mise au point de procédés nouveaux. D'où le 2ème volet de notre action qui consiste à explorer au maximum l'éventail des nouveaux procédés offerts et de trouver un procédé qui puisse devenir compétitif à long terme.

Autrement dit, en se plaçant délibérément dans une économie de marché, et en développant au maximum les possibilités de concertation internationale qui sont offertes, le programme français se présente actuellement en deux points :

1) Préparer une construction d'usine de séparation isotopique par diffusion gazeuse de 6 à 8 M UTS commençant à produire en 1978 et entièrement opérationnelle en 1980.

2) Préparer un éventail de connaissances de séparation isotopique faisant appel aux procédés qui présentent les meilleures perspectives pour les années 80.

PREMIER OBJECTIF

Construction d'une usine de diffusion gazeuse de 6 à 8 M UTS

Il est évident qu'un tel projet repose largement sur l'acquis détenu par la France dans le domaine de la séparation isotopique. Cet acquis fait l'objet d'une autre communication présentée ici même[1]. Nous nous bornerons donc à insister sur ce que signifie PIERRELATTE sur le plan de la construction (arrière-plan industriel, montage industriel, ordonnancement), sur le plan du démarrage (modèles mathématiques permettant de prédire et d'optimiser les transitoires de concentration), sur le plan de l'exploitation enfin (automatisation du contrôle analytique, conduite assistée d'ordinateurs en temps réel, entretien prévisionnel, contrats d'intéressement, etc), et nous ajouterons que les équipes qui ont apporté ainsi la preuve de leur compétence ont été maintenues.

Pour en revenir au projet lui-même, nous allons successivement décrire sa progression et approfondir quelques-uns de ses aspects économiques.

Sa progression résulte d'un faisceau d'activités convergentes: essentiellement des études de réalisation et des démonstrations de pilotes industriels.

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[1] LEDUC, C. et al., ces Actes, vol.9, mémoire 605.

Les études de réalisation comportent trois phases :
- le choix du projet,
- la définition du procédé,
- l'élaboration du projet industriel détaillé.

Voyons le choix du projet tout d'abord.
Il repose sur :
- une étude détaillée du marché dont sont extraites la capacité, la date du début de production de l'usine, et les conditions de son insertion progressive dans le marché,
- une étude approfondie du coût de l'énergie en se basant, d'une part sur les prévisions les plus solidement établies des coûts de production et en tenant compte, par ailleurs, de toutes les particularités offertes par le client de choix que représente une usine de diffusion gazeuse pour un fournisseur d'électricité,
- une expression des investissements nécessaires en fonction de la taille de l'usine,
- une étude approfondie des possibilités de financement et des conditions d'amortissement.

Cet ensemble est introduit sur un modèle mathématique souple, c'est-à-dire largement paramétré, susceptible d'explorer de multiples hypothèses et de circonscrire les solutions intéressantes.

Voyons ensuite la définition de procédé.
Elle repose essentiellement sur :
- une première définition suivie d'une optimisation de la cellule de séparation et de ses conditions de fonctionnement qui permettent d'établir une spécification générale de chacun de ses principaux constituants,
- une définition de l'agencement de l'assemblage,
- une définition de ses équipements annexes et des principales servitudes nécessaires.

Les éléments de la définition de procédé sont, eux aussi, en grande partie programmés.

Voyons enfin l'établissement du projet industriel détaillé.
C'est de loin l'étape la plus longue. Elle rassemble, en effet, sous la forme la plus détaillée, les dessins d'étages, définitions d'annexes, plans d'implantation, spécifications d'équipements, plans de distribution d'énergie, réseaux auxiliaires, dispositifs de conduite et de gestion, environnement utilitaire et logistique. Commencée en 1969, elle ne sera achevée qu'en 1972 en même temps que le dossier de construction dont elle constitue un élément essentiel.

A l'appui de ces études, et dans le but d'apporter la preuve tangible du caractère réaliste et industriel des réalisations correspondantes, nous avons construit par ailleurs un ensemble de pilotes à échelle 1 dont voici brièvement l'historique.

Les études préalables d'une usine civile nous ayant permis d'opter dès 1968 pour une usine de capacité comprise entre 6 et 10 M UTS et de préciser que cette usine serait uniquement constituée d'étages en série, nous avons alors dégagé les caractéristiques de deux étages types, respectivement parcourus par des flux nominaux de 40 kg/s et 160 kg/s d'UF_6. La décision était prise peu de temps après d'une

part de passer commande à l'industrie française de compresseurs prototypes, d'autre part d'entreprendre la construction de pilotes de démonstration sur le site de PIERRELATTE. Ces pilotes, aujourd'hui achevés, sont désignés par PP 200 et PP 300. Chacun d'entre eux comprend plusieurs circuits destinés d'une part à essayer des compresseurs de types aérodynamiques différents, d'autre part à tester des montages de diffuseurs aussi économiques que possible. Outre les appareils de base que constituent le compresseur et le diffuseur, chacun de ces pilotes comporte également un échantillon complet des autres appareillages nécessaires : échangeur, vannes, joints, etc. Deux prototypes de compresseurs de 40 kg/s essayés en 1969 et en 1970 ont donné entière satisfaction tant du point de vue aérodynamique que du point de vue mécanique. A présent, le premier prototype de 160 kg/s vient d'être livré et devrait démarrer prochainement.

La réussite technique que représente le bon fonctionnement de ces pilotes est évidemment spectaculaire. Nous voudrions insister sur l'importance des renseignements d'ordre économique qu'apporte une telle expérience car elle oblige à passer en revue toutes les phases de la construction, de la réalisation, et de l'exploitation. Ajoutés à la connaissance générale que nous possédons des usines de diffusion gazeuse, ces renseignements permettent d'établir un devis exact de l'investissement nécessaire. De là, et en fonction des valeurs attribuées au prix de l'énergie et à l'intérêt de l'argent, il est possible d'établir un coût prévisionnel de l'unité de travail de séparation extrêmement faible.

Après avoir exposé la méthode et décrit les éléments sur lesquels reposera le dossier de construction de l'usine de diffusion gazeuse, nous allons maintenant approfondir les deux éléments économiques majeurs que sont le coût de l'énergie et les risques de pénalisation au démarrage.

Des études ont été entreprises avec EDF ayant pour but de déterminer le coût moyen du kWh consommé par une usine de diffusion gazeuse.

Dans une première étape, nous avons essayé d'évaluer les coûts prévisionnels du kWh délivré aux bornes des centrales. Nous avons pris en considération plusieurs types de centrales ainsi que les paramètres qui pouvaient modifier le coût de l'énergie délivrée. Nous avons recoupé les chiffres obtenus avec les valeurs figurant dans les récents contrats de centrales nucléaires. Nous avons enfin tenu compte d'une part des actualisations économiques nécessaires, d'autre part des améliorations techniques devant accroître l'efficacité de la production d'énergie.

Dans une deuxième étape, nous avons tenu compte des avantages dont pourrait bénéficier une usine de diffusion gazeuse à savoir, le niveau et la régularité de la consommation d'une telle usine et l'absence de frais annexes dus à l'appartenance à un complexe industriel.

A cela s'ajoutent les avantages qu'offrent des possibilités de délestage en cas de pénurie temporaire d'énergie. Ce point est apparu extrêmement important pour deux raisons : la première est la nature de l'énergie ainsi rendue disponible qui s'apparente à un surcroît d'énergie hydraulique en raison de sa grande souplesse d'utilisation, la deuxième est l'implantation judicieuse de cette possibilité dans des régions dépourvues d'hydraulicité, ce qui permet une optimisation du réseau dont découlent des économies considérables.

Au total, les chiffres obtenus concernant le coût de l'énergie au cours des années 1970/75 entrent dans la fourchette comprise entre 3 et 4 centimes/kWh et les possibilités d'interruptibilité permettent d'obtenir une tarification extrêmement avantageuse.

Une autre question importante concerne la mise en production d'une usine de grande capacité. Des craintes ont été exprimées concernant le manque de souplesse d'un tel ensemble. En raison des pénalités économiques qu'un démarrage laborieux pourrait faire peser sur l'ensemble du projet, une étude approfondie destinée à explorer les conditions de démarrage a été entreprise. Plusieurs aspects ont été examinés :

- Tout d'abord, la durée théorique minimum pour placer l'usine initialement remplie en UF_6 naturel au voisinage de son régime nominal de concentration. Cette durée est relativement courte. Elle dépend bien entendu de la concentration maximum recherchée, mais n'excède pas un mois si cette concentration ne dépasse pas 5 %.
- Ensuite, la souplesse offerte par diverses stratégies de démarrage qui font alterner des périodes de production et des périodes de remplissage. Il est apparu que toute croissance moyenne comprise entre 1 et 3 M UTS/an pouvait être satisfaite économiquement.
- D'autres études ont été faites de manière à préciser les meilleures utilisations possibles de produits enrichis et appauvris soutirés au cours du démarrage : en effet, ces produits peuvent être directement utilisables dans certains types de réacteurs ou bien ils peuvent être réutilisés dans des phases ultérieures de la mise en concentration.

De cet ensemble d'études, il ressort qu'une usine de diffusion gazeuse est un instrument très souple susceptible de faire face à une grande variété de conditions qui seront à sélectionner en fonction des conditions commerciales à l'époque du démarrage.

En conclusion nous pouvons dire que le premier objectif du plan français est sur le point d'être atteint. Nous disposerons, en effet, dès 1972 d'un dossier de construction d'une usine de séparation isotopique par diffusion gazeuse de 6 à 8 millions d'UTS reposant sur 15 années d'expérience en séparation isotopique dont 8 années d'exploitation sans cesse améliorée de PIERRELATTE. Le dossier de construction lui-même aura nécessité 4 années d'études économiques et techniques y compris la réalisation et la mise à l'épreuve de pilotes à pleine échelle.

L'essentiel à présent est de mettre sur pied les structures industrielles nécessaires à la concrétisation d'un tel projet.

Pour cela, nous tenons essentiellement compte des problèmes liés au choix du site qui sont :

- une énergie à bas prix,
- une source de refroidissement,
- une main d'oeuvre adaptée, tant pour la construction que pour l'exploitation de l'usine,
- un financement approprié,
- une solide infrastructure industrielle.

Nous étudions ces problèmes dans un esprit de large ouverture européenne et mondiale, et procédons aux démarches qui permettent d'avancer rapidement dans cette voie.

Mais la France ne se limite pas aux études de diffusion gazeuse bien qu'elle pense que ce procédé soit mieux adapté au lancement d'une usine dans les premiers délais envisagés. D'où son second objectif.

SECOND OBJECTIF

Préparation d'un éventail de connaissances de séparation isotopique faisant appel aux procédés qui présentent les meilleures perspectives pour les années 1980

En effet, le CEA étudie d'autres procédés et en particulier l'ultracentrifugation du fait du potentiel d'amélioration que ce procédé peut éventuellement présenter à long terme.

Outre l'étude théorique du procédé de base et les études expérimentales effectuées sur bancs d'essai, une attention particulière a été accordée aux problèmes soulevés par l'application industrielle à grande échelle de ce procédé en raison notamment de la part prépondérante de l'investissement initial dans le coût de l'unité de travail de séparation.

Voici quelques réflexions à caractère général qui illustrent ce type d'études. Elles portent sur :

- la conception des usines,
- le facteur d'environnement,
- les prix de grande série.

Il est inutile d'insister sur toutes les caractéristiques de l'ultracentrifugation dont certaines différencient nettement ce procédé de la diffusion gazeuse. Cependant deux aspects particuliers méritent un développement car il est intéressant d'observer comment les impératifs liés à la réalisation d'un grand ensemble industriel peuvent en modifier la première apparence.

Il s'agit, tout d'abord, de l'extrême dissémination des éléments de séparation. Si on considère le bol comme élément séparateur de base dont la puissance de séparation n'excède pas quelques UTS, la réalisation d'une usine de plusieurs millions d'UTS revient à rasssembler, relier et organiser le fonctionnement de plusieurs centaines de milliers d'éléments. Ceci peut être fort coûteux en raison :

- de la multiplication des équipements annexes de l'élément séparateur,
- de la ramification des liaisons (tuyauteries et câbles),
- des impératifs de contrôle qui dépassent largement l'échelle humaine et nécessitent une automatisation pratiquement absolue.

Pour rester dans une zone de compétivité, il est indispensable de limiter au maximum le poids de ces extensions dont un indice particulièrement révélateur est le volume spécifique, c'est-à-dire le volume total de l'usine divisé par sa puissance de séparation. Nous avons entrepris une réduction systématique de ce volume spécifique, d'abord au niveau de la machine en incorporant plusieurs bols dans une même enceinte obtenant ainsi une multimachine, ensuite en groupant plusieurs multimachines afin de réaliser des modules de commutation et de conduite. Les améliorations apportées par de telles concentrations ont été mises en évidence au cours d'études successives qui ont permis de réduire le volume spécifique dans un rapport proche de 10

et de ramener ce dernier à des valeurs comparables à celles obtenues en diffusion gazeuse.

Un deuxième aspect qui a considérablement évolué est la manière de réaliser une cascade idéale. Il peut paraître séduisant en effet d'imaginer une usine rassemblant un grand nombre de petites cascades en parallèle et de chercher à identifier ces cascades avec les modules précédemment définis. Mais ce type de découpage prête à critique à mesure que l'on étudie les conséquences pratiques en matière d'implantation et que l'on recherche :

- des formes géométriques simples pour les bâtiments ainsi que l'occupation régulière et rationnelle des locaux,
- des axes de liaison et de circulation sous forme de réseaux à angles droits,
- un maximum de répétitivité pour les liaisons.

Ceci nous a conduit finalement à choisir des cascades de quelques centaines de 10^3 UTS qui résultent de l'assemblage en rangées de groupes parallèles identiques. L'empilement de rangées en nombre relativement limité et de largeurs différentes conduit à un profil de type squared-off dont l'efficacité reste supérieure à 95 %.

Un autre sujet d'étude a été le facteur d'environnement.

Le facteur d'environnement est le rapport entre le montant total des investissements de l'usine et le coût des centrifugeuses. Il demande a être défini avec soin. Il dépend des conditions dans lesquelles a été estimé le prix de fabrication des machines, conditions qui peuvent être entachées d'éléments conjoncturels. D'autre part, il dépend des limites choisies entre les machines et leur environnement. Sous réserve de ces remarques, il semble que la valeur du facteur d'environnement puisse se situer entre 2 et 3.

Enfin, nous avons étudié les prix de grandes séries de matériels répétitifs. Il est connu que l'existence de grandes séries conduit à une diminution sensible du prix des équipements dont une expression est donnée par la loi de CAQUOT. Généralisant cette loi, nous avons pu situer un certain nombre de coûts de grande série. Mais ce qui est peut-être plus important, c'est que les consultations faites en vue d'une usine de centrifugeuses ont mis en évidence un autre aspect de la question. Il apparaît que les prix de grande série ne diminuent plus en fonction inverse du nombre d'exemplaires au-delà d'une limite qui correspond à la saturation des moyens de production nécessaires à une gamme donnée (outillage, chaîne, usine). On peut admettre que ce phénomène touche la plupart des équipements importants d'une usine d'ultracentrifugation dépassant le seuil de 10^5UTS/an (machines, vannes, appareils de mesure).

Cette particularité entraîne deux conséquences :

- La première est que la concentration dans le temps et en un site donné d'une capacité de séparation ne s'impose pas comme en diffusion gazeuse. Il est envisageable de disséminer les installations sous forme d'usines locales qui n'auront pas besoin d'atteindre des tailles importantes.
- La seconde est que l'augmentation du nombre d'exemplaires ne permettant plus de gagner sur les investissements à partir d'un certain seuil, le problème se déplace alors pour se situer au niveau du compromis prix-qualité, la qualité rejaillissant sur la fiabilité du matériel ou sur la bonne marche du procédé.

Des considérations telles que celles que nous venons de citer en exemple permettent peu à peu de préciser les particularités et les contours des usines d'enrichissement de 3ème génération.

Au terme de cet exposé, nous espérons avoir donné un aperçu aussi complet que le permet une telle conférence des développements prévus par la France dans le domaine de la séparation isotopique de l'uranium, et avoir convaincu de l'étroite dépendance des efforts ainsi consentis aux futurs besoins en énergie dont il est temps d'entreprendre les réalisations de base.

CENTRIFUGE PLANTS IN EUROPE

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Abstract–Résumé–Аннотация–Resumen

CENTRIFUGE PLANTS IN EUROPE.

This paper briefly reviews the history of the development of the centrifuge in the Federal Republic of Germany, the Netherlands and the United Kingdom leading up to the agreement for collaboration between the three Governments in 1970. The initial plants now under construction which will be owned and operated by the Tripartite Enrichment Organisation are described. The potential market for this organization is reviewed and possible strategies for future expansion are discussed in the light of the technical and economic factors which will govern the commercial exploitation of the centrifuge as a uranium enrichment process.

USINES DE CENTRIFUGATION EN EUROPE.

Le mémoire fait brièvement l'historique de la mise au point de la centrifugeuse en République fédérale d'Allemagne, aux Pays-Bas et au Royaume-Uni, jusqu'à la conclusion de l'accord de collaboration entre les gouvernements de ces trois pays, en 1970. Il décrit les premières installations actuellement en construction qui seront propriété de l'Organisation d'enrichissement tripartite et dont celle-ci assurera l'exploitation. Il présente le marché qui s'ouvrira à cette organisation et analyse les politiques éventuelles de développement futur, compte tenu des facteurs techniques et économiques qui régiront l'exploitation commerciale de la centrifugeuse en tant que mode d'enrichissement de l'uranium.

СТРОИТЕЛЬСТВО ОБОГАТИТЕЛЬНЫХ УСТАНОВОК С ЦЕНТРИФУГАМИ В ЕВРОПЕ.

В докладе кратко рассмотрена история разработки центрифуги в ФРГ, Нидерландах и Соединенном Королевстве, что привело в 1970 году к заключению соглашения о сотрудничестве между тремя указанными странами. Описаны первые строящиеся в настоящее время обогатительные установки, которые будут собственностью трехсторонней международной организации по обогащению урана и будут эксплуатироваться этой организацией. В докладе рассмотрены возможность образования рынка, которым будет располагать трехсторонняя международная организация, возможные меры с целью расширения этого рынка в свете технических и экономических факторов, определяющих применение центрифугирования как метода обогащения урана.

INSTALACIONES DE CENTRIFUGACION EN EUROPA.

Este trabajo revisa brevemente la historia del desarrollo de la centrifugación en la República Federal de Alemania, los Países Bajos y el Reino Unido que condujo al acuerdo de colaboración entre los tres gobiernos en 1970. Se describen las instalaciones iniciales que se están construyendo actualmente y que pertenecerán y serán explotadas por la organización tripartita de enriquecimiento. A la vista de los factores económicos y técnicos que regirán la explotación comercial de la centrifugación como método de enriquecer uranio se revisa el mercado potencial de esta organización y se discute la estrategia más adecuada para la expansión futura.

INTRODUCTION

The concept of separating mixtures of gases of different molecular weights by means of gravitational or centrifugal fields was first tested experimentally by Bredig [1] as early as 1895. The use of the method for separating isotopes was suggested in 1919 by Lindemann and Aston

[2] who developed a separation theory applicable to ideal gases: this theory was subsequently extended by Mulliken [3], Chapman [4] and others. One of the earliest techniques considered was that using the evaporative centrifuge. In this the isotopic mixture is introduced into the rotor in liquid form, the machine is driven up to full speed and the vapour enriched in the light isotope is drawn off along the axis: the separation factor is proportional to the difference in mass of the two isotopic species.

The first successful separation of isotopes by centrifugation was achieved in 1937 following the development by Beams [5] of the convection-free vacuum type machine. Using the evaporative method, Beams and Masket [6] and Beams and Skarstrom [7] altered the concentration ratio of ^{35}Cl and ^{37}Cl in carbon tetrachloride. Humphreys [8] achieved similar success in the separation of ^{79}Br and ^{81}Br in ethyl bromide.

The evaporative centrifuge is essentially a batch process, however, and was soon replaced by the concurrent gas centrifuge in which the mixture to be separated is introduced into one end of a rotating tube and is collected as two separate fractions at the other end. The tubular centrifuge had been pioneered by Beams [9] in the late 1930's and was further developed during the 1940's [10] for the separation of the isotopes of uranium in UF_6.

The concurrent separator is of very limited application because of the severe gas handling problems and the low separation factor which arises only from the separation in the pseudo-gravitational field. The first attempts to multiply the simple process factor in a single centrifuge were made by Skarstrom et al [11] in an evaporative centrifuge and by Harteck et al [12] in an oscillatory machine; but the significant advance in this area was the development of the countercurrent centrifuge by Beams et al [13] in the U.S.A. and by Groth et al [14] in Germany. In the countercurrent centrifuge the gas at the periphery, enriched in the heavy isotope, is swept to one end of the machine whilst that in the centre is swept to the other end. In this way an isotopic concentration difference develops along the length of the machine so that the enriched and depleted streams can be taken off at opposite ends in the high pressure zone, namely the periphery: this considerably eases the gas handling problem. A second advantage is that the simple process factor is increased many times: the separation factor is proportional to the difference in mass of the isotopic species and to the length of the unit, as compared with simply the difference in mass for the evaporative and concurrent systems.

The experimental and theoretical work on the countercurrent method in the U.S.A. followed the initial suggestion of Urey [15]: Groth's work in Germany followed the theoretical work of Martin and Kuhn [16] and of Harteck and Jensen [12]. Cohen [17] published a generalised theory of isotope separation in a centrifuge in 1951. Several methods of creating the desired countercurrent have been investigated, including the use of two separate feeds and external pumping (Beams [10]) and the simpler method of initiating the required flow pattern by establishing a temperature difference along the length of the tube (Groth [14]).

The separative power of the countercurrent machine is proportional theoretically to the peripheral velocity to the fourth power and to the length. As a consequence of this relationship the obvious development aims in any countercurrent centrifuge programme are to achieve high operating speeds and/or machines of long length. The maximum operating speed is determined by the specific strength of the rotor material whereas the length depends on the problems associated with critical resonances in long rotating bodies. If a shaft or tube is rotated about its axis, then, as is well known, flexural resonances will occur at critical rotational speeds determined by the ratio of its length to diameter, the specific modulus and the end masses. A centrifuge operating below the first flexural resonance is termed a sub-critical machine. If the techniques are available for traversing the critical speeds then, for a given material system, a large increase in length, and hence output, can be achieved: such a machine is termed a super-critical centrifuge.

The early research work of Groth, Beams and their co-workers confirmed the general aspects of the process, but the machines developed by these workers were not of a form suitable for exploitation in a practical separation plant. The engineering was sophisticated, the gas handling systems were complex, and power consumption was high, particularly in the bearings. The same

was substantially true of the work carried out in the U.K. at Harwell during the years 1948-1954 by Kronberger, and in the Netherlands in the mid-1950's by Kistemaker [18].

The really significant advance was the description by Steenbeck, Zippe, and Scheffel [19] in 1958/60 of the work carried out by them in USSR during the years 1946-1954. Initially they were concerned with the development of a super-critical centrifuge with a length to diameter ratio of 50:1. This consisted of tubes connected together by flexible bellows (which eases the problem of traversing the critical rotational speeds) and operating at speeds of about 200 to 250 m/s. In 1953, work was started on a sub-critical centrifuge. Zippe and Scheffel returned to the West in the late 1950's and repeated the work on the short tube centrifuge at the University of Virginia in the U.S.A., and at Degussa in Frankfurt/Main, Fed. Rep. Germany, respectively, under a co-operative programme during the years 1958-1960; this work was freely published [19]. It was immediately clear that these types of centrifuge were of simple engineering construction and solved many of the problems of bearing power consumption and gas handling. This led to a resurgence of interest in the centrifuge process, the work in the Netherlands and the U.S.A. was continued and new programmes were undertaken in the U.K. and the Federal Republic of Germany.

In 1960, at the request of the U.S. Government, the three European countries classified their work and consequently have worked completely independently throughout the 1960's. Brief summaries of their work, leading up to Trinational collaboration, follow.

WORK IN THE UNITED KINGDOM

Centrifuge development restarted in the U.K. in late 1958 at Capenhurst, the site of the U.K.A.E.A. diffusion plant. Initially, the work was concentrated on the development of experimental machines for the investigation of rotor dynamics and separation performance. The programme was then expanded to cover the evaluation of the properties of materials and the development of components suitable for prolonged operation at very high speeds. In addition, the development of bearing systems suitable for plant use received considerable attention. Resulting from these programmes a machine design was available in 1964/65 which appeared to be suitable as the basic unit in an economic centrifuge plant. The machine was of relatively simple design and had a high separation performance: the specific power consumption was approximately an order of magnitude lower than for a typical large diffusion plant stage.

The remaining unknowns affecting its potential as an economic separator were the life and cost of the machine. Consequently, life test stands were built in 1965 for investigating the reliability and life of machines and the long term behaviour of components and materials. The first machines were installed in 1966 and are still operating. The results of these and many subsequent tests have clearly indicated that the reliability and life of the machine at design operating conditions is such that the replacement of centrifuges in a plant will not prove to be a decisive factor in the cost of separative work. During this period theoretical and practical evaluation of cascade performance factors was also undertaken.

In parallel with the centrifuge work, the development was also undertaken at Capenhurst of a large diffusion plant stage suitable for use as the feed stage for a plant of output in the range 2,000-5,000 tonnes of Separative Work per year (t.SW/y). The progress and potential of both processes were critically reviewed in 1967. It was concluded that in the European situation – partly because of the high projected cost of power compared with that available to the U.S.A.E.C. – the centrifuge method was likely to be the more economic. A further decisive factor was the view that diffusion technology had been the subject of intensive development in many countries for twenty or more years, and it was considered that further significant improvements were not to be expected. The centrifuge, however, was considered to be still at an early stage of development, giving the potential of low separative work costs through the development of machines of high output and the likely reduction in specific capital costs arising from the study and application of advanced production engineering techniques.

The scale of the project was enlarged as a result of this decision and a mass production version of the laboratory machine was designed. Machines to this design have been built in substantial numbers in a specially constructed workshop and have been subjected to rigorous test

schedules. Detailed design and costing of relatively large-scale plants was also undertaken. On the experimental side, work was continued at an increased scale on improving the separation performance of the machine, further simplifying its design and, in particular, proving plant concepts. An experimental cascade and a prototype plant cascade were built and operated. The results of this work are now to be realised in the plant under construction at Capenhurst.

In parallel with this work, the construction and evaluation of machines and plant systems of advanced concept has also been undertaken; it is estimated that machine and plant designs based on these concepts will be available for commercial exploitation in the late 1970's.

WORK IN THE FEDERAL REPUBLIC OF GERMANY

In 1950, Beyerle, Groth, Martin, and their co-workers recommenced their work – which had been interrupted by World War II – at Aachen, Bonn, Göttingen, and Kiel. Many important experimental and theoretical results were achieved [14, 20, 21, 22, 23]. The U.S./German co-operative programme already referred to was established in 1958 but was terminated due to classification of all centrifuge work in 1960.

In 1964, a new Government-owned research company "Gesellschaft für Kernverfahrenstechnik mbH" (GKT), founded at Jülich, took over and continued the Germany centrifuge programme. GKT demonstrated the feasibility of centrifuge cascades for uranium isotope separation up to a stage where industrial exploitation seemed to be possible.

In 1967, several private German companies (DORNIER, Friedrichshafen, INTERATOM, Bensberg, ERNO, Bremen and MAN, München) started work in the centrifuge field. An experimental cascade was designed in 1968 and came into operation at the beginning of 1970. Useful information has been obtained on cascade behaviour and also on hex technology and handling systems.

With the prospect of collaboration in mind, two private companies were formed in 1969; "Gesellschaft für nukleare Verfahrenstechnik mbH" (GnV) located at Bensberg, and "URANIT, Uran-Isotopentrennungs-GmbH" at Jülich. These companies took over all West German centrifuge work, including the laboratories and personnel of GKT. GnV hold one-third of the shares of the Prime Contractor and URANIT one-third in the Enrichment Organisation (URENCO).

WORK IN THE NETHERLANDS

The initial fundamental research in the Netherlands was carried out by Kistemaker who, already aware of the experiments made by Beams in the U.S.A., proposed a programme of work after attending a colloquium on centrifuges organised by the "Physikalisches Institut" at Hamburg in 1954. This work was promoted by the Netherlands Institute for Fundamental Research of Matter (F.O.M.) and was supported financially by the Netherlands Reactor Centre (R.C.N.).

Kistemaker's experiments had been hindered by the same obstacles which led Beams to abandon his research in the field but the information made available by Zippe et al [19] – particularly regarding the upper and lower bearing systems – was of material assistance. The research work continued with renewed enthusiasm and, in 1961, the centrifuge project entered a new technological phase. For this reason its further development was entrusted to R.C.N. and a special centrifuge development laboratory was installed for the purpose at Amsterdam. Particular attention was necessary in this phase of the work to problems of rotor dynamics: these were resolved quite satisfactorily.

The status of the project in 1967 required that the laboratory scale experiments be supplemented by further development work in two stages:

(a) testing of a large number of centrifuges in cascade formation and study of cascade behaviour,

(b) design of a complete pilot plant.

The first multi-machine experimental cascade became operative in the Autumn of 1968 and proved to be most successful: useful practical experience on cascade behaviour was accumulated. Machine and plant development studies have continued in parallel with this cascade work. Also

in 1968, the engineering company COMPRIMO Limited was commissioned by R.C.N. to design a pilot plant with a capacity of up to 25 t.SW/y. A complete technical design was submitted together with detailed plans and a cost estimate. All the available performance and design data were evaluated, in consultation with interested industrial parties in the Netherlands, and resulted in the formation in 1969 of the Ultra-Centrifuge Nederland N.V. (U.C.N.) in which the Netherlands Government, Dutch State Mines, Philips, Shell, Rijn-Schelde and Verenigde Machine Fabrieken are participants. The first object of this company is the construction of a semi-commercial plant with a capacity of 25 t.SW/y on a site near Almelo.

TRIPARTITE ORGANISATIONS

It has previously been stated that, because of the agreement on military classification, work in the three countries proceeded independently. In 1968, public statements, particularly by F.R. Germany and the Netherlands, indicated the possibility that these countries had reached a similar stage of development and confidence in the process as had the U.K. Hence, with a growing belief that international co-operation was necessary to achieve a fully competitive enrichment plant in Europe, approaches were made at Government level aimed at achieving a joint venture. In early 1969 a limited technical exchange and a comparison of machine costs and plant economics provided the added confidence necessary to proceed with negotiations. These culminated in the signature at Almelo on 4th March 1970 of a Treaty of Collaboration between the three countries [24]. The Agreement provides for the formation of two companies – The Prime Contractor and The Enrichment Organisation. The Prime Contractor, sited in Germany, is concerned with the development, design and manufacture of centrifuges and plants, and with the specific object of achieving a common development programme within 18 months of formation. The other company, now called the Uranium Enrichment Company (URENCO) will purchase plants from the Prime Contractor, operate these through subsidiaries and market the enriched uranium.

The treaty has been ratified by all three countries and the two companies are now fully engaged in the evaluation of the technical position, with the objective of achieving a joint development programme and a common design of centrifuge and plant for large scale exploitation. The Shareholders of the Enrichment Organisation (URENCO) are :

"The British Nuclear Fuels Limited" (B.N.F.L.)
"Ultra-Centrifuge Nederland N.V." (U.C.N.) and
"URANIT, Uran-Isotopentrennungs-GmbH"

INITIAL PLANTS

The Agreement covers the construction of initial plants to a total capacity of approximately 350 t.SW/y, sited at Capenhurst and at Almelo. Currently, a total capacity of approximately 90 t.SW/y is under construction at the two sites under the control of the individual countries. The remaining capacity of about 250 t.SW/y will be built jointly as a pre-runner to large commercial plants. The plants now under construction are principally regarded as demonstration plants aimed at obtaining detailed information on centrifuge mass production procedures and gaining plant operating experience on a significant scale.

The plant at Almelo is being constructed in two separate parts. The first is of Dutch design and is being built under the supervision of U.C.N. The design output is 25 t.SW/y, the product concentration can be varied between approximately 2·5 and 4% ^{235}U and the waste concentration will be 0·2%. Plant construction started in November 1969 and is now at an advanced stage. The centrifuges for the plant have been produced in a separate factory, on the same site, owned and controlled by U.C.N. but operated by a sub-contractor. The completed plant is scheduled to become operational in the early months of 1972.

The second part of the Almelo plant is of German design and will be erected by GnV, under contract to URANIT, in two stages. The first stage comprises a T-shaped supply building containing the complete UF_6 facilities, electrical supply and control system and a cascade hall with about

one-third of the centrifuges. The second stage will contain the residual two-thirds of the centrifuges and will bring the plant up to the total design capacity of 25 t.SW/y. This division into two stages gives the opportunity to install different designs of centrifuges and cascade, and thus make use of the advancement in technology during the construction period. The plant is designed to have a product concentration adjustable over the range 2·2 to 3·2% ^{235}U, and a waste concentration of 0·2%. It is anticipated that it will be progressively commissioned in stages between March 1973 and August 1974.

Initially the plant at Capenhurst will have a separative work capacity of 15 t.SW/y at a product concentration of 2·5% ^{235}U and a waste concentration of 0·3% ^{235}U. It is designed for extension to about 50 t.SW/y and is located in an existing building on the Capenhurst site. The primary objective in the initial plant is to confirm the technical and economic predictions; it is also designed in such a way that any remaining problems can be defined and resolved quickly. The plant will be completely commissioned by January 1973.

THE OBJECTIVES OF URENCO

The main objectives of URENCO will be to produce enriched uranium at a commercially viable cost and to sell its enrichment services as widely as possible. The scale of URENCO's activities will clearly be influenced by several factors. Among these, the most important are:

(a) the rate of technological advance resulting from the pooling of the information from the three countries.

(b) the timing of implementation of the significant improvements in technology and economics expected to arise under item (a).

(c) the availability of capital: this will largely be determined by the potential earning power of the capital investment. For a relatively modest target of, say, a final plant capacity of 10,000 t.SW/y a total investment in the region of £500M is envisaged.

With regard to the rate of capital investment, it should be pointed out that the centrifuge process has a major advantage over the diffusion process. It is relatively easy to match the rate of construction of a centrifuge plant – and hence the rate of capital investment – to the growth of the market. Consequently, if delays occur on the market, the rate of investment in the enrichment plant can readily be adjusted. For example, a plant of 10,000 t.SW/y can be built progressively over a period of, say, ten years, giving an annual investment of £50M without materially affecting the plant efficiency and hence the cost of separative work. The position with a diffusion plant is radically different: the throughput for a large plant can only be economically achieved by the use of large individual stages and sensible plant efficiencies can only be achieved when the majority of the stages are available for connection in series. Hence the major part of the capital expenditure must be invested in anticipation of the market developing: and a fair return on that capital is achieved only when the market has developed.

THE FUTURE MARKET FOR ENRICHMENT

The projected market for enriched uranium in the period 1975/1985 has been the subject of many detailed and complex studies. The uncertainties involved in these projections, which are discussed in the following paragraphs, are such that strict mathematical treatment produces an extremely wide range of requirements. The range is such that it does not alone provide an adequately firm basis for planning.

The position was admirably put by U.S.A.E.C. Commissioner W. E. Johnson [25] in his address to the 1970 Annual Conference of the Atomic Industrial Forum, when he stated "Our computers reach a point in this kind of exercise where human judgement is more useful in getting an answer than further computerised problem elaboration".

Estimates [26] of the nuclear power capacity to be installed throughout the world, excluding the U.S.A. the U.S.S.R. Eastern Europe and the People's Republic of China, are of the order of 140,000 MW(e) by 1980, rising to about 300,000 MW(e) by 1985. Well over 80% of this capacity will require enriched uranium. It must be stressed that the assumptions on which these figures

TABLE I

SEPARATIVE WORK REQUIREMENTS

Year	Approximate Annual Requirements (t.SW/y)	
	World[1]	World[1] excluding U.S.A.
1975	18,000	7,000
1980	38,000	17,000
1982	50,000	24,000
1985	70,000	35,000

are based could well prove to be optimistic and the likelihood must exist that the installed capacity will turn out to be considerably less than the figures quoted. In converting this type of projection into requirements for separative work, it is necessary to make assumptions on:

(a) the type of reactors, the rate of advancement of core design, improvement in burn-up values and neutron economy;

(b) the recycling of reprocessed uranium and plutonium and, in particular, the year in which this is likely to become a practical possibility;

(c) the change in reactor load factors with the increase in installation of nuclear power stations;

(d) the timing and rate of installation of breeder reactors (it is now generally assumed that breeder reactors will not significantly change the requirements for separative capacity in the next 10-20 years).

As an example of the effect of these assumptions, it can be shown that for a mixed system of Boiling Water and Pressurised Water reactors, and using the manufacturers' and utilities' predictions of improvements, the separative work requirements per megawatt of installed electrical capacity reduce by 40 per cent between 1970 and 1980. Further uncertainties are introduced by the uranium market and the enrichment process itself, for example:

(a) an increase in the price of uranium feed will increase the separative work requirements because, in this event, it will be economic to reduce the ^{235}U concentration in the waste flow ("tails assay" in U.S. terminology) from the enrichment plant;

(b) an increase in separative work costs will have the converse effect of reducing the separative work requirements: in this event it becomes economic to increase the ^{235}U concentration in the waste flow;

(c) the potential surplus capacity of U.S. diffusion plants arising from the possibility of the U.S.A.E.C. regulating their plant outputs by raising the waste concentration.

Using assumptions similar to those of Johnson (namely, plutonium recycle starting in 1974 and waste concentrations of 0·25 per cent) but including the U.K. market, the World[1] requirements can be approximately represented by the values given in Table 1.

[1] "World" in this report means all countries except U.S.S.R., Eastern Europe and People's Republic of China.

It is obvious that the U.S. will plan to provide its domestic requirements from internal sources. It is further being assumed in the U.S. that the U.S.A.E.C. or other future owners of the enrichment plants, will capture about 75 per cent of the non-U.S. market in 1980 and 65 per cent in 1985 – excluding the U.K. market. Whether these assumptions prove to be valid will depend on the relative costs of separative work from new installations in the U.S., in Europe, and possibly elsewhere. It is instructive, nevertheless, to examine the market which would be available to the Tripartite Companies in the event that the U.S. projection were regarded as correct. The separative work demand, external to the U.S. and excluding the Capenhurst diffusion plant, would be approximately 5,000 t.SW/y in 1980 and 13,000 t.SW/y in 1985.

The magnitude, even of this "minimum" market, indicates that major decisions and financial commitments for centrifuge plants will be required in the next two or three years.

It has frequently been suggested that the rate of production of centrifuges, even for the relatively small plants required to meet the demands just quoted, will be difficult to achieve. We do not believe this to be so. As an example, to meet an installation programme of 1,500 t.S.W. each year in the early 1980's would require the production of about ¼ - ¾ million centrifuges (depending on type) each year, or 5,000 - 15,000 machines per week. Although each machine contains sophisticated engineering components, the total number of such components is not large, and these production rates are certainly quite practicable, especially when judged against the fact that production facilities in all three countries will be available.

A number of other factors will clearly influence the scale and timing of the building of large enrichment plants by the Tripartite Companies. The more important of these, which are largely outside the control of the Companies, are likely to be:

(a) the rate of escalation of the U.S. price-scale particularly as a direct result of increases in power costs;

(b) the timing and predicted costs of new additions to the U.S. enrichment plants;

(c) the general contractual arrangements which will be found acceptable to the purchasers of toll enrichment and, in particular, the terms of cancellation, because this will affect the ease or otherwise with which capital for plant construction can be obtained;

(d) the possible entry of the U.S.S.R., France, Japan, or South Africa into the enrichment market.

The final factor is clearly the rate of advance of centrifuge technology within the next few years; the achievement of the full development potential would place the Companies in a strong position to attack the market worldwide.

REFERENCES

[1] BREDIG, A., The influence of centrifugal force on chemical systems, Z. phys. Chem. 17 (1895) 459.

[2] LINDEMANN, F. A., ASTON, F. W., The possibility of separating isotopes, Phil. Mag. 37 6 (1919) 523.

[3] MULLIKEN, R. S., The separation of isotopes by thermal and pressure diffusion, J. Am. chem. Soc. 44 (1922) 1033
and The separation of liquid mixtures by centrifuging, J. Am. chem. Soc. 44 (1922) 1729
and The separation of isotopes: application of systematic fractionation to mercury in a high speed evaporation - diffusion apparatus, J. Am. chem. Soc. 45 (1923) 1592.

[4] CHAPMAN, S., The possibility of separating isotopes, Phil. Mag. 38 6 (1919) 182.

[5] BEAMS, J. W., PICKELS, E. G., The production of high rotational speeds, Rev. Sci. Instr. 6 (1935) 299.

[6] BEAMS, J. W., MASKET, A. V., The concentration of chlorine isotopes by centrifuging Phys. Rev. 51 (1937) 384.

[7] BEAMS, J. W., SKARSTROM, C., The concentration of isotopes by the evaporative centrifuge method, Phys. Rev. 56 (1939) 226.

[8] HUMPHREYS, R. F., Separation of Br. isotopes by centrifugation, Phys. Rev. 56 (1939) 684.
[9] BEAMS, J. W., High rotational speeds, J. appl. Phys. 8 (1937) 795 and High speed centrifuging, Rev. mod. Phys. 10 (1938) 245.
[10] BEAMS, J. W., SNODDY, L. B., KUHLTHAU, A. R., Tests of the theory of isotope separation by centrifuging, Int. Conf. peaceful Uses atom. Energy (Proc. Conf. Geneva 1958) 4 (1958) 428.
[11] SKARSTROM, C., CARR, H. E., BEAMS, J. W., Concentration of the chlorine isotopes by centrifuging, Phys. Rev. 55 (1939) 591.
[12] HARTECK., P., JENSEN, H., Calculations of the separation effect and the yield of various arrangements of centrifuges in order to improve the efficiency of a single centrifuge, Unpublished Report, 1942.
[13] BEAMS, J. W., SNODDY, L. B., The separation of the isotopes of uranium in UF_6 by the refluxing counter-flow centrifuge method, American report 3391 (1943). Declassified 1960.
[14] GROTH, W., NANN, E., WELGE, K. H., Enrichment of uranium isotopes in a counter-current gas centrifuge, Z. Naturf. 12A (1957) 81.
[15] UREY, H. C., Rep. Prog. Phys. VI (1939) 72.
[16] MARTIN, H., KUHN, W., Multiplikationsverfahren zur Trennung von Gasgemischen, insbesondere bei Anwendung von Schwerefeldern, Z. phys. Chem. A. 189 (1941) 219.
[17] COHEN, K., The Theory of Isotope Separation as applied to the Large Scale Production of U^{235}, McGraw-Hill (1951) 103.
[18] KISTEMAKER, J., LOS, J., VELDHUYZEN, E. J. J., The enrichment of uranium isotopes with ultracentrifuges, Int. Conf. peaceful Uses atom. Energy (Proc. Conf. Geneva, 1958) 4 (1958) 435.
[19] ZIPPE, G., SCHEFFEL, R., STEENBECK, M., Schnellaufende Gaszentrifuge, German Patent DBP 1 071 593 (1957).
ZIPPE, G., STEENBECK, M., Schleudertrommel für schnellaufende Zentrifugen, German Patent DBP 1 120 986 (1957).
ZIPPE, G., The development of short bowl ultracentrifuges: final report USAEC Report ORO.315 (1960).
[20] BEYERLE, K., GROTH, W., IHLE, H., MURRENHOFF, A., NANN, E., WELGE, K . H., Enrichment of uranium isotopes with the gas centrifuge. (Proc. Int. Symp. on Isotope Separation, Amsterdam 1957) North Holland Publishing Co. (1958) 667.
[21] GROTH, W., BEYERLE, K., NANN, E., WELGE, K. H., Enrichment of the uranium isotopes by the gas centrifuge method, Int. Conf. peaceful Uses atom. Energy (Proc. Conf. Geneva 1958) 4 (1958) 439.
[22] MARTIN, H., Gas convection in ultracentrifuges and its importance for the development of gas centrifuges, Chemie-Ingr. Tech. 31 (1959) 73.
[23] BULANG, W., GROTH, W., NANN, E., Thermally controlled countercurrent centrifuges with axial and radial temperature gradients, Z. Physik. Chem. 25 (1960) 283.
[24] U.K. Edition, Agreement between the United Kingdom of Great Britain and Northern Ireland, the Federal Republic of Germany and the Kingdom of the Netherlands, on Collaboration in the development and exploitation of the Gas Centrifuge process for producing Enriched Uranium, Almelo 4th March 1970, Miscellaneous No. 5 (1970) Cmnd. 4315. H.M. Stationery Office.
[25] JOHNSON, W. E., Report of Conference, Nuclear Industry 17 11/12 (1970) 15.
[26] ENEA/IAEA, Joint Report, Uranium: Resources, Production and Demand, September 1970.

OBJECTIVES AND PROGRESS IN THE CENTRIFUGE ENRICHMENT PLANT INDUSTRY

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Abstract–Résumé–Аннотация–Resumen

OBJECTIVES AND PROGRESS IN THE CENTRIFUGE ENRICHMENT PLANT INDUSTRY.

The international Prime Contractor company was formed in the Federal Republic of Germany in order to undertake on a commercial basis the development, design and construction of gas centrifuge plants for the enrichment of uranium. The company will initially co-ordinate and later take control of the research and development programs in various laboratories and other centres in the United Kingdom, the Netherlands and the Federal Republic of Germany. The activity leads to certain organizational problems which are briefly considered. The paper also examines the possible avenues of centrifuge and enrichment plant development and points to economic, production and business restraints on development. Some early conclusions including the economics of enrichment plant production are drawn and discussed.

OBJECTIFS DES INSTALLATIONS D'ENRICHISSEMENT PAR CENTRIFUGATION ET PROGRES REALISES.

Une société internationale a été créée en République fédérale d'Allemagne afin d'entreprendre, sur une base commerciale, l'étude et la construction d'installations de centrifugation gazeuse pour l'enrichissement de l'uranium. Cette société assurera d'abord la coordination puis la direction des programmes d'étude et de réalisation de différents laboratoires et autres centres de la République fédérale d'Allemagne, du Royaume-Uni et des Pays-Bas. Cette activité soulève certains problèmes d'organisation que le mémoire expose brièvement. Il étudie aussi les possibilités offertes par la mise au point d'installations d'enrichissement par centrifugation et indique les facteurs d'ordre économique, commercial et de production qui y font obstacle. Il établit et analyse des conclusions préliminaires notamment sur l'économie de la production de ces installations.

ЗАДАЧИ И ДОСТИЖЕНИЯ НА ПРЕДПРИЯТИЯХ ПО ОБОГАЩЕНИЮ УРАНА С ПОМОЩЬЮ ЦЕНТРИФУГ.

В Федеративной Республике Германии создана международная компания – "Главный подрядчик", призванная осуществлять на коммерческой основе разработку, проектирование и строительство заводов по обогащению урана с помощью газовых центрифуг. Компания будет первоначально координировать, а затем осуществлять контроль за выполнением программ в области научных исследований и разработок в различных лабораториях и других ядерных центрах Соединенного Королевства, Нидерландов и Федеративной Республики Германии. Эта деятельность вызывает некоторые организационные проблемы, которые кратко рассматриваются в докладе. В нем рассматриваются также возможные направления развития заводов газовых центрифуг для обогащения урана и указываются некоторые экономические, производственные и практические факторы, сдерживающие это развитие. Приводятся и обсуждаются некоторые предварительные выводы, включая экономические аспекты производства обогащенного урана на заводах по его обогащению.

OBJETIVOS Y PROGRESOS EN LA INDUSTRIA DE INSTALACIONES DE ENRIQUECIMIENTO POR CENTRIFUGACION.

Se ha constituido en la República Federal de Alemania, con carácter de contratista principal, une sociedad internacional encargada de emprender, sobre una base comercial, el desarrollo, diseño y construcción de instalaciones

centrífugas de gas para el enriquecimiento del uranio. La empresa coordinará inicialmente y más adelante controlará los programas de investigación y desarrollo de diversos laboratorios y otros centros del Reino Unido, los Países Bajos y la República Federal de Alemania. Se consideran brevemente ciertos problemas de organización que se presentan en este tipo de actividades. Se analizan también en esta memoria las posibles vías de desarrollo de las instalaciones centrífugas de enriquecimiento, y se señalan sus limitaciones de carácter económico, de fabricación y comercial. Se exponen y debaten algunas de las primeras conclusiones, entre las que se incluye la economía de las instalaciones de enriquecimiento.

1. INTRODUCTION

Development carried out independently within a number of countries over several years has indicated the possibility that the gas centrifuge could provide a commercially attractive method of the production of enriched uranium.

Consequently an agreement on collaboration in the development and exploitation of the gas centrifuge process for producing enriched uranium was signed at Almelo in the Netherlands on 4th March, 1970, by Ministers of the Netherlands, German and British Governments [1]. Essentially, the three Governments undertook to collaborate with a view to the enrichment of uranium by this process.

The commercial exploitation of the gas centrifuge process under this agreement involves

1. the pooling of all the gas centrifuge information so far generated in the three countries;
2. the formulation and putting into operation of an integrated research and development programme;
3. the establishment of a first joint centrifuge enrichment plant design;
4. the construction and operation of process plants;
5. the exploitation of the pooled industrial rights.

The Tripartite Centrifuge Agreement represents a more farreaching and ambitious venture than any so far attempted in the nuclear field. This is no mere agreement to exchange research information or technology, nor is it limited to the execution of one specific development project or programme. By their agreement, the three Governments promoted the formation of two commercial companies which, under the supervision of a Joint Committee of Government representatives, will exercise commercial freedom and act according to commercial standards of profitability. The agreement made between the three Governments involves also three nominated shareholders in each of the two companies. It involves also specific commitments to investment, and carefully worked out provisions concerning industrial rights.

The agreement relates to a technology which is still classified. It is necessary therefore to follow strict security precautions, and also to ensure that all operations carried out under the agreement are in full compliance with the Non-Proliferation Treaty.

After the establishment of the Joint Enterprises it may be possible to bring in companies from other countries with similar interests. A prerequisite would be the conclusion of an agreement between the Governments concerned.

2. ORGANISATION

The Agreement provides that the tasks referred to above should be undertaken by "Joint Industrial Enterprises". In fact, two such enterprises have been established, an "Enrichment Organisation" and a "Prime Contractor". As this paper is concerned with the organisation and role of the Prime Contractor, it will suffice to say of the Enrichment Organisation that its role is to own and operate enrichment plants, to sell enrichment services, and to exercise responsibility for research and development on centrifuge plant operation.

The Prime Contractor is a company established under German law, with its headquarters at Bensberg near Cologne in Germany. The capital is held in equal shares by British Nuclear Fuels Limited, the Gesellschaft für nukleare Verfahrenstechnik m.b.H., and the Ultra-Centrifuge Nederland N.V.

The objective of the Prime Contractor is to design, develop, and construct economic plants which will be owned and operated by the Enrichment Organisation or eventually by operators in other countries. In setting about this task it will be necessary to implement the co-ordinated development programme mentioned in the Introduction after initially assessing the work already undertaken in the three countries. The Agreement requires that the existing development and manufacturing facilities should be utilised as far as possible.

In the early years therefore the majority of tasks of the P.C. will be sub-contracted to organisations in the three countries already working in this field. Whether it will at a later stage find necessary to acquire its own laboratories or manufacturing facilities to supplement national facilities will depend on the succes of the centrifuge process and on commercial consideration.

3. ECONOMIC TARGET

At the present time there is an excess capacity of uranium enrichment facilities in the western world, due primarily to the giant U.S. diffusion plant complex. This situation, however, is quickly changing with the rapid expansion in the installation of nuclear power stations. Approximately each 1,000 MWe nuclear power station using enriched uranium requires to be supported by a 100 t/year enrichment capacity; thus a projected nuclear Installation programme for western Europe of 85,000 MWe by 1980 implies an enrichment plant requirement of 8,500 t/year. Even if only part of this requirement is satisfied within Europe, this implies an installation rate of over 1,000 t/year of enrichment plant in the late 1970's.

The world price for separative work is primarily determined by the published U.S. price scale using product from the American diffusion plant complex. At the present time the price quoted is about $ 32/kg f.o.b.[1] in the U.S., which allowing for transportation costs, would be equivalent to about $ 36/kg f.o.b. at a factory in Europe. The present U.S. plant capacity is 17,000 t/year.

In order to meet the increasing demand of enriched uranium some enlargement of the present capacity is under discussion. Such enlargement could be achieved by modification of present plants or building new plants. The cost of enrichment from U.S. diffusion plant is difficult to predict because it is sensitive to both scale of operation and cost of electrical power. Nevertheless, a realistic target for European separative work (S. W.) costs from a European plant will probably be of the order of $ 34/kg.

[1] f.o.b. = free on board.

4. DEVELOPMENT AND DESIGN

Work already undertaken confirms that the specific power consumption (power consumption per separative work output) of the centrifuge process is much lower than of the diffusion process. Therefore the economics of the centrifuge process and the corresponding costs of separative work depend to a large extent on the specific investment costs (costs of plant and centrifuges per separative work output) and the lifetime of the components.

As the separative work output of a single centrifuge is of the order of a few kilograms large numbers of centrifuges have to be manufactured and assembled in order to meet the predicted future separative work demands. Typically this means the production of several hundred thousands of centrifuges per year, which allows the application of mass production techniques to substantially lower the unit costs. The overall separative work costs may be further reduced by an increased separative work output from each centrifuge, provided this can be achieved without increasing these machine costs.

Increased output can be obtained by increasing the circumferential speed of the rotor and/or by increasing the separation length. The achievement of higher speeds presents problems because of the higher stresses in the rotating components. One of the properties required from the materials is a high specific strength (i.e. high ratio of tensile strength to density).

Possible materials of interest include:

- Aluminium alloys
- High strength steels
- Titanium
- High strength fibre composites.

Another problem associated with the selection of materials is compatibility in the process environment which includes uranium hexafluoride. The results of the programmes already in progress in the member laboratories where these materials problems are being studied, and other relevant technologies such as bearing design, will be available for the Prime Contractor. The design of a single, optimum machine can then be established taking into account data from these programmes together with the available centrifuge operating experience which will enable life predictions to be made.

The centrifuge machine, although perhaps the most critical, is only one of the components making up the process plant. The centrifuges must be interconnected into cascades to give the required enrichment and output and special plant components, such as valves, have been developed and tested. Similarly, electrical drive systems have been designed and proven. In the three countries the design, construction, and operation of several experimental cascades have demonstrated the feasibility of the design criteria and the control and stability of cascades. Furthermore, the continued operation of these cascades contributes significantly to the life testing of machines and plant components.

As mentioned in Section 1, the Prime Contractor will initially examine and co-ordinate the national development programmes of the member organisations. This will involve a progressive rationalisation of the three programmes with optimisation of the effort and the use of development facilities, leading to an increased pace of development coupled

with cost savings in each of the member organisations. At the same time, the P.C. will be determining the nature and extent of a combined development programme to meet jointly agreed commercial objectives. This programme, which should be instituted within eighteen months of the setting up of the P.C., will be both financed and controlled by the new company.

The selection of this joint development programme amounts to more than assembling the best features from the three national programmes. It involves initially the definition and timing of the principal commercial objectives and then the planning of the development programme to achieve these objectives, taking into account the extent to which the existing national programmes can be matched to the joint requirement. It is not only technical merit which will be important but also suitability for large-scale application, and the inherent potentiality for longer term development. The development programme could include a number of promising alternatives, but since the enriched uranium market will be limited by the commercial introduction of fast reactors, it is important to concentrate on designs which offer clear promise on an appropriate time scale. The earlier a design can be introduced the better the prospects of maximising the return on the design, development, and capital expenditure.

5. CONSTRUCTION

The Tripartite Agreement [1] provides the construction of 350 t S.W. during an initial programme phase. The construction of the first prototype enrichment plants (90 t S.W.) has already begun at Almelo and Capenhurst under the responsibility of the national organisations. It is planned that the Enrichment Organisation will assume responsibility for operation of these plants at a later date.

5.1. Progress at Capenhurst

A prototype enrichment plant is being built at Capenhurst by British Nuclear Fuels Limited. This is to have a nominal output of 40 t separative work per year, with construction being handled in two distinct phases separated by about a year in time. The building of the first phase of the plant commenced earlier this year and is programmed for completion in 1972. It will be situated in part of the existing building containing the main diffusion plant and will have its own process services. These, together with control facilities and electrical supplies, have been laid out to serve both phases of the plant. The Workshop for the assembly and inspection of centrifuge machines is situated in the same building. The construction of the Workshop is timed to supply machines for use in the prototype enrichment plant at a rate consistent with running the first cascade in 1972.

The building of the plant and workshops by phases has the important advantage of advancing construction and operating experience whilst permitting design improvements under proving tests to be incorporated into the later stages of the plant design. It is also suited to a collaborative project in that some flexibility is available until a fairly late stage.

5.2. Progress at Almelo

A prototype enrichment plant with a nominal output of 25 t separative work per year is being built at Almelo by Ultra-Centrifuge Nederland N.V. The building of this plant started in November 1969 and is programmed for

completion in 1971. The centrifuges for this plant are presently being manufactured in a separate adjacent workshop, the construction of which was started in the second half of 1969 and was completed in May 1970.

At the same site a German prototype plant with a nominal capacity of 25 t is being built by Gesellschaft für nukleare Verfahrenstechnik m.b.H. as prime contractor for Uranit. The construction of the plant is divided in two phases. The first phase comprises construction of the process service building for the required capacity and the first cascades. In the second phase the construction of an advanced design will follow. The construction of the first phase starts Summer 1971 and will be completed at the end of 1972.

The centrifuges for the plant are being manufactured in Germany and will be transported to the Almelo site, where they are assembled into modules and checked out for insertion into the cascades.

6. FUTURE PROSPECTS

The future prospect for centrifuge enrichment plants depends primarily on their competitiveness in relation to other sources of supply which will determine their share of the market for enriched uranium. It is necessary to consider whether, within this constraint, centrifuge plants could offer an adequate return on investment.

It appears that on the basis of present technology and using available manufacturing capacity in the three countries the specific investment for a 300 t plant in operation from about 1976 would be in the order of $ 150-200/kg S.W. Such a plant would however be smaller than the minimum economic size. In the later 70's it is expected that the specific investment could be reduced to about $ 120/kg S.W. with the introduction of mass production techniques and improved centrifuge and plant designs. Taking into account this low-specific investment and the known low specific energy consumption (approximately 300kWh/kg S.W.) it is clear that the centrifuge process provides an attractive method for the enrichment of uranium.

REFERENCE

[1] Agreement on Collaboration in the Development and Exploitation of the Gas Centrifuge Process for producing Enriched Uranium. Almelo, 4th March 1970.

LES ETUDES DE PROCEDES DE FABRICATION ET LES USINES D'EAU LOURDE SUIVANT LE PROCEDE FRANÇAIS

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Abstract–Résumé–Аннотация–Resumen

RESEARCH ON HEAVY-WATER PLANTS AND MANUFACTURING PROCESSES BASED ON THE FRENCH SYSTEM.

Over the last seven years the research conducted by the Commissariat à l'énergie atomique on the production of heavy water has concerned water/hydrogen sulphide isotopic exchange, with particular emphasis on the ammonia/hydrogen exchange process (in conjunction with several industrial companies); there has also been a study of other versions of the process and ways of improving production capacity. The water/hydrogen sulphide exchange process, a topic of research at an earlier stage, has recently been revived in the light of enlarged production capacity. At the present time the basic elements are available for a plant producing 400 t/yr, suitable for the needs of a heavy-water reactor system, if constructed. Ammonia hydrogen exchange, first applied in practice at the specially constructed plant at Mazingarbe (France) has been investigated in great detail both in the laboratory as well as on a semi-industrial level, and also on the basis of optimization studies. The information gained from the thoroughly satisfactory operation of the plant and pilot projects conducted in parallel have resulted in improvement in the technological and economic aspects of the monothermal version; data have also been made available on which to base a bithermal version when local conditions permit. A second monothermal plant producing 67 t/yr, in process of construction at Baroda in India, is superior in several respects to the Mazingarbe plant. The construction of additional plants is at present under discussion by the parties concerned. Other research under way, with a longer-range objective in view, concerns hydrogen/amine exchange, which appears to offer attractive prospects. To ensure that the process is self-contained, the possibility of re-enrichment by hydrogen/water exchange through homogeneous catalysis and the chemical conversion of feedwater into hydrogen is under consideration.

LES ETUDES DE PROCEDES DE FABRICATION ET LES USINES D'EAU LOURDE SUIVANT LE PROCEDE FRANÇAIS.

Au cours des sept dernières années, les études effectuées par le Commissariat à l'énergie atomique en vue de la production d'eau lourde ont porté sur le procédé d'échange isotopique eau-hydrogène sulfuré, mais surtout sur le procédé ammoniac-hydrogène en association avec plusieurs firmes industrielles, sur les variantes de ce procédé et sur les moyens permettant d'en accroître la capacité de production. Le procédé d'échange eau-hydrogène sulfuré, qui avait fait l'objet d'études antérieures, a été réexaminé récemment dans l'optique de productions plus importantes. On dispose aujourd'hui des éléments d'un projet d'usine de 400 t/an, répondant aux besoins d'une filière éventuelle de centrales à eau lourde. Le procédé d'échange ammoniac-hydrogène, concrétisé par la construction d'une première usine à Mazingarbe (France), a fait l'objet d'études très détaillées, à la fois au laboratoire, à l'échelle semi-industrielle et sur le plan des calculs d'optimisation. Les informations tirées de l'exploitation particulièrement satisfaisante de cette usine et les études à l'échelle pilote poursuivies

simultanément ont conduit à améliorer la version monotherme sur les plans technique et économique; elles ont aussi fourni les données permettant d'envisager la formule bitherme lorsque les conditions locales s'y prêtent. Une seconde usine monothermie de 67 t/an, en cours de construction à Baroda (Inde), présente par rapport à celle de Mazingarbe plusieurs améliorations. La construction d'autres usines est en cours de négociation avec divers partenaires. D'autres recherches sont en cours avec un objectif plus lointain; elles portent sur le procédé d'échange hydrogène-amines, qui semble ouvrir des perspectives intéressantes. Pour assurer l'autonomie du procédé, on examine les possibilités du « regonflage » par échange hydrogène-eau réalisé par catalyse homogène, et de la conversion chimique en hydrogène de l'eau d'alimentation.

РАЗРАБОТКА НОВЫХ МЕТОДОВ ПРОИЗВОДСТВА ТЯЖЕЛОЙ ВОДЫ ВО ФРАНЦИИ.

На протяжении семи последних лет Комиссариат по атомной энергии занимался исследованием методов производства тяжелой воды, основанных на реакциях химического обмена: вода-сероводород и, главным образом, водород-аммиак. Последний метод был разработан совместно с промышленными фирмами. Исследованы также способы увеличения мощности заводов по производству тяжелой воды. Процесс вода-сероводород, который изучался ранее, недавно был вновь исследован с целью применения его на установках высокой производительности. В настоящее время Франция располагает достаточным запасом исходных продуктов для сооружения заводов мощностью 400 т в год, которые могут потребоваться в случае строительства крупных тяжеловодных реакторов. Обменный процесс аммиак-водород, который применяется на первом французском заводе в Мазенгарбе, был детально исследован в лабораторных условиях и на полупромышленных установках; выполнены также экономические оценки. Успешная работа завода, а также одновременные исследования этого процесса на полупромышленных установках привели к усовершенствованию технологии и повышению экономичности этого процесса по сравнению с простым циклом. Эти исследования обеспечили также получение данных, которые можно использовать для рассмотрения процесса с двойным циклом.

Второй завод (с простым циклом, производительность – 67 т в год), строящийся в Барода (Индия), имеет ряд преимуществ, связанных с введением некоторых усовершенствований по сравнению с заводом в Мазенгарбе. Обсуждается вопрос строительства других заводов по производству тяжелой воды. В настоящее время продолжаются дальнейшие исследования, имеющие целью разработать перспективные методы производства тяжелой воды. В первую очередь исследуется обменный процесс водород-амин, который представляется весьма перспективным. Для обеспечения автономии этого процесса исследуется возможность использования метода "нагнетания газа" путем обменной реакции вода-водород с гомогенным катализатором и метода химического превращения воды в водород.

ESTUDIO DE LOS METODOS DE FABRICACION Y LAS FABRICAS DE AGUA PESADA SEGUN EL PROCEDIMIENTO FRANCES.

En el curso de los siete últimos años, los estudios de producción de agua pesada efectuados por la Comisaría de Energía Atómica de Francia se han centrado en el método de intercambio isotópico agua-sulfuro de hidrógeno, pero, sobre todo, en el procedimiento amoniaco-hidrógeno, en colaboración con diferentes firmas industriales, en las variantes de este procedimiento y en los medios que permiten aumentar la capacidad de producción. El procedimiento de intercambio agua-sulfuro de hidrógeno, que había sido objeto de estudios anteriores, ha sido reconsiderado últimamente bajo la óptica de producciones más importantes. Hoy día se dispone de los elementos de un proyecto de fábrica de 400 t/año, cifra que respondería a las necesidades de una posible cadena de centrales de agua pesada. El procedimiento de intercambio amoniaco-hidrógeno, materializado por la construcción de una primera fábrica en Mazingarbe (Francia), ha sido objeto de estudios muy detallados, tanto en el laboratorio, a escala semiindustrial, como sobre el plano de estudios de optimización. Las informaciones obtenidas de la explotación particularmente satisfactoria de esta fábrica y los estudios a escala piloto realizados simultáneamente, han llevado a mejorar la versión monotérmica sobre los planos técnico y económico; ellos han suministrado, también, los datos que permiten considerar la fórmula bitérmica donde las condiciones locales lo permitan. Una segunda fábrica monotérmica de 67 t/año, en construcción en Baroda (India), presenta, en relación con la de Mazingarbe, muchas mejoras. La construcción de otras fábricas está en curso de negociación con diferentes socios. Hay otras investigaciones en curso con un objetivo más lejano; se basan sobre el procedimiento de intercambio hidrógeno-aminas, que parece abrir perspectivas interesantes. Para asegurar la autonomía del procedimiento, se examinan las posibilidades del «relanzamiento» por intercambio hidrógeno-agua, realizado por catálisis homogénea, y de la conversión química del agua de alimentación en hidrógeno.

1. INTRODUCTION

La France a mis en service à la fin de 1967 l'usine d'eau lourde dont le schéma avait été décrit lors de la dernière Conférence de Genève [1]. Le succès de son fonctionnement, les progrès réalisés par les études détaillées de certaines de ses parties, ont conduit le gouvernement indien à confier la construction de deux nouvelles usines basées sur les mêmes principes, mais trois fois plus grosses, aux industriels ayant construit la majeure partie de l'usine de Mazingarbe avec la S.C.C. La construction de plusieurs ateliers d'eau lourde du même type est encore envisagée.

Nous allons décrire la marche de l'usine de Mazingarbe, en tirer les enseignements, et mentionner les améliorations réalisées sur le procédé d'échange à une température entre l'ammoniac et l'hydrogène. Nous résumerons ensuite les études relatives à des variantes de la mise en oeuvre de cet échange, ou à d'autres procédés, qui ont été poursuivies en France pour permettre à l'industrie de continuer à l'avenir à construire, en toute connaissance des différentes possibilités techniques et économiques, les usines les mieux adaptées aux problèmes posés.

2. L'USINE D'EAU LOURDE DE MAZINGARBE

2.1. Caractéristiques générales

L'usine de Mazingarbe était en 1964, lorsque sa construction fut décidée, l'unité optimale susceptible d'assurer la production de 20 tonnes par an nécessaire à la France, et permettait le passage à la taille industrielle du procédé NH_3-H_2. Dans cette première réalisation, on s'est entouré de nombreuses marges de sécurité. Citons seulement le choix d'un nombre de systèmes de contact gaz-liquide et de machines tournantes supérieur au minimum strictement requis en fonction des performances observées lors des essais pilotes, la limitation de la pression du cracking et du facteur d'enrichissement primaire à des valeurs modérées.

L'unité (Fig. 1) est liée à un nouvel atelier de synthèse d'ammoniac d'une capacité de 650 tonnes par jour. En réalité, la production n'est que de 535 tonnes, les 115 tonnes supplémentaires servant au reflux liquide des tours d'échange isotopique. Celles-ci fonctionnent à une pression voisine de la pression de synthèse d'ammoniac (400 à 450 bars). La température dans les tours d'échange est de -25°C et de -10°C. L'enrichissement primaire est de cent fois la teneur naturelle. La dissociation de l'ammoniac s'effectue à 55 - 60 bars. La température sur le catalyseur de dissociation est voisine de 550 à 580°C. La concentration finale est obtenue par une distillation d'ammoniac conduite au voisinage de la pression atmosphérique, à une température de -30°C.

La capacité de production nominale a été volontairement limitée à vingt tonnes par an.

2.2. Fonctionnement

La première production d'eau lourde de qualité nucléaire (99,8%) a eu lieu en janvier 1968, c'est-à-dire après le temps minimum de montée à l'équilibre. Nous disposons donc aujourd'hui d'une expérience industrielle de trois années, et un bilan peut être tenté.

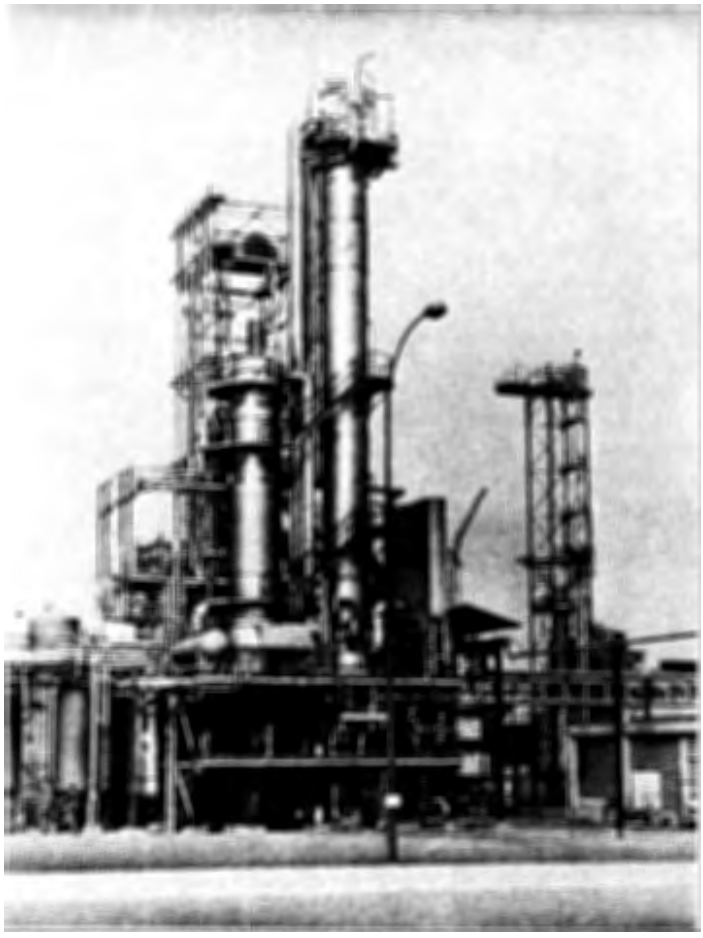

FIG. 1. Usine de Mazingarbe.
Au premier plan, installation de finition par distillation; derrière, bâti des tours d'échange; à l'arrière-plan à droite, tube de synthèse de l'usine d'ammoniac.

Dès le démarrage, la conduite de l'unité s'est trouvée aisée. Ceci est dû principalement au fait que le procédé monotherme présente par lui-même une grande stabilité. Les principales causes de perturbation n'entraînent que de faibles variations de la production. C'est ainsi qu'une modification des températures des tours d'échange isotopique de $\pm$ 5°C ne diminue la production que de deux pour cent. Il en résulte une parfaite régularité de marche. Signalons que le taux de disponibilité de l'installation, calculé sur 8760 heures, a été de 92 % au cours de la dernière année de fonctionnement.

Les problèmes de liaison avec l'atelier de synthèse d'ammoniac se concrétisent principalement par la qualité du gaz de synthèse. Celui-ci ne peut être traité dans l'usine d'eau lourde que si sa teneur en impuretés oxygénées est très

inférieure à une partie par million. C'est pourquoi nous avons été contraints de construire un atelier où la purification est menée en trois phases :

- une transformation de l'oxygène en eau sur catalyseur,
- une déshydratation du gaz sur alumine sous haute pression,
- l'élimination de l'oxygène, de l'eau résiduelle et de l'oxyde de carbone par un lavage à l'amidure de potassium.

La consommation moyenne de potassium au cours de cette dernière phase a été de l'ordre de 0,6 kilogrammes pour 100 tonnes de gaz à traiter. Elle est cependant très sensible à la teneur en oxyde de carbone du gaz de synthèse. La technologie générale de l'usine d'eau lourde ressemble à celle d'une synthèse d'ammoniac : les problèmes posés par la haute pression et l'hydrogène sont identiques. Les températures froides choisies restent d'un niveau assez élevé (-25°C) pour ne nécessiter que des aciers faiblement alliés.

2.3. Problèmes particuliers

La partie la plus complexe et aussi la plus originale est constituée par les tours d'échange isotopique. Nous rappellerons que chaque plateau d'échange, décrit en détail par ailleurs [2], comporte une couronne d'éjecteurs et deux pompes noyées dans l'amidure, destinées à vaincre la perte de charge créée par les éjecteurs. Un démontage complet ayant été effectué en 1969 pour modifier les orifices et permettre ainsi l'augmentation du débit de gaz traité, nous n'avons constaté au cours de cette visite aucune usure des paliers des pompes d'étage. Depuis le démarrage, en 1967, celles-ci ont fonctionné sans interruption.

L'atelier de dissociation d'ammoniac n'a nécessité aucune révision depuis la construction. Le catalyseur n'a pas été remplacé et semble avoir gardé son activité. C'est dans cette partie de l'installation, qui fonctionne sous pression, à haute température, et qui traite du gaz enrichi en deutérium, que le problème des fuites se pose avec le plus d'acuité. Celles-ci, que nous avons limitées par des ensembles soudés, sont restées inférieures aux limites fixées par les études du procédé (elles correspondent à quelques pour cent de la production).

Enfin, il faut remarquer que l'amidure de potassium ne provoque aucune corrosion sur les aciers courants. Le comportement des garnitures, isolants, paliers et autres matériaux susceptibles de se trouver en présence d'amidure avait fait l'objet de recherches technologiques assez poussées et n'ont causé aucune surprise.

La fabrication, le transport, le rinçage des solutions d'amidure, qui doivent être réalisés en l'absence d'oxygène et d'eau, ne présentent pas de réelles difficultés. Seules les prises d'analyses et les purges de certains appareils doivent faire l'objet de consignes particulières de sécurité. Il n'y a pas eu d'accident d'exploitation depuis le démarrage de l'unité.

3. PERFECTIONNEMENT DU PROCEDE MONOTHERME

Les études se sont poursuivies à la fois au laboratoire et dans les installations pilotes et ont abouti à des modifications de schéma [3,4].

3.1. Les essais de laboratoire

Ils ont permis de connaître avec précision la cinétique de l'étape purement chimique de la réaction d'échange, apportant ainsi une partie des

éléments nécessaires à l'interprétation des résultats obtenus en installations pilotes sur les dispositifs de contact à Mazingarbe ou à Grenoble.

Par ailleurs, ces essais ont permis de trouver un certain nombre d'adjuvants au catalyseur permettant de multiplier la vitesse de la réaction par un facteur de l'ordre de 2 à 3. Seul est envisageable l'emploi des adjuvants dont la stabilité chimique est suffisante ; à cet égard, les amines tertiaires, triméthyl- ou triéthylamine, donnent toute satisfaction. Par ailleurs, des mesures ont montré que la solubilité de l'amidure de potassium dans l'ammoniac n'était abaissée que faiblement par addition de triméthylamine. Enfin, les incidences indirectes de la présence d'adjuvants sur l'économie du procédé ont été examinées : moyens d'extraire l'adjuvant du gaz envoyé vers la synthèse d'ammoniac (étudiés en laboratoire, puis en pilote), sensibilité du catalyseur de synthèse d'ammoniac aux traces d'adjuvant résiduelles.

3.2. Les essais pilotes

Ils restaient indispensables pour étudier l'influence des paramètres de fonctionnement et de construction, que l'on n'aurait pu faire varier sur l'usine sans en perturber la marche. L'installation pilote de Mazingarbe, qui avait servi à mettre au point les éléments de l'usine, poursuivit un programme d'essai dans un grand intervalle de pression (100 à 600 bars) et de température (-25°C à + 20°C) et avec le gaz de synthèse.

Différents systèmes de contact (éjecteurs et plateaux) ont été étudiés, ainsi que différents catalyseurs d'échange isotopique, avec ou sans adjuvants.

De plus, à Grenoble, un nouveau pilote, de formule bitherme, fut construit, dont la pression maximum fut limitée à 100 bars, mais qui permettait d'expérimenter du côté basse température jusqu'à - 70°C en service continu, avec des moyens d'analyse isotopique "en ligne" ; ce pilote peut de plus fonctionner en hydrogène pur (Fig. 2).

Les deux installations de Mazingarbe et de Grenoble, travaillant suivant un programme concerté, donnent en particulier les mesures d'efficacité de contacts.

3.3. Résultats des essais - Nouveaux projets

L'interprétation des résultats des pilotes et du laboratoire et les études systématiques menées depuis six ans ont permis de construire une théorie générale qui rend compte parfaitement du fonctionnement des éjecteurs. Les efficacités de ceux-ci peuvent être, de ce fait, considérablement augmentées et les améliorations apportées seront profitables au fonctionnement des nouvelles usines indiennes. Même avant, considérant que les synthèses d'ammoniac produisant 1 000 tonnes par jour deviennent de plus en plus nombreuses, un projet détaillé d'usine pouvant servir de base à la construction d'ateliers produisant entre 60 et 70 tonnes d'eau lourde par an avait été établi.

Dans cette étude étaient incorporés :

- l'avantage économique du facteur taille par rapport à Mazingarbe (20 tonnes par an) et celui de l'augmentation du rendement d'extraction du deutérium, poussé à 80 % ou 90 %,
- l'allègement ou la simplification de certains éléments par rapport à la première réalisation,
- l'adoption de valeurs plus élevées pour l'enrichissement primaire et la pression de cracking, qui réduisent respectivement le coût de la finition et les frais de recompression du gaz.

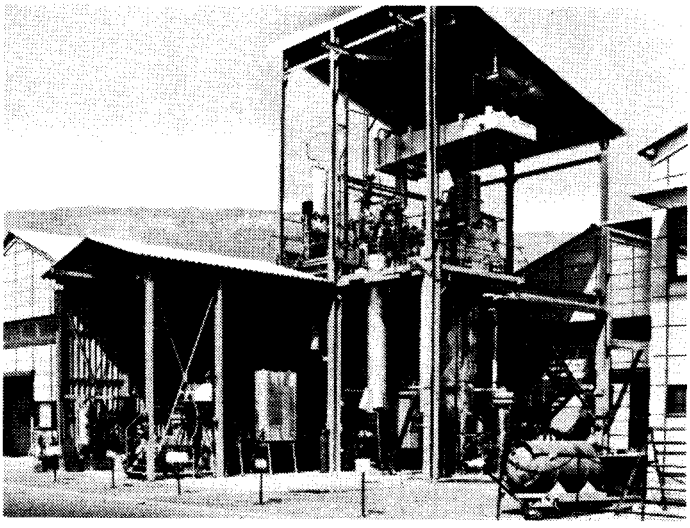

FIG. 2. Installation pilote bitherme de Grenoble.
A droite, colonnes d'échange; à gauche, hangar des machines; à l'extrême droite, salle de contrôle.

4. LES NOUVELLES USINES INDIENNES

Lorsque le gouvernement indien décida de développer sa production d'eau lourde, les industriels de l'Association NH_3-H_2, auxquels s'associa étroitement la Société Sulzer Frères, purent donc faire des propositions, qui furent retenues après une comparaison technique et économique soigneuse de toutes les solutions compétitives. Une première usine indienne fonctionnant suivant le procédé d'échange isotopique monotherme est en cours de construction à Baroda (Inde). Elle fournira 67,2 tonnes d'eau lourde par an à partir du gaz de synthèse d'une usine produisant 1 000 tonnes d'ammoniac par jour. Quelques points importants différencient cette installation de celle de Mazingarbe (Fig. 3) :

4.1. L'ammoniac liquide indispensable pour assurer le reflux dans la tour d'extraction TA est fourni par une synthèse dite "supplémentaire", propre à l'usine d'eau lourde, puisque celle-ci sera branchée sur une usine de synthèse existante. A Mazingarbe, la construction simultanée de l'usine d'ammoniac et de l'atelier d'eau lourde avait permis une incorporation des deux synthèses. Mais leur dissociation est parfaitement possible.

4.2. Dans la tour d'extraction TA la pression a la valeur imposée par l'usine de Baroda, soit 650 bars, alors qu'à Mazingarbe, elle n'était que de 450 bars.

4.3. La température d'échange, qui reste basse dans la tour TA (-27°C) a été portée à 0°C dans la tour d'enrichissement TE. L'adoption de cette dernière valeur résulte des calculs d'optimisation, qui tiennent compte de deux facteurs jouant en sens inverse : une élévation de température entraîne, certes, une diminution du facteur d'échange à l'équilibre, mais en revanche la cinétique est meilleure, donc l'efficacité du contact augmente.

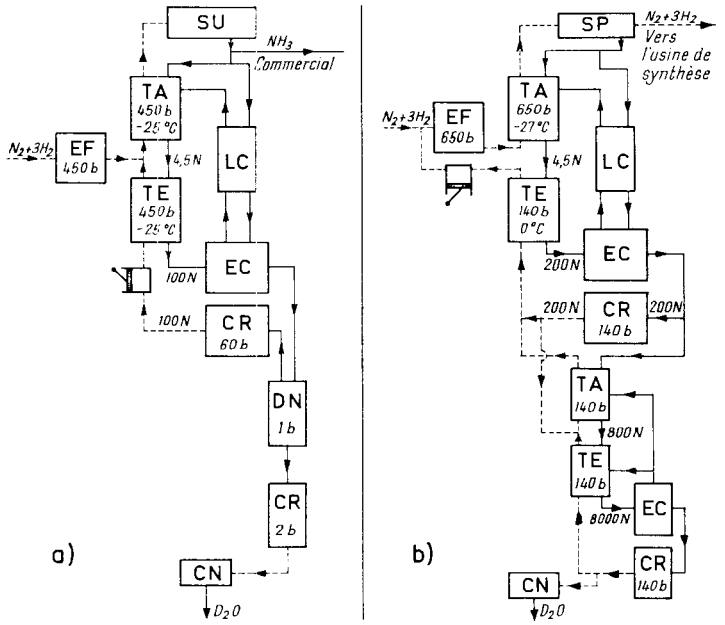

FIG. 3. Schémas comparatifs simplifiés des usines d'eau lourde de Mazingarbe a) et de Baroda b).

EF :	Epuration finale et saturation en ammoniac	CR :	Cracking de l'ammoniac
TA :	Tours d'appauvrissement	EC :	Extraction du catalyseur
TE :	Tours d'enrichissement	LC :	Lavage du catalyseur
SU :	Synthèse de l'usine d'ammoniac	DN :	Distillation de l'ammoniac
SP :	Synthèse propre à l'atelier d'eau lourde	CN :	Combustion du deutérium.

4.4. Pour diminuer l'énergie de recompression du gaz dissocié dans le cracking, la pression de celui-ci a été relevée de 60 à 140 bars. Cette valeur étant suffisante pour assurer un fonctionnement correct de la tour d'enrichissement TE, le compresseur de gaz cracké a pu être placé en aval de cette tour. Un compresseur lubrifié classique a pu ainsi être utilisé : son refoulement a été branché en amont de la station d'épuration finale qui élimine les traces d'huile entraînées. En outre, les fuites éventuelles de ce compresseur n'intéressent qu'un gaz de faible teneur isotopique.

4.5. La teneur de coupure entre la tour d'enrichissement TE et la finition a été relevée de 95 fois la teneur naturelle à 200 fois cette teneur.

4.6. L'enrichissement final est obtenu non par distillation, mais par un deuxième échange isotopique, du type monotherme comme le premier, fonctionnant entièrement à 140 bars.

4.7. Le taux d'extraction garanti, qui, pour Mazingarbe était de 63% environ, est porté à Baroda à 89%. Cette augmentation tient, pour une part, comme mentionné plus haut, aux progrès réalisés dans la connaissance des échanges par plateaux à éjecteurs et pompes en plusieurs années d'essais sur pilote et sur la première usine elle-même.

4.8. En plus de ces principales innovations, d'autres améliorations peuvent être rapidement mentionnées, en particulier : augmentation de la teneur en catalyseur dans les tours, d'où accroissement de l'efficacité ; simplification de l'unité d'extraction du catalyseur ; d'une façon générale meilleure optimisation grâce à une connaissance plus précise des divers paramètres pris en considération.

5. LES ETUDES DE PROCEDES DIFFERENTS

5.1. Généralités

Sous sa forme actuelle le procédé NH_3-H_2 est mis en oeuvre en symbiose avec des usines d'ammoniac. La limitation de production unitaire des ateliers qui en résulte ne serait pas un obstacle à un développement de la production d'eau lourde dans des pays comme la France, qui disposent d'une demi-douzaine d'usines d'ammoniac modernes capables de produire 1000 tonnes par jour, et qui pourraient donc fournir comme sous-produit jusqu'à 400 tonnes d'eau lourde par an. Mais nous avons voulu examiner si la multiplication d'ateliers était la seule solution, et comment elle se comparait à la construction d'usines de grandes tailles.

Ces études sont encore en cours.

5.2. Le procédé H_2S-H_2O

Le CEA a d'abord réexaminé dans les conditions françaises le procédé bien connu d'échange entre hydrogène sulfuré et eau. Seul, puis avec la Société pour l'obtention du deutérium, il y avait consacré des efforts importants de 1956 à 1961, et il en avait suivi le développement par l'étude de différents schémas d'échange de chaleur et l'examen de la transposition aux conditions économiques européennes des projets de Proctor et Thayer [5].
Aussi en 1968, désireux de réactualiser les évaluations technico-économiques du procédé, il confia à la Société Technip l'étude d'usines de 100, 200 et 400 tonnes/an.

Les résultats de ces travaux sont :

- que les données acquises permettraient d'entreprendre la construction de telles usines en France,
- que dans des conditions de financement équivalentes, les prix de revient français devraient être compétitifs avec ceux annoncés outre-Atlantique,
- que le coût de l'eau lourde produite diminue notablement en fonction de la taille de l'usine jusqu'à une capacité de 400 tonnes/an,
- enfin que les colonnes du plus grand diamètre réalisable aujourd'hui ne correspondent pas obligatoirement à l'optimum économique.

Mais, si l'échange H_2S-H_2O, sous réserve d'un certain nombre de précautions de mise en oeuvre, offre des avantages très importants [6], les limites du procédé, qui proviennent du facteur d'échange et du taux d'extraction du deutérium qui ne peut dépasser 20 % environ, ne semblent guère pouvoir être reculées.

5.3. Les procédés d'échange amine-hydrogène

Contrairement au cas précédent, les limites d'utilisation possible de ces procédés sont encore loin d'être bien connues [6, 7].

FIG.4. Dispositif pour l'étude de l'échange H_2 - H_2O en phase liquide.
Vue partielle. L'enceinte cylindrique calorifugée, de 40 cm de diamètre, contient l'ensemble des éléments sous pression (vaporisateur-surchauffeur; saturateur; réacteur d'échange).

5.3.1. On sait depuis longtemps que l'échange entre l'hydrogène et les amines obéit à une cinétique encore plus favorable que celle de l'échange entre l'hydrogène et l'ammoniac, avec des coefficients d'échange à l'équilibre équivalents. Il permet le travail à des températures plus basses où les séparations sont plus importantes ; enfin, la pression de vapeur des amines étant plus faible que celle de l'ammoniac, la recirculation des vapeurs serait moins coûteuse.

C'est pourquoi des études de laboratoire ont été entreprises en vue de déterminer la valeur des grandeurs nécessaires à la mise en oeuvre du procédé : cinétique et mécanisme de l'étape chimique, effet d'adjuvants, solubilité des catalyseurs, stabilité chimique, étude des produits de décomposition (important surtout pour la tour chaude d'un procédé bitherme). Des évaluations préliminaires de procédé ont été faites à partir des premiers résultats. Elles ont montré que certaines parties importantes des usines d'eau lourde pourraient en principe être réalisées de façon économiquement intéressante grâce à cet échange.

Aussi des études plus complètes sont en cours à Grenoble à l'échelle pilote, en particulier pour prouver la "faisabilité" du procédé. L'installation de formule bitherme ayant servi à l'étude de l'échange NH_3-H_2 a été aménagée à cet effet. De plus, en vue d'étudier la récupération des amines saturant le gaz épuisé en deutérium avant son introduction dans le tube de synthèse, une installation d'essais susceptible de travailler à 250 bars a été construite.

Mais des projets détaillés doivent être réalisés à l'aide de l'ensemble des résultats d'études avant que des industriels ne puissent formuler des offres sur ce procédé.

5.3.2. L'alimentation d'un échange NH_3-H_2 ou amines-H_2 à partir de l'eau permettrait de construire des unités de taille aussi grande que voulue sur un seul site. On sait que d'après certaines études ces procédés seraient d'ici quelques années économiquement plus intéressants que tous les procédés concurrents [8].

Deux procédés ont été examinés simultanément à l'échelle du laboratoire pour réaliser cette alimentation :

- Le procédé d'échange H_2-H_2O réalisé en phase liquide par catalyse homogène, pour lequel un dispositif expérimental fonctionnant jusqu'à 300°C sous 150 bars a été construit (Fig. 4) ; les premiers essais ont porté sur la catalyse basique ; ce procédé présente l'inconvénient de fournir de l'hydrogène à une teneur (90 ppm) très inférieure à la teneur naturelle, ce qui entraîne un accroissement de la taille de l'usine principale.
- Le procédé d'oxydation et réduction alternées d'une masse de contact métallique; ce procédé est prometteur par l'avantage qu'il présente de fournir de l'hydrogène à teneur naturelle ; mais il se heurte encore pour l'instant à des difficultés technologiques sérieuses ; une expérimentation sous pression atmosphérique a permis de faire un choix préalable parmi des masses à base de fer présentant à la fois une vitesse de réaction suffisante et une bonne résistance au frittage à haute température.

6. CONCLUSION

Les études de fabrication d'eau lourde en France ont permis à l'industrie de faire face aux besoins français depuis la dernière Conférence de Genève, et à l'Inde de développer son programme de production d'eau lourde. Les enseignements des réalisations en cours et les nouvelles études devraient permettre aux procédés décrits dans cette communication d'assurer la fabrication de quantités beaucoup plus importantes d'eau lourde si le besoin s'en fait sentir.

REFERENCES

[1] LEFRANCOIS B., LERAT J. M., ROTH E., Etudes de production d'eau lourde en France, Conf. int. util. énergie atom. fins pacif., Genève (1964) 28/P/91 et Rap. CEA R-2646 (1964)

[2] ROTH E., BEDHOME A;, LEFRANCOIS B., LE CHATELIER J., TILLOL A., L'usine d'eau lourde de Mazingarbe, Energie Nucléaire 10 3 (1968) 1

[3] ROTH E., Recent developments in isotope separation and heavy water preparation in France, J. Atomic Energy Soc. Japan 12 2 (1970) 80

[4] LEFRANCOIS B., L'usine d'eau lourde de Mazingarbe, Symposium nucléaire sur les aspects techniques et économiques de la production d'eau lourde, Turin, septembre 1970.

[5] PROCTOR, J.F., THAYER, V.R., Economics of heavy water production, Chem. Engng Prog. 58 4 (1962) 53-61.

[6] ROTH E., ROSTAING M., Etudes sur la production d'eau lourde en France, Symposium nucléaire sur les aspects techniques et économiques de la production d'eau lourde, Turin, Septembre 1970.

[7] BANCROFT A.R., RAE H.K., Heavy water production by amine-hydrogen exchange, Symposium nucléaire sur les aspects techniques et économiques de la production d'eau lourde, Turin, septembre 1970.

[8] BANCROFT A.R., The canadian approach to cheaper heavy water, Rapport AECL-3044 (1968) Chalk River.

DISCUSSION ON AGENDA ITEM 2.5

Developments in isotope enrichment techniques and trends in costs for enrichment services

DISCUSSION ON THE FOLLOWING GROUP OF PAPERS:

P/383 FRG Presented by E.W. Becker

P/605 France Presented by C. Leduc

P/066 USA Presented by W.E. Johnson

P/606 France Presented by M. Pecqueur

M. PECQUEUR: I have a question for Mr. Johnson concerning Fig. 3 of paper P/066, where the cumulative requirement curves are accompanied by the legend 'Reduced non-U.S. market: 5% in 1975 to 35% in 1985'. This represents a fairly considerable portion of the market and by 1985 could represent a volume in excess of 20 million units of separative work. How do you think that this market can be covered, or do you think work by plants outside the United States of America will be involved? I think the question may be of importance. In your presentation you referred to the differences in view concerning the time when the United States capacity will become saturated. I think that one of the reasons for this divergence in estimates may be related to the 'reduced non-U.S. market' component.

W.E. JOHNSON: In its projections, the United States has simply assumed that foreign separative capacity would increase during the late 1970's and 1980's to the extent that by 1985, 35% of the world demand outside of the United States, the United Kingdom and the Soviet nations would be supplied from such capacity. We also assumed that United Kingdom requirements would be met by the United Kingdom.

F. BRADY: Would Mr. Johnson care to comment, in the light of United States experience, on the technical and economic feasibility of an interruptible (variable) type of power supply to a gaseous diffusion plant. This question relates not to an unreliable power supply but to one in which planned, and possibly short-term, interruptions are used to achieve economies in the operation of a power utility system.

W.E. JOHNSON: I shall ask Mr. Sapirie, the co-author of paper P/066, to answer this question.

S.R. SAPIRIE: The key to this question is the quantity of power adjustment and the amount of time permitted for making the adjustments, through reduction of process gas inventory and pressure. We have had experience in making seasonal adjustments in power utilization for the purpose of reducing load during the season of the year in which the power suppliers have their peak demand. We thereby obtain a reduction in the power demand charge. We do not consider it economically attractive to use interruptible power that requires prompt reduction, without notice, in our existing plants.

G.R.H. GEOGHEGAN: In making his plant cost estimates, did Mr. Becker use the very good separation results obtained with a hydrogen/UF_6 mixture which are reported in his paper (P/383) or his earlier results with helium/UF_6 mixture?

E.W. BECKER: As pointed out in the paper, the estimates are based on the results achieved with hydrogen/UF_6 mixture. This mixture has been tested extensively in laboratory-scale equipment. For safety reasons, we have not been allowed, up to now, to use H_2/UF_6 mixtures within the technical stage now available.

DISCUSSION ON THE FOLLOWING GROUP OF PAPERS:

P/493 UK Presented by J.V.L. Parry

P/382 Netherlands Presented by M. Bogaardt

I.A. WEISBRODT: In connection with the developmental work on gas centrifuges in the United States of America and France, could Mr. Johnson and Mr. Pecqueur please indicate: the present state of this work; what it is expected to accomplish within the next 2-4 years; and the economic prospects or forecasts for enrichment by a gas centrifuge plant?

W.E. JOHNSON: In the United States, information concerning work on the centrifuge is still classified as restricted data and therefore I am unable to give a report on its state of development. In any event, we have not yet reached the point of proven economic performance.

M. PECQUEUR: In view of the potential importance of centrifuging, France is making a substantial effort in relation to theoretical, experimental and industrial studies of this process. However, it would appear that not all the problems in the large-scale utilization of centrifuging can be rapidly solved, and therefore the effort is essentially a long-term one. Once an experiment performed on a significant scale enables us to resolve the remaining problems (relating mainly, we believe, to the endurance of materials and the behaviour of the concentrations in the aerodynamic system constituted by a cascade of centrifuges), it is conceivable that the economic prospects of a centrifuging plant will appear in a more favourable light.

C. FREJACQUES: I should like to make a comment on the comparative investment costs for centrifugation and gaseous diffusion. Steady improvements in technology have been resulting for several years in a reduction of approximately 4% per annum in the specific investment costs for gaseous diffusion. In France, the improvement in centrifugation technology is more rapid at present, but the difference in specific investment is still two to one and should make us very cautious in estimating the date by which the labour costs for separation by the two techniques will intersect.

M. PECQUEUR: I should like to comment briefly on the respective start-up characteristics of gaseous diffusion plants and of centrifuging plants. The time required for the construction of a diffusion plant is of the same order of magnitude as the period separating the date of commissioning and the date of fuel loading in nuclear power plants. Consequently, it cannot be said that the start-up of a gaseous diffusion plant is subject to drawbacks resulting from market fluctuations.

I also have the following question. Mr. Bogaardt indicated in his presentation that, in order to cope with large-scale production, it would be necessary to manufacture a large number of machines every year – in other words, to set up a fairly elaborate system of means of production permitting operation on a serial basis. But is not the establishment of such a system indicative of a rather considerable rigidity in the programs for the development of centrifuging?

M. BOGAARDT: If we embark upon a program of mass production and assembly of large numbers of centrifuges, we would obviously make use of existing facilities for the production of components, as far as possible. There are a lot of parts, e.g. electric motors, which we would not dream of building in special facilities. It can therefore be stated that the changes in the production rate of centrifuges would not be very dramatic.

A second and even more important point is that since a large proportion of the components would be produced outside the factory, the total investment in a factory for large numbers of ultracentrifuges would be, relatively, very small compared with the specific investment in the separation facility. So I think the effect of a fluctuating rate of construction of an ultracentrifuge separation plant would not be a very serious matter.

T. TUOHY: At what price does France need to purchase electricity for the proposed French diffusion plant in order to be able to match United States prices for enrichment?

M. PECQUEUR: A great deal of caution has to be exercised in trying to answer this question. In the first place, there must be agreement as to what is meant by price. Are we talking about a price expressed in terms of constant money or of current money? Then, in determining this price, we must take into account the advantages which may result from optimization of grid and plant as a whole, a task to which Electricité de France has been devoting itself for a long time. And lastly, allowance must be made for other factors which can be affected by optimizing the cost of separation operations. Against this background, all that can be said is that the rate schedules as now planned will enable us to set an enrichment price that is competitive. It can also be shown that, in view of the growing share of nuclear energy in power generation, there is no reason for expecting that ten years or more from now there will be very different rate schedules for electricity in areas of high industrial density, such as the United States, Europe or Japan. The question that has been asked relates to the field of technology and here we think that we are in a competitive position.

P. DELAROUSSE: I should like to ask Mr. Parry whether the cost estimates presented in his paper (P/493) take into account the numerous internal isotopic mixtures which will be produced in a separation plant by the stopping, removing and replacing of elementary machines which are damaged (e.g. 10^6 machines in service with a 5-year lifetime result in 200 000 being removed annually)?

J.V.L. PARRY: Yes. There are, however, two points which should be made. Firstly, even if one accepts Mr. Delarousse's assumption on centrifuge life as correct, there is no reason to suppose that the failure pattern of centrifuges is unlike that of other machinery in giving a perfectly uniform rate of failure leading to the figure of 200 000 failures per annum stated in the question. The failure made is expected to follow the usual pattern of a small infant mortality followed by a small annual failure rate,

and then after many years the rate of failure will increase as the mean lifetime is approached. As the lifetime is known to be long, it will be possible to carry out preventative maintenance before the onset of a failure pattern signifying the approach of the mean lifetime.

Secondly, the characteristics of a centrifuge plant are quite different from those of a diffusion plant. The hold-up is extremely small – kilograms as compared with tonnes – so that incidents causing the mixing of material at different isotopic concentration will not give rise to a significant cost penalty. Similarly, the isotopic equilibrium time is measured in hours as compared with weeks or even months for the diffusion process – hence again the cost penalty for interruption of operation of parts of the plant for maintenance is small.

DISCUSSION ON THE FOLLOWING PAPER:

P/607 France Presented by E. Roth

N. AYBERS: What will be the capacity of a heavy-water plant using NH_3 and supplying power at a price competitive with that produced by the methods used at present in the United States of America and Canada?

E. ROTH: In the present version of the NH_3-H_2 process, the cost of heavy water depends on the ammonia plant with which the heavy-water facility is linked up, and more particularly on its production capacity. The most favourable price is obtained by using the ammonia syntheses having the largest available capacity. As regards competitiveness, sales by heavy-water plants this year would appear to show that for the customers, in the conditions obtaining at their plants, the process is competitive.

AGENDA ITEM 2.7

Uranium-plutonium fuel cycle for thermal and fast reactors

Cycle du combustible uranium-plutonium pour les réacteurs à neutrons thermiques et à neutrons rapides

Уран-плутониевый топливный цикл для реакторов на тепловых и быстрых нейтронах

Ciclo del combustible uranio-plutonio para reactores térmicos y para reactores rápidos

Chairman

R. B. DUFFIELD, United States of America

Vice-Chairman

W. RUTKOWSKI, Poland

Scientific Secretaries

H. A. HUGHES, United Kingdom
K. N. KOSTADINOV, IAEA

Союз Советских Социалистических
Республик
A/CONF.49/P/709

ПУТИ ЭФФЕКТИВНОГО ИСПОЛЬЗОВАНИЯ ГОРЮЧЕГО В АТОМНОЙ ЭНЕРГЕТИКЕ С БЫСТРЫМИ РЕАКТОРАМИ

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Abstract–Résumé–Аннотация–Resumen

EFFICIENT USE OF FUEL IN NUCLEAR POWER GENERATION WITH FAST REACTORS.

Sodium-cooled fast power reactors at present occupy an important place in almost all long-term nuclear power development programs. This is due, as is well known, to their capacity for fuel breeding and to the possibility of generating electricity at minimum capital cost. The refinement of nuclear data in recent years and the consequent greater accuracy of the performance data for fast reactors have shown that, in spite of somewhat poorer performance, this type still retains its advantages over thermal reactors.

The paper considers the various aspects of nuclear power development using fast reactors. It discusses the problem of reactor performance as a function of nuclear data together with the latter's effect on the fuel cycle parameters of the developing nuclear power industry. The natural fuel-cycle parameters for the period up to the year 2000 are considered in the context of using different reactor types and combinations. An evaluation is made of the influence of technical progress in reactor engineering and of the operating conditions of nuclear power stations on such parameters as the volume of natural uranium consumption.

In the light of the various methods available for further improving the performance of fast power reactors, the paper discusses the problems of optimizing this performance together with the main problems and procedures associated with building economically efficient large fast reactors. It is shown that the ceramic-fuel fast reactors now being designed can ensure the necessary long-range rate of progress in power generation at minimum consumption of natural nuclear fuel.

UTILISATION EFFICACE DU COMBUSTIBLE DANS LES REACTEURS DE PUISSANCE A NEUTRONS RAPIDES.

A l'heure actuelle, les réacteurs de puissance à neutrons rapides refroidis au sodium occupent une place de choix dans tous les programmes à longue échéance établis pour le développement de la production d'énergie d'origine nucléaire. Ce fait s'explique de toute évidence par un rapport de régénération élevé et par la possibilité de produire de l'énergie électrique avec un minimum de dépenses. Les précisions apportées pendant ces dernières années aux constantes nucléaires et, partant, aux caractéristiques des réacteurs à neutrons rapides, ont montré que malgré une certaine baisse des caractéristiques, ces réacteurs conservent intégralement leurs avantages par rapport aux réacteurs à neutrons thermiques.

Les auteurs examinent divers aspects du développement de la production d'énergie d'origine nucléaire à l'aide de réacteurs à neutrons rapides. Ils étudient la variation des caractéristiques des réacteurs en fonction des constantes nucléaires et l'influence exercée par ces constantes sur les spécifications du cycle du combustible d'un réseau en voie de développement. Ils examinent les caractéristiques naturelles d'un tel cycle jusqu'à l'an 2000 pour différentes filières de réacteurs ou des combinaisons de ces filières. Ils évaluent l'influence que le progrès technique réalisé dans la construction des réacteurs et le régime de fonctionnement des centrales nucléaires exercent sur un facteur tel que la consommation d'uranium naturel.

Il existe différentes possibilités et méthodes pour améliorer les caractéristiques des réacteurs de puissance à neutrons rapides; à ce propos les auteurs étudient les questions relatives à l'optimisation de ces caractéristiques et les principaux problèmes que pose la réalisation de grands réacteurs à neutrons rapides économiquement viables. Ils montrent que les réacteurs à neutrons rapides utilisant du combustible céramique, qui sont actuellement à l'étude, pourront assurer le développement de la production d'énergie à longue échéance au rythme voulu et réduire au minimum la consommation de combustible nucléaire naturel.

ПУТИ ЭФФЕКТИВНОГО ИСПОЛЬЗОВАНИЯ ГОРЮЧЕГО В АТОМНОЙ ЭНЕРГЕТИКЕ С БЫСТРЫМИ РЕАКТОРАМИ.

В настоящее время быстрые энергетические реакторы с натриевым теплоносителем занимают прочное место почти во всех перспективных программах развития атомной энергетики. Это, как известно, объясняется их способностью к расширенному воспроизводству горючего и возможностью производства электроэнергии с минимальными затратами средств. Проведенное в последние годы уточнение ядерных данных и соответственно характеристик быстрых реакторов показало, что, несмотря на некоторое ухудшение характеристик, быстрые реакторы полностью сохраняют свои преимущества по отношению к реакторам на тепловых нейтронах.

В докладе рассматриваются различные аспекты развития атомной энергетики с использованием быстрых реакторов. Обсуждается вопрос о зависимости реакторных характеристик от ядерных данных и о влиянии последних на показатели топливного цикла развивающейся ядерной энергетики. Рассматриваются натуральные показатели топливного цикла атомной энергетики на период до 2000 года при использовании различных типов реакторов и их комбинаций. Дается оценка влияния технического прогресса в реакторостроении и режима работы атомной электростанции на такой показатель как объем потребления природного урана.

Существуют различные пути и способы дальнейшего улучшения характеристик энергетических реакторов на быстрых нейтронах, в связи с этим обсуждаются вопросы оптимизации характеристик реактора и основные проблемы и пути создания экономически эффективных больших быстрых реакторов. В докладе показывается, что быстрые реакторы с керамическим топливом, разрабатываемые в настоящее время, могут обеспечить необходимые в перспективе темпы развития энергетики при минимальном расходе природного ядерного топлива.

METODOS DE UTILIZACION EFICAZ DEL COMBUSTIBLE EN LAS CENTRALES CON REACTORES RAPIDOS.

En la actualidad los reactores rápidos de potencia con sodio como refrigerante ocupan un lugar destacado en casi todos los programas de gran porvenir para la utilización de la energía atómica. Esto, como se sabe, se explica por su capacidad para una reproducción a gran escala de combustible y la posibilidad de producir energía eléctrica con un mínimo de gastos. La precisión, obtenida en los últimos años, de los datos nucleares y, por tanto, de las características de reactores rápidos, ha puesto de manifiesto que, a pesar de cierto empeoramiento de las características, los reactores rápidos mantienen plenamente sus ventajas con respecto a los reactores térmicos.

En la memoria se consideran diferentes aspectos del desarrollo de la energía atómica con utilización de reactores rápidos. Se discute el problema de la relación entre las características del reactor y sus parámetros nucleares y de la influencia de estos últimos sobre los índices del ciclo de combustible en la industria de la energía nuclear que se está desarrollando. Se consideran los índices naturales del ciclo de combustible en la industria de la energía nuclear durante el período que abarca hasta el año 2000, con la utilización de diferentes tipos de reactores y de sus combinaciones. Se evalúa la influencia del progreso técnico sobre la construcción de reactores, y del régimen de trabajo de las centrales nucleares sobre los índices tales como el volumen de utilización de uranio natural.

Existen diferentes vías y métodos para el perfeccionamiento ulterior de las características de los reactores de potencia rápidos; en relación con ello se discuten los problemas de optimización de las características del reactor, y los problemas básicos y los métodos de desarrollo de grandes reactores rápidos, económicamente rentables. En la memoria se demuestra que los reactores rápidos con combustible cerámico que se desarrollan actualmente, pueden asegurar los necesarios y prometedores ritmos del desarrollo de la energía nuclear con un consumo mínimo de combustible nuclear natural.

1. ВВЕДЕНИЕ

Хорошо известно, что запасы урана на земле достаточны для обеспечения энергетических потребностей людей путем использования ядерных реакторов деления на любой обозримый период времени. Однако, по современным представлениям, лишь ничтожная доля этих запасов может быть отнесена к категории пригодных для экономического использования в ядерных реакторах на тепловых нейтронах. Реактор, охлаждаемый обычной водой (ЛВР), служащий сейчас основой развития ядерной энергетики

большинства стран, потребляет 0,6-0,8 кг ^{235}U в расчете на 1 МВт электрической мощности в год. При современных ценах на природный уран $C_U \approx 20$ долл/кг это приводит к урановой рудной составляющей стоимости 1 квт·ч электроэнергии 0,04 цент/квт·ч, что примерно на порядок ниже затрат на органическое горючее и обусловливает экономическую эффективность ядерной энергетики.

Переход же по мере расходования ресурсов к урану по цене 50-100 50-100 долл/кг означал бы заметное повышение затрат на атомную электроэнергию.

Таким образом, ядерная энергетика, призванная решить топливно-энергетическую проблему, сама оказывается перед такой проблемой.

Известны две основные концепции решения проблемы ядерного горючего. Некоторые специалисты предлагают вкладывать средства в разведку новых месторождений, разработку дешевых способов извлечения урана из бедных руд, усовершенствование тепловых реакторов и развитие ториевого топливного цикла. Другие, в том числе авторы настоящего доклада, видят кардинальное решение топливной проблемы в реакторах-размножителях на быстрых нейтронах.

При достижении определенных показателей воспроизводства эти реакторы способны вовлечь в энергетику огромные запасы урана и тория в будущем, но и в ближайшее время они позволят развить энергетику до крупных масштабов в рамках известных ресурсов дешевого урана.

Исследования и опытные работы, выполненные в некоторых странах в течение последних двух десятилетий, дают возможность рассчитывать на решение проблемы быстрых реакторов с натриевым охлаждением, включая освоение крупных атомных электростанций (АЭС) с такими реакторами, уже в ближайшие годы. Быстрые реакторы составляют сейчас основу перспективных программ развития ядерной энергетики.

Внедрение быстрых реакторов с коэффициентом воспроизводства КВ > 1 еще не решает проблемы ядерного горючего. Для этого необходимо, чтобы КВ превышал единицу с достаточным запасом, позволяющим обеспечить вторичным горючим как догрузку существующих, так и ввод новых реакторов. Другими словами, собственный темп роста системы быстрых реакторов ω_0 должен быть по крайней мере равным темпу роста энергетики ω. Для того чтобы быстрые реакторы успели придти на смену тепловым реакторам, пока те не исчерпали запасов дешевого урана, собственный темп их роста должен быть в достаточной мере выше:

$$\omega_0 > \omega \qquad (1)$$

Определению требований к быстрым реакторам, вытекающих из задачи экономии урановых ресурсов, и исследованию их возможностей с этой точки зрения посвящен настоящий доклад. При этом ограничимся рассмотрением быстрых реакторов с натриевым охлаждением и керамическим горючим, получающих наибольшее развитие. Что касается мероприятий по усовершенствованию показателей расхода горючего, то здесь будут рассмотрены лишь те из них, которые в настоящее время изучаются всерьез. Основанием для такого "практицизма" служит то, что уже и этих усовершенствований оказывается достаточно.

Оценке и оптимизации реакторов с точки зрения расходования природного урана, взятого в натуральном выражении, в сравнении с известными его ресурсами иногда противопоставляется "экономический" под-

ход. Экономические критерии позволяют наиболее полно учесть разнородные затраты на производство электроэнергии и в конечном счете определяют выбор реакторов. Однако, будучи рассмотренными при существующих ныне ценах на уран, они не отражают в достаточной мере необходимости в экономии урановых ресурсов, не учитывают одно из главных экономических преимуществ быстрых реакторов – возможности развития энергетики без существенного расширения топливной базы в ближайшие десятилетия. Этим и оправдывается интерес наряду с экономическими и к натуральным показателям расхода урана.

Поскольку при оптимизации желательно (а в строгой постановке необходимо) иметь единый критерий, попытаемся в дальнейшем объединить содержание натуральных и экономических критериев, исходя из предположения об ограниченности запасов дешевого урана.

Развитие ядерной энергетики можно условно разделить на два этапа. На первом из них ядерная энергетика быстро развивается, занимая скромное место в производстве электроэнергии. На втором этапе, достигнув масштабов одного из главных производителей энергии, она замедлит темпы своего роста до общеэнергетических. Рассмотрим сначала требования к быстрым реакторам, выдвигаемые энергетикой отдаленного будущего.

2. ПЕРСПЕКТИВЫ АТОМНОЙ ЭНЕРГЕТИКИ

Учитывая масштабы энергетики будущего, необходимо потребовать, чтобы быстрые реакторы работали в режиме самообеспечения горючим, т.е. чтобы их собственный темп был равен темпу роста энергетики:

$$\omega_0 = \omega$$

В этом случае потребление урана ядерной энергетикой ограничивается использованием его в качестве сырья для зон воспроизводства быстрых реакторов. В течение длительного времени, пока используется обогащенный уран, это не потребует дополнительных затрат урана, так как отвала обогатительного производства будет вполне достаточно. Проблема природного урана (а может быть, и тория) как сырья возникает много позже и вполне разрешима, так как для этой цели экономически допустимо применять и очень дорогой уран. Величина ω_0 зависит как от собственных характеристик реакторов и топливного цикла, так и от условий их работы в энергетической системе (коэффициента нагрузки φ). Чтобы выделить эту зависимость, введем в качестве характеристики собственно реактора время удвоения при $\varphi = 0{,}8$ ($T_2^{0{,}8}$). Тогда

$$\omega_0 = \frac{\ln 2}{T_2^{0,8}} \cdot \frac{\varphi(1 + 0{,}8T_п/T_a)}{0{,}8(1 + \varphi T_п/T_a)} \qquad (2)$$

и требуемое время удвоения в зависимости от ω и φ равно

$$T_2^{0,8} = \frac{\varphi(1 + 0{,}8T_п/T_a)\ln 2}{0{,}8(1 + \varphi T_п/T_a)\,\omega} \qquad (3)$$

Здесь $T_п$ – время внешнего топливного цикла; T_a – кампания активной зоны при $\varphi = 1{,}0$.

Когда ядерная энергетика достигнет больших масштабов, ей придется хотя бы частично взять на себя функции регулирования мощности в энергосистемах. Хотя относительно низкие топливные и высокие капитальные затраты АЭС будут стимулировать мероприятия, препятствующие снижению φ, будем считать, что коэффициент нагрузки окажется в пределах 0,6-0,8.

Ниже приведены требуемые величины $T_2^{0,8}$ в зависимости от темпов роста энергетики при φ, изменяющемся в указанном диапазоне:

ω, 1/год	$T_2^{0,8}$, годы
0,10	6-7
0,08	7-9
0,06	9-11

Таким образом, даже современные быстрые реакторы с окисным горючим, которые при времени внешнего топливного цикла $T_п$ = 0,5 года могут иметь время удвоения $T_2^{0,8}$ = 9 лет, обеспечивают темпы роста энергетики ~ 6% в год.

Более высокие темпы роста энергетики, характерные для нашей страны в настоящее время ($\omega \approx 8\%$ в год), потребуют усовершенствования окисных реакторов или перехода на новые виды горючего, например монокарбид, что дает снижение $T_2^{0,8}$ до 6-8 лет. Поэтому не вызывает сомнений возможность использования быстрых реакторов разрабатываемого сейчас типа для обеспечения перспектив развития ядерной энергетики.

Более актуален вопрос о более высоких темпах роста ядерной энергетики в ближайшие десятилетия. Для того чтобы в обозримый период можно было бы перейти на строительство только быстрых бридеров и прекратить сооружение тепловых реакторов, собственный темп роста быстрых бридеров ω_0 должен быть несколько более высоким, чем ω. Из анализа развития системы тепловые конвертеры – быстрые бридеры следует, что время выхода в такой режим

$$t_0 \approx \frac{1}{r_т/g_б - (\omega_0 - \omega)} \ln \frac{r_т/g_б}{\omega_0 - \omega} \qquad (4)$$

Здесь $r_т$ – удельное производство плутония в тепловом конвертере; $g_б$ – удельная загрузка плутония в топливном цикле быстрого бридера.

При $r_т \approx 0,2$ кг/МВт(эл) год и $g_б \approx 4$ кг/МВт(эл) можно вывести ядерную энергетику в такой режим (т.е. прекратить строительство тепловых реакторов) в обозримый срок ($t_0 \approx 30$ лет), если $\omega_0 - \omega \approx 0,2$, т.е. $\omega_0 \approx 10\%$ в год, так что требуемые времена удвоения составляют 6-7 лет. Формула для времени удвоения в наиболее полном виде получена в работе [1]:

$$T_2 = \frac{\bar{g}_а\left(1 + \varphi\frac{T_п}{T_а}\right) + \bar{g}_{э.т}\left(1 + \varphi\frac{2T_п}{T_а}\right) + \rightarrow}{\rightarrow + \bar{g}_{э.б}\left(1 + \varphi\frac{T_п}{T_{э.б}}\right)} \ln 2 \qquad (3')$$

$$\varphi[\,y(КВ - 1) - Y(1 - \epsilon)]$$

Здесь $\bar{g}_а$, $\bar{g}_{э.т}$, $\bar{g}_{э.б}$ – среднее количество плутония в активной зоне, в торцовом экране и в боковом экране на единицу мощности.соответственно;

y – количество плутония, сгорающее в год на единицу мощности; Y – полное количество плутония, выгружаемое из реактора в год на единицу мощности; ϵ – коэффициент возврата из предела, учитывающий потери; $T_{э.б}$ – среднее время задержки плутония в боковом экране; КВ – полный коэффициент воспроизводства реактора.

Основными путями снижения величины T_2 являются:

1) уменьшение количества плутония в топливном цикле за счет сокращения времени внешнего цикла;

2) снижение количества горючего в цикле за счет увеличения удельной мощности горючего при помощи выравнивания поля тепловыделения;

3) увеличение воспроизводства горючего при переходе к новым его видам и усовершенствовании твэлов и пакетов;

4) увеличение глубины выгорания горючего.

Результаты расчетов показывают, что рассматриваемые усовершенствования быстрых реакторов позволяют сохранить расход природного урана для развития ядерной энергетики на уровне известных ресурсов дешевого урана.

Дополнительная экономия урана, если это потребовалось бы, может быть достигнута путем усовершенствования тепловых реакторов или же внедрения, особенно в начальный период, быстрых урановых конвертеров вместо части ЛВР.

В следующем разделе анализируются способы снижения расхода горючего за счет увеличения его удельной мощности при помощи выравнивания радиального поля тепловыделения в активной зоне и стабилизации его во времени для реакторов большой мощности.

3. ЭНЕРГЕТИЧЕСКИЕ РЕАКТОРЫ НА БЫСТРЫХ НЕЙТРОНАХ БОЛЬШОЙ МОЩНОСТИ

Для АЭС с ядерными реакторами на быстрых нейтронах характерным для ближайшего будущего явится рост единичных мощностей до 1000–2000 МВт(эл). Это целесообразно как в отношении снижения удельных капитальных вложений, так и с точки зрения экономики топливного цикла [2].

Основные особенности физики быстрых реакторов большой мощности обусловлены следующими факторами:

1) ростом внутреннего коэффициента воспроизводства реактивности и близостью его к единице, что упрощает проблему компенсации изменения реактивности при выгорании и обеспечивает возможность длительной непрерывной работы реактора без перегрузок (до ~ 1 года и более);

2) необходимостью выравнивания профиля поля тепловыделения в активной зоне реактора, что обеспечивает снижение удельного количества горючего в цикле и сохранение полного коэффициента воспроизводства с ростом мощности реактора.

Как известно, наибольшее распространение в настоящее время имеет способ выравнивания тепловыделения путем создания зон разного обогащения в активной зоне реактора. Изучение зависимости физических характеристик реакторов от их мощности проводилось с участием С.Т.Лескина, А.И.Новожилова.

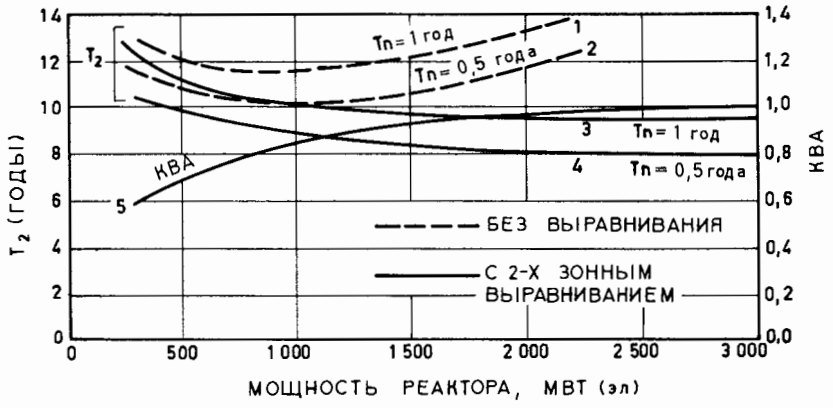

Рис.1. Зависимость времени удвоения и внутреннего коэффициента воспроизводства от электрической мощности реактора при временах внешнего топливного цикла $T_п$ = 1 год и 0,5 года, топливо UO_2-PuO_2, максимальная глубина выгорания 100 000 МВт·сут/т:
1 – время удвоения T_2, без выравнивания тепловыделения, внешний цикл $T_п$ = 1 год;
2 – время удвоения T_2, без выравнивания тепловыделения, внешний цикл $T_п$ = 0,5 года;
3 – время удвоения T_2, с выравниванием тепловыделения, внешний цикл $T_п$ = 1 год;
4 – время удвоения T_2, с выравниванием тепловыделения, внешний цикл $T_п$ = 0,5 года;
5 – внутренний коэффициент воспроизводства.

Результаты этих исследований показаны на рис.1. Как видно из рисунка, увеличение мощности реактора сопровождается улучшением его физических характеристик.

Для реактора типа БН-600 с окисным горючим с выравниванием тепловыделения зонами разного обогащения при времени внешнего цикла 0,5 и 1 год время удвоения получается равным 9 и 11 годам соответственно. Интересы развития ядерной энергетики требуют, чтобы это время составляло 6-8 лет.

Изучение возможностей реакторов на быстрых нейтронах требует учета определенного технического прогресса в реакторостроении (усовершенствование конструкции топливных кассет и твэлов, улучшение технологии и разработка новых топливных композиций) [2].

С другой стороны, использование наиболее распространенного способа выравнивания тепловыделения путем создания подзон разного обогащения приводит к значительной временно́й нестабильности профиля поля тепловыделения при длительной работе реактора большой мощности без перегрузок (0,5 года и более). В процессе работы такого реактора происходит относительный рост тепловыделения в центре активной зоны из-за пространственного перераспределения концентраций делящихся изотопов и нейтронного потока, что в конечном счете становится определяющим условием продолжительности непрерывной работы реактора. Вопросы временно́й нестабильности профиля поля тепловыделения исследовались при участии С.Б.Боброва, В.М.Мурогова, А.И.Новожилова, Л.В.Точенова, А.Н.Шмелева, В.Г.Илюнина, И.Д.Ракитина и др. [3-5]. С.М.Зарицким, В.М.Муроговым и А.Н.Шмелевым был разработан способ выравнивания профиля тепловыделения в реакторах на быстрых нейтронах, обеспечивающий его временну́ю стабильность [6] за счет создания подзон с различными делящимися и воспроизводящими изотопами.

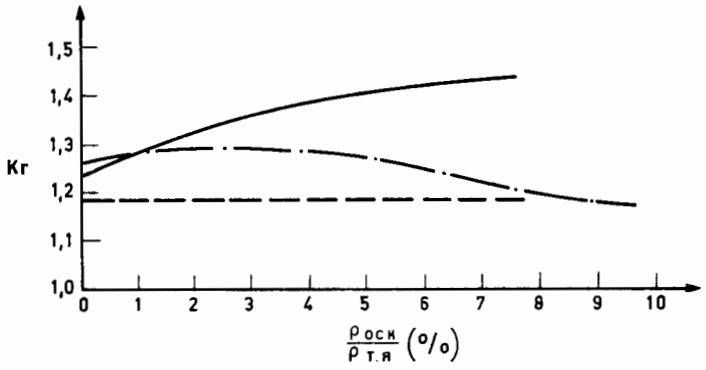

Рис.2. Зависимость радиального коэффициента неравномерности тепловыделения K_r от времени непрерывной работы реактора (на оси абсцисс указана глубина выгорания % вес):
—— – выравнивание двумя обогащениями,
—·— – выравнивание составом,
- - - – реактор со стабильным полем тепловыделения.

На основе указанного способа выравнивания тепловыделения Е.И.Гришаниным, В.М.Муроговым, В.В.Орловым и Л.В.Точеным был предложен быстрый энергетический реактор со стабильным во времени профилем поля тепловыделения (БСР – быстрый стабильный реактор).

Исследования, проведенные при участии В.Г.Илюнина, А.М.Кузьмина, В.М.Мурогова, А.Н.Шмелева, Ю.В.Силаева, показали, что в таком реакторе достигается как наименьшее значение коэффициента неравномерности тепловыделения, так и неизменность поля тепловыделения в процессе кампании (рис.2). В результате обеспечивается возможность длительной непрерывной работы без перегрузок в процессе кампании реактора и значительное повышение энергонапряженности горючего в топливном цикле. Рис.2 иллюстрирует изменение радиального коэффициента неравномерности $K_т$ для различных способов выравнивания в зависимости от глубины выгорания, достигаемой в реакторе за время работы без перегрузки. Существенным является отличие поведения поля тепловыделения в плутониевом реакторе, выравненном обогащением, и в стабильном реакторе.

Дополнительные преимущества в реакторах большой мощности имеет способ выравнивания составом (разный шаг твэлов или разный диаметр твэлов в зонах).

В результате исследований, проведенных при участии А.И.Новожилова, С.Б.Боброва, В.Г.Илюнина, А.М.Кузьмина, В.М.Мурогова, Ю.В.Силаева, А.Н.Шмелева, было установлено, что стабильность профиля поля тепловыделения при выравнивании составом, достигаемая за счет равенства зонных коэффициентов воспроизводства, обеспечивает достаточно высокий темп воспроизводства и энергонапряженность горючего в цикле (см.рис.2). При этом способе выравнивания упрощается также решение некоторых вопросов безопасности, что связано с более отрицательным значением натриевого и допплеровского коэффициентов реактивности.

Что касается способа выравнивания обогащением, то работы, выполненные при участии С.Б.Боброва и А.И.Новожилова, показали, что оптимальное размещение плутониевого горючего с различным содержанием

высших изотопов плутония (^{240}Pu, ^{241}Pu и ^{242}Pu) в значительной мере улучшает временно́е поведение поля тепловыделения и повышает энергонапряженность и темп воспроизводства горючего.

Корректное и разностороннее изучение различных способов выравнивания невозможно без проведения комплексного расчета активной зоны быстрого реактора (т.е. теплового, гидравлического, прочностного и нейтроннофизического расчета в комплексе). Для успешного решения этих задач при участии А.М.Кузьмина, А.А.Кашутина, Ю.В.Силаева, Л.В.Точеного, В.В.Хромова, В.А.Апсэ был разработан комплекс программ для оптимизационных исследований быстрых реакторов [7-9]. Оптимизационная программа "РОКБАР" позволяет проводить указанный комплексный расчет активной зоны быстрого реактора с натриевым охлаждением с определением оптимальных (по тем или иным критериям) композиций и параметров реакторов: размеров активной зоны; шага решетки твэлов, диаметра и толщины оболочки твэлов; скорости теплоносителя и объемной доли стенок кассет; высоты газовой полости; обогащения горючего.

При этом вводятся ограничения на некоторые параметры, функционально зависящие от управлений, например температуру горючего и оболочки наиболее напряженного твэла (с учетом факторов перегрева), напряжение в оболочке твэла и стенках кассет, величину среднего подогрева теплоносителя (с учетом дросселирования).

Физический расчет в программе "РОКБАР" проводится эффективным малогрупповым методом условного разделения пространственных и энергетических переменных. Поиск оптимального варианта реактора осуществляется при помощи итерационного метода. На каждом шаге итераций исходная задача линеаризуется (с использованием теории малых возмущений), а полученная при этом линейная задача решается методом последовательного сокращения невязок [10].

В табл.I на основе результатов оптимизационных исследований больших быстрых реакторов, проведенных С.Б.Бобровым, В.Г.Илюниным, А.М.Кузьминым, В.М.Муроговым, А.И.Новожиловым, Ю.В.Силаевым, А.Н.Шмелевым, приведены характеристики быстрых реакторов большой мощности [1500 МВт(эл)] с различными способами выравнивания (с учетом ожидаемого прогресса реакторостроения): БОП-2 – усовершенствованный окисный урановый реактор, выравненный обогащением; БСР – усовершенствованный реактор со стабильным полем тепловыделения; БОБ-1 – усовершенствованный окисный плутониевый реактор, выравненный обогащением; БОБ-2 – усовершенствованный окисный плутониевый реактор, выравненный составом; БКБ – усовершенствованный карбидный плутониевый реактор, выравненный обогащением.

Исходные технологические и конструкционные параметры, положенные в основу оптимизации, представлены в табл.II.

Некоторые из принятых здесь для оптимизационного расчета исходных данных взяты с учетом определенного технического прогресса (значительное гидравлическое сопротивление пакетов, твэлы с выпуском газовых осколков). Что касается глубины выгорания, то в расчете использованы умеренные перспективные оценки этой величины (максимальная глубина выгорания 100 000 МВт·сутки/т). В связи с этим надо заметить, что обнаруженное в последние годы явление радиационного распухания сталей может потребовать дополнительных технических мероприятий для достижения принятых глубин выгорания.

ТАБЛИЦА I. ХАРАКТЕРИСТИКИ ПЕРСПЕКТИВНЫХ БЫСТРЫХ РЕАКТОРОВ БОЛЬШОЙ МОЩНОСТИ

Параметр	БОП-2	БСР	БОБ-1	БОБ-2	БКБ
Тепловая мощность, МВт	3750	3750	3750	3750	3750
Электрическая мощность, МВт	1500	1500	1500	1500	1500
Плотность тепловыделения, кВт/л	650	760	550	540	650
Способ выравнивания тепловыделения	Два обогащения	По данным работы [6]	Два обогащения	Составом	Два обогащения
Коэффициент неравномерности:					
радиальный	1,27	1,18	1,37	1,26	1,3
аксиальный	1,26	1,27	1,24	1,25	1,25
Загрузка активной зоны, т	3,45	2,50	2,93	2,85	2,74
Удельная загрузка активной зоны, т/млн.кВт(эл)	2,3	1,65	1,95	1,90	1,83
Диаметр твэлов, мм	5,8	5,8	6,8	5,8-6,25	7,0
Коэффициент воспроизводства	0,9	1,1	1,4	1,38	1,55
Время удвоения при коэффициенте нагрузки 0,8:					
$T_п$ = 1 год	–	–	9	9,5	7,3
$T_п$ = 0,5 года	–	–	7,5	7,5	6

ТАБЛИЦА II. ИСХОДНЫЕ ТЕХНОЛОГИЧЕСКИЕ И КОНСТРУКЦИОННЫЕ ДАННЫЕ ДЛЯ ОПТИМИЗАЦИОННОГО РАСЧЕТА

Параметр	Двуокись	Монокарбид
Максимальная температура горючего, °С	2450	1600
Максимальная температура оболочки, °С	725	725
Плотность горючего, г/см³	8,7	11
Толщина оболочки, мм	0,4	0,3
Материал оболочки	Нержавеющая сталь	Нержавеющая сталь
Максимальная глубина выгорания, МВт·сутки/т	100 000	100 000
Тип твэла	Герметичные	С выпуском газа
Предельная относительная деформация оболочки	0,002	0,002
Перепад давления в реакторе, ата	12	12
Теплопроводность горючего, ккал/м.ч.°С	2,2	16

4. РАСХОД ПРИРОДНОГО УРАНА

Примем, что развитие ядерной энергетики описывается выражением

$$N(t) = \frac{A}{\omega}\left(e^{\omega t} - 1\right) + N(0) \qquad (5)$$

где $N(t)$ – уровень мощности в момент времени t; A – постоянная, в данном случае $A = 2{,}38$; ω – заданный асимптотический темп роста мощности, в данном случае $\omega = 0{,}08$; $N(0)$ – начальный уровень мощности, в данном случае $N(0) = 2$. Рассматриваемый отрезок времени – 30 лет. Расчеты потребления урана выполнены при участии Л.С.Ануфриенко.

На рис.3 показан диапазон изменения расхода природного урана за 30 лет при использовании комбинаций урановых и плутониевых реакторов с различными характеристиками.

Кривая 1 дает расход урана для урановых плутониевых реакторов (БОБ + БОП) с "современными" характеристиками, кривая 2 относится к быстрым реакторам со стабильным полем тепловыделения (БСР) и, наконец, кривая 3 показывает, как сильно может снизиться расход урана при использовании монокарбидного бридера с перспективными характеристиками (БКБ). Во всех трех вариантах принят постоянный коэффициент нагрузки $\varphi = 0{,}8$ и постоянная концентрации ^{235}U в отвальном уране $x_0 = 0{,}25\%$; время внешнего цикла также постоянно и равно 1 году. В случае сокращения длительности внешнего топливного цикла до 0,5 года (что представляется технически возможным) расход урана может быть снижен еще на 25-35%.

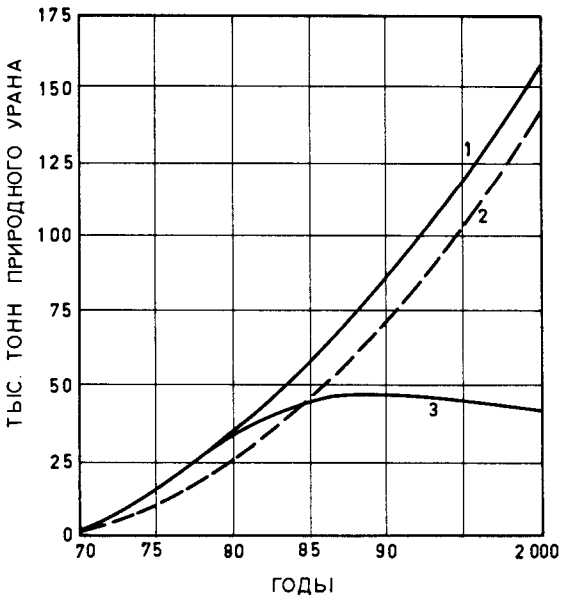

Рис.3. Суммарный расход природного урана в зависимости от характеристик быстрых реакторов:
1 – реакторы БОП + БОБ; 2 – реакторы БСР; 3 – реакторы БОП + БКБ.

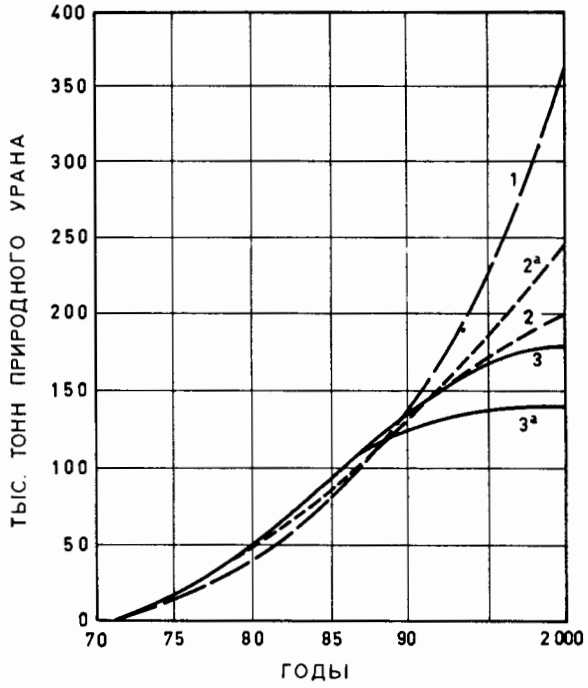

Рис.4. Суммарный расход природного урана для различных комбинаций реакторов: 1 – реакторы ЛВР-1 + ЛВР-2; 2 – реакторы ЛВР-1 + ЛВР-2 + БОБ + БКБ; 2а – реакторы ЛВР-1 + ЛВР-2 + БОБ + БКБ, $T_п$ = 0,5 года; 3 – ЛВР-1 + БОП + БОП-2 + БОБ + БКБ; 3а – ЛВР-1 + БОП + БОП-2 + БОБ + БКБ, $T_п$ = 0,5 года.

Рассмотренные выше варианты представляют в основном теоретический интерес для анализа возможностей одних быстрых реакторов. В реальных условиях ядерная энергетика начинает свое развитие с реакторов на тепловых нейтронах, так как они сейчас наиболее отработаны и составляют основу ядерной энергетики во всех странах. Известно, что развитие ядерной энергетики на одних тепловых реакторах с циркуляцией плутония потребовало бы значительно больше природного урана, чем для вариантов развития энергетики с быстрыми плутониевыми реакторами, так как в тепловых реакторах плутоний используется неэффективно. Причем это различие должно возрастать с увеличением рассматриваемого интервала времени. Указанный эффект проявляется уже достаточно сильно в пределах ближайших 30 лет.

На рис.4 представлены кривые суммарного расхода природного урана для трех вариантов развития ядерной энергетики в соответствии с выражением (5). Приведенные цифры носят чисто иллюстративный характер.

В качестве тепловых реакторов взяты два варианта легководных реакторов – современный (ЛВР-1) и перспективный (ЛВР-2); их характеристики даны в табл.III (они примерно соответствуют данным работы [11]). Предполагается, что первые 10 лет все АЭС работают с коэффициентом нагрузки $\varphi = 0{,}8$, а следующие 20 лет коэффициент нагрузки равен 0,7.

ТАБЛИЦА III. ХАРАКТЕРИСТИКИ ТЕПЛОВЫХ РЕАКТОРОВ

Параметр	ЛВР-1	ЛВР-2
Загрузка, т природного урана/млн. кВт(эл)	650	400
Расход на подпитку (φ = 0,8), т природного урана/млн. кВт(эл)·год	160	120
Производство плутония (φ = 0,8), т делящегося Pu/млн. кВт(эл)·год	0,200	0,200

Если в системе есть быстрые реакторы, после первых 10 лет коэффициент нагрузки снижается только для АЭС с тепловыми реакторами; быстрые реакторы все время работают с коэффициентом нагрузки 0,8; при этом φ тепловых реакторов в зависимости от их числа снижается настолько, что общий средний коэффициент нагрузки составляет ~0,7. Если не оговорено особо время внешнего топливного цикла, оно принято равным 1 году.

Рассмотрены следующие варианты (см.рис.4).

1. Легководные реакторы ЛВР-1 первые 10 лет плюс легководные реакторы ЛВР-2 следующие 20 лет; вырабатываемый плутоний используется в этих же реакторах; эффект использования плутония учтен путем уменьшения общего расхода урана на 20%.

2. Легководные реакторы ЛВР-1 первые 10 лет плюс легководные реакторы ЛВР-2 последующие 20 лет совместно с быстрыми плутониевыми реакторами; быстрые плутониевые реакторы на окисном горючем (БОБ) начинают вводиться по прошествии первых 10 лет, затем еще через 5 лет вместо них строятся плутониевые быстрые реакторы на карбидном горючем (БКБ).

2а. То же, что и вариант 2, но время внешнего топливного цикла $T_п$ = 0,5 года.

3. Легководные реакторы ЛВР-1 строятся первые 10 лет, затем вместо них совместно с быстрыми реакторами (БОБ) по мере надобности сооружаются быстрые урановые реакторы (БОП); через 15 лет после начала развития энергетики вместо окисных бридеров (БОБ) строятся карбидные бридеры (БКБ) и вместо реакторов БОП по мере необходимости – усовершенствованные реакторы БОП-2.

3а. То же, что и вариант 3, но время внешнего топливного цикла уменьшено с 1 до 0,5 года после 1985 г.

Результаты расчетов показывают, что в варианте с одними тепловыми реакторами осуществляется несколько меньший суммарный расход урана по сравнению с остальными вариантами в течение примерно первых 17 лет, однако к концу второго десятилетия все кривые расхода пересекаются и расход урана в варианте с одними тепловыми реакторами после этого становится больше, чем в остальных вариантах, и эта разница быстро нарастает со временем.

К концу третьего десятилетия экономия в расходе урана для вариантов с быстрыми реакторами по сравнению с "всетепловым" вариантом 1 составляет 33-62%.

Наиболее экономичными по расходу урана оказываются варианты, в которых по истечении первых 10 лет вместо тепловых урановых реакто-

ров строятся по мере надобности в дополнение к быстрым бридерам быстрые урановые реакторы.

При использовании урановых быстрых реакторов обеспечивается не только наилучшая экономия урана за 30 лет, но и минимальный его расход в дальнейшем за пределами этого периода. Имеется в виду расход урана на подпитку АЭС с урановыми реакторами, введенными за рассматриваемый 30-летний интервал, до конца их срока службы. Расход этот для варианта 1 с одними тепловыми реакторами будет намного больше, чем в вариантах 3 и 3а, где тепловых реакторов мало и, кроме того, имеется возможность применять для подпитки после 2000 г. избыточный плутоний из быстрых реакторов.

Как уже отмечалось, для перспективных быстрых реакторов здесь приняты умеренные величины максимальной глубины выгорания, которые достижимы для окисного горючего уже сейчас.

Увеличение глубины выгорания остается, таким образом, возможным резервом дальнейшего повышения эффективности топливного цикла. Здесь не рассмотрены возможности дополнительного улучшения характеристик при использовании металлического горючего, так как пока нет достаточных опытных данных о допустимых его характеристиках (температура, плотность, легирование, глубина выгорания и т.д.).

Что касается влияния на приведенные результаты по расчету потребности в природном уране масштабов развития ядерной энергетики, то можно считать, что конечные итоговые цифры в определенных пределах пропорциональны уровню мощности на конец рассматриваемого периода.

Следует остановиться еще на влиянии неточности ядерных данных на результаты расчета характеристик быстрых реакторов и объемов потребления природного урана. Настоящие расчеты физических характеристик реакторов сделаны на основе 26-групповой системы констант БНАБ [12], скорректированной в 1969 г. по последним экспериментальным данным для ^{239}Pu, ^{238}U и ^{235}U [13-15].

В результате коррекции сечений произошло небольшое возрастание удельной загрузки (~ на 6%) и снижение коэффициента воспроизводства рассматриваемых типов быстрых реакторов (на 0,05); характеристики реакторов в табл.I даны с учетом этих изменений.

По оценкам, приведенным в работе [16], точность расчета таких характеристик большого быстрого реактора, как коэффициент воспроизводства, составляет ±0,1, а критической массы – ±8%. Это объясняется существующей неопределенностью ядерных данных. Если взять за основу при расчете реакторных характеристик пессимистические оценки, т.е. уменьшение коэффициента воспроизводства на 0,1 и увеличение загрузки активной зоны на 8%, то расход урана за 30 лет в системе с тепловыми и быстрыми реакторами возрастет по сравнению с представленными на рис.4 цифрами не более чем на 8-15%.

5. ЭКОНОМИЧЕСКИЕ КРИТЕРИИ ЭФФЕКТИВНОСТИ ИСПОЛЬЗОВАНИЯ ЯДЕРНОГО ГОРЮЧЕГО

Как отмечалось выше, экономические критерии должны учитывать фактор ограниченности ресурсов дешевого природного урана, приводящий к росту цен на уран со временем. При этом характеристики реакторов сказываются на темпах потребления урана и динамике его цены, которые оказывают обратное влияние на экономические показатели реакторов.

При определении динамики цен на уран здесь не будет учитываться неизбежный "нормальный" прогресс в технологии, ведущий к снижению затрат, так как при оптимизации и сравнениях важны не абсолютные, а относительные величины, которые более стабильны.

Если $G(t)\,dt$ – расход природного урана за время dt, $\mathcal{G}(C)$ – ресурсы урана, затраты на который не превышают C (долл/кг), то зависимость $C(t)$ может быть получена из выражения

$$\mathcal{G}[C(t)] = \int_0^t G(t)\,dt \tag{6}$$

При оценке экономической эффективности будем исходить из суммарных, приведенных к "настоящему моменту" $t = 0$ затрат на природный уран:

$$З = \int_0^\infty dt e^{-pt} G(t)\,C(t) \tag{7}$$

где p – процент дисконтирования.

Варьируя выражения (6) и (7), можно найти, что при изменении расхода урана на $\delta G(t)$ изменение суммарных затрат составит

$$\delta З = \int_0^\infty dt e^{-pt} \delta G(t)\,\tilde{C}(t) \tag{8}$$

где

$$\tilde{C}(t) = C(t) + \int_t^\infty dt' e^{-p(t'-t)} \frac{dC}{dt} \tag{9}$$

Величина

$$\tilde{C}(t) = e^{pt} \frac{\delta З}{\delta G(t)} \tag{10}$$

и должна быть взята в качестве цены урана в условиях ограниченных ресурсов. Она учитывает как затраты на данный килограмм урана, так и увеличение затрат в будущем при израсходовании дополнительного килограмма урана в момент t. Аналогично цена плутония

$$\tilde{C}_{Pu}(t) = e^{pt} \frac{\delta З}{\delta G_{Pu}(t)} = \int_t^\infty dt' e^{-p(t'-t)} C(t') \frac{\delta G_U}{\delta G_{Pu}} \approx \alpha\gamma\,C(t) \tag{11}$$

где α – относительная эффективность ^{239}Pu и ^{235}U в реакторах;

$\gamma = \dfrac{1 - \frac{x_0}{x}}{0{,}0071 - x_0} \approx 200$; x_0 – концентрация ^{235}U в отвале; x – концентрация ^{235}U в обогащенном горючем. Здесь под $\delta З$ понимается экономия в затра-

тах на уран при введении дополнительно 1 кг плутония в момент t. Величины $\delta З$ и $\widetilde{C}_{Pu}$, конечно, зависят от того, каким образом используется плутоний (например, в тепловых или быстрых реакторах), и поэтому однозначное их определение требует рассмотрения конкретной системы. Подчеркнем лишь, что $\widetilde{C}_{Pu}(t)$ определяет лишь одну составляющую цены плутония, связанную с потреблением природного урана.

При этом приведенные затраты на природный уран для реактора i-го типа окажутся равными

$$з = e^{pt}\frac{\delta З}{\delta N_i} = \gamma\left[q_i\widetilde{C}(t) + q_i\int_t^\infty dt' e^{-p(t'-t)}\widetilde{C}(t') - r_i\int_t^\infty dt' e^{-p(t'-t)}C_{Pu}(t')\right] \tag{12}$$

где g_i(кг/МВт); g_i (кг/МВт·год); r_i (кг/МВт·год) – удельные расходы горючего (^{235}U или ^{239}Pu) на загрузку, подпитку реактора и производство избыточного плутония соответственно; δN_i – прирост мощности. При ограниченных ресурсах стоимость производства электроэнергии будет расти со временем одновременно с ценой урана. Аналогичный анализ показывает, что урановая составляющая стоимости 1 кВт·ч имеет вид

$$S = \left(p\widetilde{C} - \frac{d\widetilde{C}}{dt}\right)\gamma g + \gamma q\widetilde{C} - r\widetilde{C}_{Pu} \tag{13}$$

что при интегрировании сводится к суммарным приведенным затратам [(см.выражение (12)].

Вычисленные таким образом затраты учитывают изменение цены урана вследствие ограниченности ресурсов и в этом смысле объединяют содержание экономических и натуральных критериев. Учет роста цены приводит к увеличению абсолютных величин затрат и к относительному уменьшению роли начальных вложений.

Что касается тепловых реакторов, то полученные формулы указывают на повышение их экономичности с вводом в действие быстрых реакторов из-за повышения цены плутония, а также на целесообразность сдвига оптимальной их конструкции и режимов работы в сторону увеличения производства плутония.

Сделаем грубую количественную оценку изменений экономических показателей реакторов, связанных с учетом фактора ограниченности урановых ресурсов.

В варианте развития ядерной энергетики[1] только на легководных реакторах затраты на природный уран растут за 30 лет с C = 20 до C = 60 долл/кг, так что в среднем $dC/dt \approx 1{,}3$ долл/кг·год.

При современных ценах приведенные затраты на уран для ЛВР составляют ~30 долл/кВт при p = 6% в год, а по формулам (7), (4) ~80 долл/кВт. В случае развития энергетики на ЛВР и быстрых реакторах при p = 10% в год эти цифры равны 20 и 40 долл/кВт, соответственно.

Затраты на горючее для быстрых реакторов оказываются меньшими по величине (в пределах ±10 долл/кВт) и слабее зависят от цены урана.

[1] В мировых масштабах.

Затраты, вычисленные по формулам (9), (12), (13), вместе с затратами на обогащение, изготовление, химическую переработку твэлов и т.п. могут служить критерием экономической эффективности при оптимизации реакторов в условиях ограниченных ресурсов урана.

ЛИТЕРАТУРА

[1] ЛЕЙПУНСКИЙ, А.И. и др., Ш Женевская конференция (1964) доклад №369.
[2] ЛЕЙПУНСКИЙ, А.И., Атомная энергия 28 (1970) 297.
[3] НОВОЖИЛОВ, А.И. и др., Доклад на симпозиуме СЭВ "Состояние и перспективы работ по созданию АЭС с реакторами на быстрых нейтронах" (Обнинск, 1967).
[4] БОБРОВ, С.Б. и др., В сб. "Физика ядерных реакторов". Вып.2. Атомиздат, М., 1970, стр.121.
[5] БОБРОВ, С.Б. и др., Там же, стр.129.
[6] ЗАРИЦКИЙ, С.М., МУРОГОВ, В.М., ШМЕЛЕВ, А.Н., "Бюллетень по делам изобретений и открытий", №15 (1970).
[7] КАШУТИН, А.А. и др., Доклад на симпозиуме СЭВ "Состояние и перспективы работ по созданию АЭС с реакторами на быстрых нейтронах" (Обнинск, 1967).
[8] ХРОМОВ, В.В. и др., В сб. "Физика ядерных реакторов". Вып.1. Атомиздат, М., 1970, стр.159.
[9] ХРОМОВ, В.В. и др., Там же, вып.2, стр.3.
[10] КУЗЬМИН, А.М., Там же, вып.1, стр.92.
[11] ФЕЙНБЕРГ, С.М., Атомная энергия 25 (1968) 363.
[12] АБАГЯН, Л.П. и др., Система многогрупповых констант для расчета энергетических быстрых реакторов. Атомиздат, М., 1964.
[13] ВЕСЕЛОВА, Г.М. и др., IAEA-CN-26/75 (1970).
[14] АБАГЯН, Л.П. и др., IAEA-CN-26/80 (1970).
[15] JAMES, G., SCHOMDERG, M., AERE-M-2157 (Harwell, 1969).
[16] GREEBLER, P. et al., Report 2nd International Conference on Nuclear Data for Reactors (Helsinki, 1970).

PLUTONIUM UTILIZATION IN THERMAL AND FAST REACTORS

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Abstract–Résumé–Аннотация–Resumen

PLUTONIUM UTILIZATION IN THERMAL AND FAST REACTORS.

Large quantities of plutonium are made available in thermal reactor operation, thus constituting a major alternative source of fissile material. The advantages of using plutonium as an alternative to enriched uranium and of improving exploitation of the source material are incentives to recycle plutonium in thermal reactors. This assumes greater importance in the long term in connection with the advent of commercial fast reactors. In Italy, the Government Agency for Power Production, ENEL, and that for Nuclear Research, CNEN, tackled the problems associated with plutonium utilization by means of programs that can be considered complementary. In 1966, ENEL, in co-operation with EURATOM, launched an extensive research program to establish the feasibility of recycling plutonium in thermal reactors by means of UO_2 - PuO_2 fuel assemblies. These assemblies were manufactured to standard fabrication techniques and were loaded in one of the ENEL reactors to verify their performance under actual operating conditions. The behaviour of these assemblies in normal operation, combined with the results of criticality experiments and gamma scanning, confirmed the validity of this approach. In the same year, CNEN launched the Plutonium Program and the Fast Reactor Program. The former aimed at acquiring know-how in the manufacture of ceramic-grade plutonium fuel in a specially built laboratory. As part of the development work, an extensive series of irradiation experiments on plutonium fuel was performed in prototype reactors. The Fast Reactor Program included research and development activities concerning fuel for fast reactors with a view to obtaining the necessary know-how to fabricate fuel assemblies capable of operating reliably at high specific power and high burn-ups.

UTILISATION DU PLUTONIUM DANS LES REACTEURS THERMIQUES ET DANS LES REACTEURS RAPIDES.

De grandes quantités de plutonium sont produites par l'exploitation des réacteurs thermiques, qui représentent ainsi une source importante de matières fissiles. Les avantages qui découlent à la fois de l'utilisation du plutonium à la place de l'uranium enrichi et d'une meilleure exploitation de l'uranium lui-même encouragent à recycler le plutonium dans les réacteurs thermiques. A long terme, le problème acquiert une importance majeure, compte tenu de la venue des réacteurs rapides sur le marché. En Italie, l'ENEL et le CNEN se sont attachés à résoudre les problèmes liés à l'utilisation du plutonium dans des programmes que l'on peut considérer complémentaires. En 1966, l'ENEL, en collaboration avec EURATOM, a entrepris un vaste programme de recherche afin de déterminer la possibilité technique de recycler le plutonium dans les réacteurs thermiques sous forme d'éléments combustibles en oxyde mixte UO_2-PuO_2. Ces éléments, fabriqués selon les techniques habituelles, ont été chargés dans un des réacteurs de l'ENEL, pour en vérifier la tenue en conditions réelles d'utilisation. Le comportement de ces éléments en exploitation normale, ainsi que les résultats des expériences de criticité et d'analyse gamma ont confirmé la validité de ces recherches. La même année, le CNEN a entrepris le programme plutonium et le programme réacteurs rapides. Le premier visait à acquérir l'expérience en matière de fabrication de combustible céramique au plutonium, dans un laboratoire spécialement construit à cet effet. Dans la phase préparatoire, une longue série d'expériences d'irradiation sur le combustible au plutonium a été exécutée dans des réacteurs prototypes. Le programme de réacteurs rapides comprend des études et des recherches sur le combustible destiné aux réacteurs rapides, afin d'acquérir les connaissances nécessaires à la fabrication d'éléments combustibles offrant une grande sécurité d'exploitation à une puissance spécifique et des taux de combustion élevés.

ИСПОЛЬЗОВАНИЕ ПЛУТОНИЯ В ТЕПЛОВЫХ И БЫСТРЫХ РЕАКТОРАХ.

При работе реакторов на тепловых нейтронах накапливаются большие количества плутония, которые могут служить основным альтернативным источником делящегося материала. Преимущества, связанные с применением плутония вместо обогащенного урана и улуч-

шеннем использования природных ресурсов, побуждают использовать в реакторах на тепловых нейтронах плутониевый топливный цикл. Это приобретает большое значение в связи со строительством в будущем промышленных реакторов на быстрых нейтронах. В Италии Национальное энергетическое управление и Национальный комитет по ядерной энергии решали проблемы использования плутония по программам, которые можно рассматривать как дополняющие друг друга. В 1966 году Национальное энергетическое управление совместно с Евратомом начало осуществлять обширную программу исследований с целью выяснения возможности использования плутониевого цикла в реакторах на тепловых нейтронах с применением сборок, топливом в которых служат UO_2 - PuO_2. Эти сборки были изготовлены по стандартной заводской технологии и поставлены в один из реакторов Национального энергетического управления для проверки их характеристик в реальных рабочих условиях. Результаты испытаний этих сборок в нормальных условиях, а также результаты критических экспериментов и гамма-сканирования подтвердили правильность такого подхода. В этом же году Национальный комитет по ядерной энергии начал исследования по программе изучения плутониевого цикла и по программе разработки реакторов на быстрых нейтронах. Первая касается получения сведений относительно керамического плутониевого топлива в специально построенной лаборатории. В качестве части исследовательской работы была выполнена обширная серия экспериментов по облучению плутониевого топлива в прототипных реакторах. Программа по реакторам на быстрых нейтронах включала исследования и разработки, касающиеся топлива для реакторов на быстрых нейтронах, с целью получения необходимой информации по изготовлению тепловыделяющих элементов, способных работать при высокой удельной мощности и больших глубинах выгорания.

UTILIZACION DEL PLUTONIO EN REACTORES TERMICOS Y RAPIDOS.

En el funcionamiento de los reactores térmicos se producen grandes cantidades de plutonio, constituyendo así una fuente adicional importante de material fisionable. Las ventajas de usar plutonio como alternativa del uranio enriquecido y de explotar más eficientemente el material de fuente son incentivos para reciclar el plutonio que se produce en los reactores térmicos. Esta cuestión reviste, a largo plazo, la máxima importancia a cuenta del advenimiento de los reactores rápidos comerciales. En Italia, la Agencia Estatal de Producción de Energía, ENEL y la de Investigación Nuclear, CNEN, atacaron los problemas referentes a la utilización del plutonio mediante programas que pueden considerarse complementarios el uno del otro. En 1966, el ENEL, en cooperación con la EURATOM, puso en marcha un extenso programa de investigación para establecer las posibilidades del reciclado del plutonio en los reactores térmicos a base de elementos combustibles de UO_2-PuO_2. Estos elementos se fabricaron con técnicas clásicas y fueron cargados en uno de los reactores del ENEL para comprobar su actuación en condiciones de funcionamiento real. El comportamiento de estos elementos en operación normal, junto con los experimentos de criticidad y la exploración gamma confirmaron la validez de este procedimiento. En el mismo año, el CNEN lanzó el Programa del Plutonio y el Programa de Reactores Rápidos. El primero pretendía adquirir experiencia en la manufactura de elementos combustibles de plutonio en forma cerámica; con este fin, se construyó un laboratorio especial. Como parte del trabajo de desarrollo se llevó a cabo una extensa serie de experimentos de irradiación sobre elementos combustibles de plutonio en reactores prototipo. El Programa de Reactores Rápidos incluía la investigación y desarrollo del combustible para reactores rápidos con la intención de obtener la experiencia necesaria para fabricar elementos combustibles dignos de confianza, en funcionamiento a alto grado de quemado y elevada potencia específica.

1. INCENTIVES TO UTILIZE PLUTONIUM IN POWER REACTORS

The large quantities of plutonium currently available from reactors in operation, and which will grow significantly in future as the nuclear installed capacity increases, are a major alternative source of fissile material [1-3]. One possible short-term utilization of plutonium is in thermal reactors, which, among other things, would avoid heavy financial outlay pending the advent of fast reactors. In fact, for a fissile plutonium value of, say, US $7/g, the credit on fuel cost for a 1000-MW(e) enriched-uranium reactor would be over a million dollars a year.

Apart from economic considerations, the incentive to recycle plutonium in thermal reactors lies in the possibility of using this material as an alternative to enriched uranium, which, on the one hand, would afford a certain independence in the procurement of fissile material and, on the

TABLE I. CHARACTERISTIC PARAMETERS OF SOME NUCLEAR REACTORS

Reactor	Initial specific inventory of fissile material (kg/MW(e))	Initial specific inventory of natural uranium (kg/MW(e))	Fissile material consumption [a] (kg/MW(e)-yr)	Natural uranium consumption (kg/MW(e)-yr)	Natural uranium consumption [a] (kg/MW(e)-yr)
Garigliano (BWR)	5.8	1020	0.60	170	125
Latina (Magnox)	9.0	1260	0.45	350	190
Trino V. (PWR)	5.0	915	0.64	180	130
Present generation:					
BWR	3.0	530	0.52	140	105
PWR	2.5	445	0.58	155	120
AGR	2.7	450	0.59	145	120
CANDU	1.0	150	0.33	130	60
Future generation:					
FBR	1.6-2.5	40-60	-(0.25-0.35)	-	1.5-3

[a] Inclusive of the energy potential of the recovered fissile plutonium

other, improve exploitation of the source material. Of course, in a mixed system of thermal and fast reactors, these advantages would be greatest if the plutonium were used in fast reactors only.

With regard to exploitation of source material, Table I gives some of the characteristic parameters [4] of the three ENEL reactors now in operation and of other reactors of the present and future generations. From columns 4 and 5 it can be seen that:

(a) in a given reactor type, the consumption of natural uranium per unit of electricity generated decreases as fuel burn-up increases, owing to the larger amount of plutonium burnt in situ;
(b) in proven reactor types, the fraction of potential energy of natural uranium actually utilized is about 1%;
(c) since the exploitation of source material in the reactors of the present generation is relatively small, the various efforts aimed at recycling plutonium in thermal reactors and, to an even greater extent, at developing advanced converter reactors as well as fast reactors are amply justified.

2. ITALIAN PROGRAMS FOR PLUTONIUM UTILIZATION

The problems associated with plutonium utilization were tackled in Italy by both CNEN and ENEL, and the related research programs may be considered complementary. ENEL carefully investigated the feasibility

of plutonium recycling by loading UO_2-PuO_2 assemblies manufactured with standard fabrication techniques (sintered pellets) in one of its thermal reactors and directly verifying their performance. Simultaneously, CNEN devoted its efforts to subsidiary aspects of UO_2-PuO_2 fuel technology, such as physics, thermohydraulic and mechanical design, and fabrication methods. The latter included advanced, as well as standard, technologies, such as those for preparation and vibrocompaction of dense powders in fuels for thermal and fast reactors. At present, CNEN is carrying out irradiation experiments in prototype reactors.

2.1. CNEN programs

The CNEN activities concerning plutonium fuel are linked with the Plutonium Program, the Fast Reactor Program, the Eurex Fuel Reprocessing Program, the Industrial Chemistry Laboratory, the Reactor Physics Laboratory, and the Health Physics Service and the Medical Service of the Nuclear Research Centre at Casaccia, near Rome.

The Plutonium Program is aimed mainly at acquiring know-how and developing the equipment necessary to manufacture ceramic-grade plutonium fuel within the shortest possible time.

The Fast Reactor Program sponsors, among other activities, long- and short-term research and development of fuel for future fast reactors to obtain the know-how to fabricate fuel assemblies capable of operating reliably at high specific power and high burn-up. For this purpose, the PEC (Prova Elementi di Combustibile, i.e. Fuel Assembly Testing) Reactor will be built for development of a fast reactor fuel. Much of the work, especially that concerning experimental tests, is at present devoted to fuel assemblies for the first PEC cores, the fuel of which will initially be enriched uranium and subsequently UO_2-PuO_2.

The Eurex Program includes a pilot plant built at Saluggia, which is now being operated for experimental reprocessing of irradiated fuel and recovery of both uranium and plutonium. The plant is equipped with a number of lead cells and gloveboxes, and is capable of receiving spent fuel, extracting the plutonium content, and converting it from nitric solution into oxide, i.e. ready for fuel preparation.

The Industrial Chemistry Laboratory of the Casaccia Centre is engaged mainly in physical chemistry work on aqueous plutonium solutions, besides performing supporting work in the analytical chemistry of plutonium compounds.

The Reactor Physics Laboratory of the Casaccia Centre performed the neutronic design of UO_2-PuO_2 fuel in co-operation with the Battelle Northwest Laboratory, within the framework of the USA-Italy bilateral agreement. The co-operative program included the following three phases:

(a) Follow-up at Argonne of the performance of the EBWR plutonium core followed by post-irradiation measurements at Hanford. This permitted a systematic check of the computer codes describing the operational behaviour of plutonium fuel as a function of burn-up.
(b) A series of criticality, power distribution and control rod worth measurements in the PRCF reactor at Hanford, in which General Electric personnel also participated, particularly for the study of hafnium control rods.

(c) A series of power distribution, spectral index and reactivity measurements with and without water gap and with Gd-poisoned fuel cladding tubes, shrouds and control rods in the RITMO reactor at Casaccia, on UO_2-PuO_2 fuel rods supplied by the USAEC (since the Casaccia plutonium laboratory was under construction at the time). Most of the above results proved useful in planning the experiments in the Garigliano reactor, in which CNEN also participated.

The Health Physics Service at the Casaccia Centre performs routine surveillance on plutonium-exposed personnel, while the Medical Service of the same Centre is responsible for first aid and for the biochemical determination, treatment and removal of plutonium.

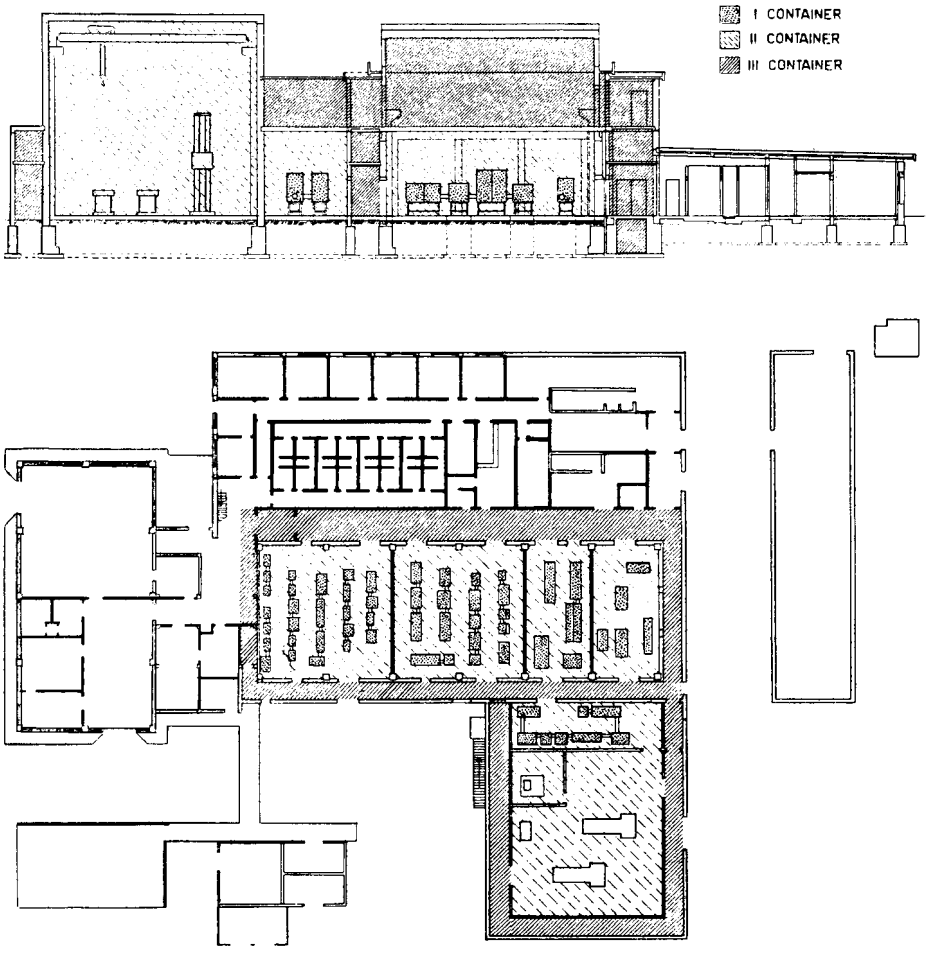


FIG. 1. Elevation plan and section of the Plutonium Laboratory, showing the three containment systems.

2.1.1. Laboratory for the development of plutonium fuel technology [5]

The Plutonium Laboratory covers an area of approximately 2400 m^2, of which about 1400 m^2 comprise the alpha laboratories for work on ceramic materials containing plutonium (Fig. 1).

While the design and construction of the plutonium laboratory at Casaccia were under way, an alpha laboratory was rented in Belgium to start the plutonium research work, to train personnel and to ascertain the possibility of adopting the sol-gel method, based on the CNEN process, and the soluble-salt hydrolysis method, based on the SNAM process, for the preparation of mixed oxides.

CNEN's plutonium activities at Mol ended in the middle of 1968, when the new laboratory was ready at the Casaccia Centre. At that time, all development activities were transferred to Italy. The first plutonium batch was introduced into the gloveboxes in November 1968.

This laboratory is used at present to try out various techniques for the fabrication of nuclear plutonium fuel, and in particular to carry out:

(a) technological research into various methods of plutonium fuel preparation for both thermal and fast reactors;
(b) analyses necessary for examination of the intermediate fabrication products as well as the final products;
(c) subsidiary research on plutonium-bearing materials (studies on various compounds, sintered ceramics, etc.).

The total expenditure incurred for the Plutonium Laboratory was about two million US dollars for special equipment and tools, and about half a million for civil engineering and conventional facilities. Although the plant is not intended to meet the requirements for industrial production, its capacity varies between 10 and 80 kg of ceramic material per day (in one shift), which is typical of large pilot plants, and it will provide useful information for the construction of any future production plants that might become necessary.

2.1.2. Irradiation experiments

The irradiation experiments on plutonium fuel produced by conventional pelletizing techniques and by the sol-gel method were aimed at determining its behaviour with regard to the particular design characteristics required for use in both thermal and fast reactors. Table II shows the irradiation experiments performed or in progress.

In the development of fast reactor fuel, the irradiation tests included experiments to investigate the release of gaseous fission products to the primary circuit (VENCA series), experiments to compare the behaviour of UO_2 and UO_2-PuO_2 fuel assemblies fabricated with different techniques (GIANO series), and sub-assembly experiments in a fast neutron flux (DFR series).

In the development of thermal reactor fuel, short-term experiments were carried out on small samples in the Swedish R-2 reactor at Studsvik. Additional long-term experiments on fuel rods are being carried out at Halden and Saluggia. Two test sections, loaded with sol-gel material,

TABLE II. SUMMARY TABLE OF IRRADIATION TESTS IN PROGRESS ON MIXED-OXIDE FUEL

Test Code name	Reactor (Location)	Burn-up as of April 1971 (Target), MWd/tonne	No. of pins	Fuel grams; form
Venca G-2	RS1 (Saluggia)	6200	1	32 g Pu; vibrocompacted
Venca 6, 7, 8	Siloé (Grenoble)	about 2000	1 each	19 g Pu; vibrocompacted
Giano-1	RS1 (Saluggia)	15 000	2	7 g ^{235}U + 6 g Pu; vibrocompacted
Giano 2, 3, 4	RS1 (Saluggia)	Up to 496 max.	6	4 g ^{235}U + 14 g Pu; vibrocompacted
IFA-124	HWBR (Halden)	400	2	23 g Pu; vibrocompacted
IFA-170	HWBR (Halden)	10 000	8	81 g Pu; vibrocompacted
A.P.-1	HWBR (Agesta)	5000 (15 000)	16	1440 g ^{235}U + 800 g Pu; pelletized
K.P.-1	BWR (Kahl)	Just started 25 000	36	About 1630 g Pu; pelletized
S.T. -43 to 54	R-2 (Studsvik)	Negligible	12	Pelletized
S.T. -55 to 66	R-2 (Studsvik)	Negligible	12	Vibrocompacted
Cyrsum PPCR	Siloé (Grenoble)	6500 (50 000)	1	6 g Pu; pelletized
Cyrsum PPTC	Siloé (Grenoble)	6500	1	6 g Pu; pelletized
DFR-2	DFR (Dounreay)	53 631	1	71 g ^{235}U + 13 g Pu; vibrocompacted

were irradiated in the Norwegian HWBR at Halden (Fig. 2). The non-instrumented section was removed after two months (at 500 MWd/tonne) because of a suspected failure which was not subsequently confirmed by post-irradiation examinations; the other fully instrumented section is still being irradiated.

To integrate these experiments based on short samples or a few rods, a program was launched to experiment on fuel prototypes. A normal 6 × 6 fuel assembly was fabricated (1600 mm long) for insertion in the boiling water reactor at Kahl, Federal Republic of Germany. A similar irradiation experiment is under way on a 4 × 4 fuel assembly (with four plutonium rods and twelve uranium rods) in the Swedish BWR at Ågesta.

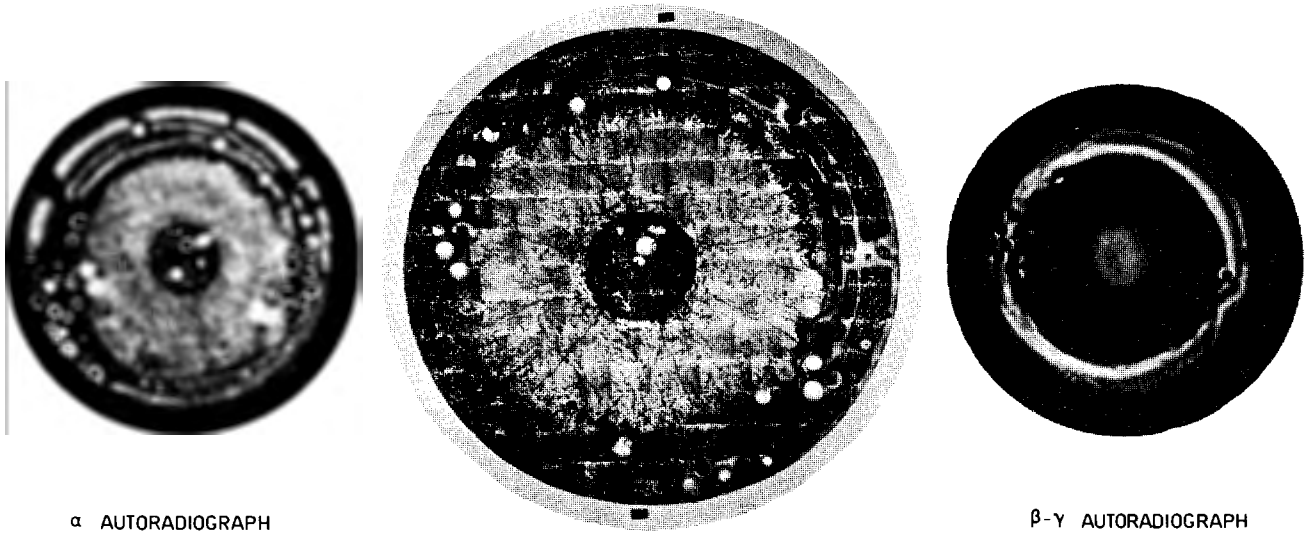


FIG. 2. Photo mosaic of a section in the region of maximum linear power rating of a UO_2-1.8 wt% PuO_2 experimental pin (IFA-124) containing sol-gel fuel.

2.2. ENEL program

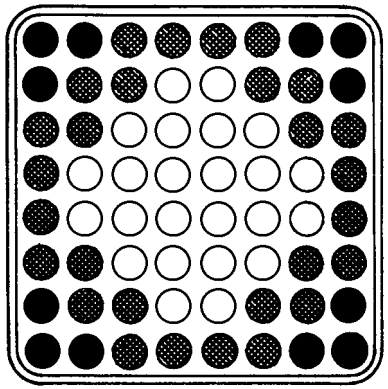
2.2.1. Plutonium recycle demonstration program in the Garigliano reactor

In 1966, after an exhaustive investigation on the utilization of the plutonium produced in its stations, ENEL launched an extensive program on plutonium recycling in thermal reactors in co-operation with EURATOM. The program was implemented through a number of phases, for each of which several choices had to be made from various solutions, all of which were often technically valid. These alternatives, and the related choices, are briefly described below:

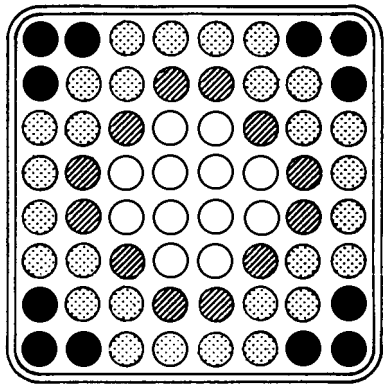
(a) Of the two reactors available – a BWR (Garigliano) and a PWR (Trino) – ENEL chose the former for several reasons, the main one being that at that time the Garigliano reactor had greater unutilized design margins [6].
(b) The number of mixed UO_2-PuO_2 prototype fuel assemblies (about 600 rods) was chosen to ensure statistical representation of the metallurgical behaviour in the reactor and to enable experiments on neutronics to be performed.
(c) Preference was given to pellet-type fuel, although other forms, such as vibrocompacted powder, were also used in small amounts.
(d) The fuel assemblies were of both the standard type (Fig. 3(a)), containing only plutonium rods, and the mixed type (Fig. 3(b)) containing plutonium rods at the centre and ^{235}U-enriched rods at the periphery.
(e) Rather than optimize the nuclear design of the prototype plutonium assembly, it was considered preferable to maintain the same lattice, the same reactivity lifetime and a local power peak within the same limits as the uranium assemblies.
(f) As regards the experiments to be conducted on the fresh plutonium assemblies, the choice fell on reactivity measurements through the replacement technique and on local power distribution measurements by means of rod gamma-scanning.
(g) A checkerboard loading pattern was selected in which the plutonium assemblies were scattered instead of being concentrated in one core region.
(h) The experiments on the plutonium assemblies at the end of the first cycle at a burn-up of about 7000 MWd/tonne were chiefly macroscopic power distribution measurements by means of assembly gamma-scanning and local power distribution measurements by means of rod gamma-scanning.

In the summer of 1968, open-vessel minimum critical experiments (item (f)) were performed on ensembles formed of enriched-uranium assemblies and prototype plutonium assemblies. The purpose of these experiments was to ascertain the reliability of the calculation methods used in the nuclear design of plutonium assemblies; in addition, since they were performed on real assemblies, they supplemented the data obtained from other experiments on critical facilities. It was thus possible to verify the reliability of the calculations for the determination of criticality conditions and power distribution in mixed lattices and for the evaluation of the influence of water gaps and contiguity of plutonium and enriched-uranium assemblies (power sharing).

For this purpose, a sufficiently large number of assemblies was discharged from the reactor to clear a core quadrant in which to form the critical ensemble with a gap over 60 cm wide to segregate the ensemble



(a)



(b)

Rods containing 2.85 wt% of fissile plutonium

Rods containing 1.40 wt% of fissile plutonium

Rods containing 0.74 wt% of fissile plutonium

UO_2-PuO_2 rods containing natural uranium and 2.89% of fissile plutonium

UO_2-PuO_2 rods containing natural uranium and 1.80% of fissile plutonium

UO_2 rods enriched to 2.41% in U-235

UO_2 rods enriched to 1.83% in U-235

FIG. 3. Distribution of plutonium content in the rods of: (a) a standard type fuel assembly; (b) a mixed type fuel assembly.

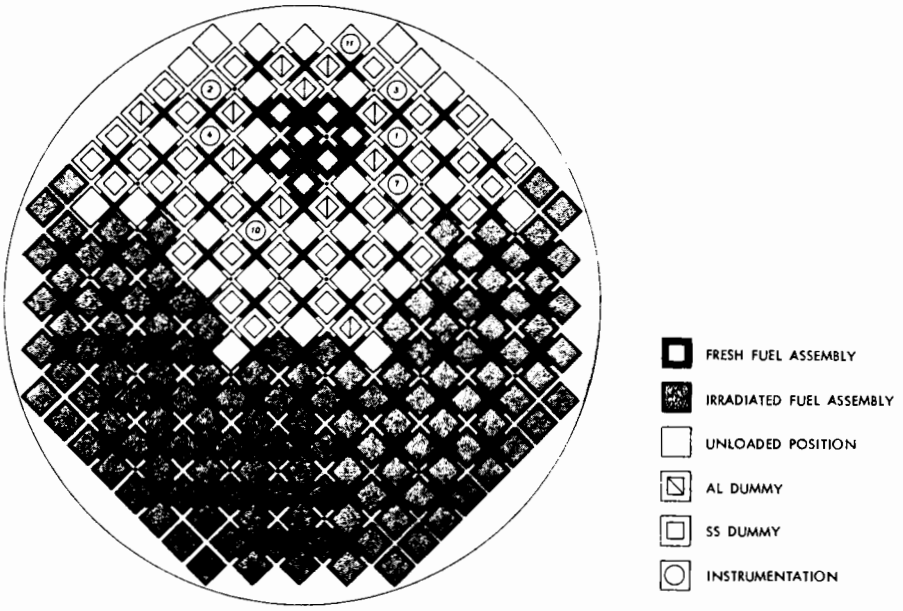


FIG. 4. Location of the assemblies of the critical configuration in the vessel.

from the rest of the core (Fig. 4). To obtain meaningful information, the experiments were started with a critical ensemble of all fresh enriched-uranium (2.3% ^{235}U) assemblies; subsequently, a uranium assembly was replaced with a plutonium assembly and the replacement was repeated for all the positions in the ensemble, one at a time, so as to obtain different configurations. In this manner, the reactivity variations associated with the different types and positions of the substituting elements could be assessed fairly accurately.

For the power distribution measurements, a 3×3 ensemble was formed with four enriched-uranium and four mixed-type assemblies and one standard plutonium assembly at the centre. The configuration presented an octant symmetry, so that it was possible to measure the power distribution by removing only three assemblies, one of each type. The three assemblies, which had been irradiated at 10^9 nv for an hour, were transferred to the fresh fuel pool for disassembly, and gamma scanning was then performed on each rod.

The values predicted for criticality were within 0.5% of the measured value; the standard deviation between theoretical and experimental values of the power distribution was less than 2%.

In the meanwhile, ENEL had made arrangements with the Laboratories of the Common Research Centre at Karlsruhe for the chemical and isotopic analyses of a few rods of a uranium assembly that had been irradiated to 9500 MWd/tonne in the Garigliano reactor; here again, the purpose was to verify the calculation methods in relation to burn-up.

The purpose of the gamma scans mentioned in item (h) above was to verify the validity and degree of precision of the simplified three-dimension codes normally used by ENEL to calculate the power distribution in the Garigliano reactor. In order to have meaningful results, fifty-two assemblies – one fourth of the core – were all subjected to gamma scanning on the four corners at eight equally spaced points along the assembly length. Eight of these fifty-two were plutonium assemblies so that their radial and axial power distributions in a mixed core could be established. The standard deviation between the calculated and experimental power distribution values was 2.5%, which proved the validity of the FLARE code, as adapted by ENEL, as a tool for the design and operational follow-up, even in the case of mixed cores.

To check the local power distribution in the plutonium assemblies, one assembly was taken apart, and the power distribution was measured on each rod at two different elevations. The experimental results were quite close to the values obtained with the code in the x, y geometry used by ENEL for the nuclear design of the plutonium assemblies in respect of the variation in local power shape with irradiation.

At the end of the next cycles, in addition to gamma scanning for the determination of the power distributions, long-term reactivity measurements and thorough inspections will be performed to determine the mechanical performance of the prototype assemblies. Consideration is being given to the desirability of performing isotopic and metallurgical analyses on partially irradiated rods of the prototype assemblies.

2.2.2. Program for industrial utilization of plutonium

Because of the quantities of plutonium that are becoming available, ENEL is giving serious consideration to plutonium recycling in its water reactors, subject, on the one hand, to the economic convenience of such recycling – which is mainly conditioned by fuel fabrication costs – and, on the other, to the plutonium requirements of any fast reactor prototypes that Italy may be interested in.

At any rate, if plutonium recycling in one or more thermal reactors in an integrated system of nuclear stations results in an industrial value of plutonium that justifies this form of utilization, there will be a number of alternatives to choose from, such as:

(a) open cycle or closed cycle;
(b) partial load or total reload of plutonium, with or without burnable poisons;
(c) fuel forms in relation to the fabrication costs;
(d) all-plutonium or mixed-oxide assemblies;
(e) nuclear design either optimized for plutonium or not;
(f) use of natural uranium or recycle uranium (depleted or slightly enriched).

3. ECONOMICS OF PLUTONIUM UTILIZATION IN POWER REACTORS

The advantage of recycling plutonium in thermal reactors pending commercial operation of fast reactors depends on many factors. One

way to assess this advantage is provided by the so-called "indifference method". In an economy of thermal reactors only, in which other applications of plutonium are not contemplated it becomes economically advantageous to recycle plutonium in a given reactor when this will result in a lower kWh cost than is obtainable with only uranium fuel and no credit for plutonium. In this case, it is "indifferent" whichever fissile material is used.

The insertion of fast reactors in a system of thermal reactors – apart from any direct or indirect advantages associated with the better exploitation of the source material – is only economically profitable when it will lower the kWh cost of the whole system. In this case it is "indifferent" whether a uranium-fuelled thermal reactor or a plutonium-fuelled fast reactor is used, but the value of plutonium is higher than in a system of thermal reactors. Because of the higher value of plutonium in a mixed system of thermal and fast reactors and of the stocks required for the initial loads of fast reactors, it will be necessary to interrupt plutonium recycle far enough in advance unless, of course, excess plutonium is available [3].

REFERENCES

[1] SANI, L., "Considerations on Plutonium Utilization in Power Reactors", Plutonium Utilization (Rep. Panel Vienna, 1968) unpublished document IAEA 112, IAEA Vienna, (1969) 145.

[2] SANI, L., "Riciclo del plutonio nei reattori termici", FAST Nuclear Energy Days, Milan, December 1968.

[3] SANI, L., SCHILEO, G., "Italian programs for plutonium utilization", Foratom Symp. on Industrial Aspects of the Nuclear Fuel Cycle in Europe, Stockholm, September 1970.

[4] SANI, L., "Technical aspects of the fuel cycles of the three ENEL nuclear power stations", BNES Int. Conf., London, June 1967.

[5] SCHILEO, G., "Attività ed infrastrutture del CNEN per la riutilizzazione del plutonio", FAST Nuclear Energy Days, Milan, December 1968.

[6] ARIEMMA, A., BELELLI, U., SANI, L., "Assessment and experimental investigation of plutonium potential in ENEL water reactors", Plutonium as a Reactor Fuel (Proc. Symp. Brussels, 1967) IAEA, Vienna (1967) 795.

UNITED KINGDOM
A/CONF.49/P/496

THE TECHNICAL PROBLEMS AND ECONOMIC PROSPECTS ARISING FROM THE ALTERNATIVE METHODS OF USING PLUTONIUM IN THERMAL AND FAST BREEDER REACTOR PROGRAMS

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Abstract–Résumé–Аннотация–Resumen

THE TECHNICAL PROBLEMS AND ECONOMIC PROSPECTS ARISING FROM THE ALTERNATIVE METHODS OF USING PLUTONIUM IN THERMAL AND FAST BREEDER REACTOR PROGRAMS.

The methods available for calculating fuel requirements for recycling plutonium in thermal reactors are first described, comments are given on their likely accuracy and results presented from calculations on plutonium flows and isotopic concentration effects for a range of gas-cooled and water-cooled thermal reactors. The general conclusions are discussed and related to other practical aspects of the use of plutonium in thermal reactors, including fuel fabrication and reprocessing requirements, reactor design, operational and fuel management problems. Similar information is then presented on the use of plutonium in sodium cooled fast reactors for metal oxide and carbide fuels. The case is considered for initiating fast reactors with ^{235}U. Conclusions are drawn about the effects of plutonium isotopic compositions on the initial core and feed requirements for different reactor systems and the re-use of rejected material. The logistic and economic consequences of recycling plutonium and its value in thermal or fast reactors are assessed, taking the UK program as a particular example, and the important factors affecting alternative strategies are identified. The general conclusion is drawn that the policy of the early introduction of fast reactors being followed in the United Kingdom provides the most economic utilization of plutonium. Recycling of plutonium in thermal reactors is likely to be attractive only in countries where the introduction of fast reactors is likely to occur much later.

PROBLEMES TECHNIQUES ET PREVISIONS ECONOMIQUES QUE SOULEVE LE CHOIX DES METHODES D'UTILISATION DU PLUTONIUM DANS LES PROGRAMMES DE REACTEURS THERMIQUES ET DE REACTEURS A NEUTRONS RAPIDES.

Les méthodes dont on dispose pour calculer les quantités et les qualités de combustible nécessaires au recyclage du plutonium dans les réacteurs thermiques sont décrites, avec des observations sur la probabilité de leur exactitude, et les auteurs présentent les résultats des calculs concernant les flux de plutonium et les effets de la concentration isotopique pour une série de réacteurs thermiques refroidis à l'eau ou par un gaz. Les conclusions générales sont analysées et rapportées à d'autres aspects pratiques de l'utilisation du plutonium dans les réacteurs thermiques, comme la fabrication du combustible et son retraitement, les concepts de réacteurs et les problèmes d'exploitation et d'utilisation du combustible. On présente ensuite des renseignements analogues sur l'utilisation du plutonium sous forme d'oxyde ou de carbure dans les réacteurs à neutrons rapides refroidis au sodium. On considère la possibilité de démarrer des réacteurs rapides avec une charge de ^{235}U. On tire des conclusions quant aux effets de la composition isotopique du plutonium sur le cœur initial et sur les conditions nécessaires à l'alimentation de différentes filières de réacteurs ainsi que du réemploi des matières rejetées. Les auteurs déterminent les conséquences logistiques et économiques du recyclage du plutonium et son importance dans les réacteurs thermiques ou rapides en prenant le programme britannique comme exemple particulier. Ils dégagent les facteurs importants affectant l'un ou l'autre des procédés et tirent la conclusion générale que la construction accélérée de réacteurs à neutrons rapides procurerait le mode d'utilisation le plus économique du plutonium au Royaume-Uni. Le recyclage du plutonium dans les réacteurs thermiques intéressera vraisemblablement les pays où l'introduction des réacteurs à neutrons rapides sera plus tardive.

ТЕХНИЧЕСКИЕ И ЭКОНОМИЧЕСКИЕ ПРОБЛЕМЫ, ВОЗНИКАЮЩИЕ ПРИ АЛЬТЕРНАТИВНЫХ СПОСОБАХ ИСПОЛЬЗОВАНИЯ ПЛУТОНИЯ В ПРОГРАММАХ РАЗРАБОТКИ ТЕПЛОВЫХ И БЫСТРЫХ РЕАКТОРОВ-БРИДЕРОВ.

Впервые описываются методы расчета потребностей в топливе при повторном использовании плутония в тепловых реакторах, приводятся некоторые замечания о их возможной точности, а также результаты расчетов расхода плутония и эффектов изотопной концентрации для ряда тепловых реакторов с газообразным и водяным теплоносителем. Обсуждаются общие выводы, касающиеся других аспектов использования плутония в тепловых реакторах, включая требования к изготовлению и переработке топлива, конструкцию реактора, проблемы эксплуатации и перегрузки топлива. Дается аналогичная информация по использованию плутония в быстрых жидкометаллических реакторах с топливом из окиси металла и карбида. Рассматривается случай использования быстрых реакторов с топливом из урана-235 на ранней стадии. Приводятся данные по влиянию изотопного состава плутония на требования к первоначальной и последующим загрузкам активной зоны для различных реакторных систем и на повторное использование отработанного материала. Приводится качественная и количественная оценка логических и экономических последствий повторного цикла плутония в тепловых и быстрых реакторах на примере программы Соединенного Королевства. Определяются основные факторы, влияющие на выбор альтернативных стратегических направлений. В заключении делается вывод, что проводимая в Соединенном Королевстве политика строительства быстрых реакторов на ранней стадии обеспечивает наиболее экономичное использование плутония. Повторное использование плутония в тепловых реакторах, по-видимому, привлекательно только в странах, где строительство быстрых реакторов начнется на более поздней стадии.

PROBLEMAS TECNICOS Y PERSPECTIVAS ECONOMICAS RESULTANTES DE LOS METODOS ALTERNATIVOS DEL EMPLEO DEL PLUTONIO EN PROGRAMAS DE REACTORES TERMICOS Y REPRODUCTORES RAPIDOS.

Se describen primero los métodos disponibles para calcular las necesidades de combustible para la recirculación de plutonio en reactores térmicos y a continuación se hacen comentarios sobre su posible exactitud, presentándose a continuación resultados de los cálculos sobre el flujo de plutonio y los efectos de la concentración isotópica en un surtido de reactores térmicos refrigerados por gas y por agua. Se discuten las conclusiones generales, relacionándolas con otros aspectos prácticos del uso del plutonio en reactores térmicos, incluyendo la fabricación de combustible, la regeneración, diseño de reactores y problemas operativos y sobre distribución de combustible. Se presenta a continuación información análoga sobre el empleo del plutonio en reactores rápidos refrigerados con sodio para combustibles de óxido metálico y carburo. Se considera también la propuesta para la iniciación de reactores rápidos con ^{235}U. Se llega a ciertas conclusiones sobre los efectos de las composiciones isotópicas del plutonio y las necesidades del núcleo inicial en la alimentación para diferentes sistemas de reactores, así como el nuevo uso del material rechazado. Se evalúan también las consecuencias logísticas y económicas de la recirculación del plutonio y su valor en reactores térmicos o rápidos, tomando como ejemplo particular el programa en el Reino Unido. Se identifican además los factores importantes que afectan las posibilidades alternativas. Se saca la conclusión general de que la norma de la pronta introducción de reactores rápidos que se sigue actualmente en el Reino Unido, proporciona el empleo más económico del plutonio. La recirculación del plutonio en reactores térmicos podría ser atrayente solamente en países donde la introducción de reactores rápidos sólo podría ocurrir mucho más tarde.

This paper describes United Kingdom progress with technical and economic assessments of uses for plutonium from uranium fuelled reactors arising from improvements in theoretical techniques and experimental data.

Fuel cycle studies are reported with Pu replacing U235 for the Mark II Gas Cooled Reactor (AGR), the Mark III Gas Cooled Reactor (HTR), the Steam Generating Heavy Water Reactor (SGHWR) and the Pressurised Water Reactor (PWR); the associated operational and fuel plant problems are discussed. Use of Pu in each thermal reactor (possibly temporarily prior to use in fast reactors) is considered and equivalent information presented for using Pu entirely in fast reactors (in some cases with supplementary U235 initiation).

Reactor system analyses require consideration in the light of specific national programme, fuel logistic and economic factors. For illustration the consequences of alternative Pu applications in typical UK programmes are presented.

THERMAL REACTORS

Methods and data

UK fuel cycle studies use the classical separation of lattice cell and overall reactor events. For thermal reactors the WIMS code is used including common nuclear data, geometric options for different types of fuel and self-shielding effects. The WIMS method has flexibility for the pilot studies required to establish group structure. Treatment of the growth of fission product poisons must recognise changes in fission yield with fissile isotope composition and the americium isotopes, 241 and 243, with a fissile contribution from Am 242, must not be neglected; studies with a substantial content of higher isotopes show an end of life reactivity loss exceeding 3% $\Delta k/k$ due to americium.

The WIMS output can be combined with a reactor fuel management scheme using two or three-dimension diffusion theory calculations to follow the changes in power distribution, typically the ODYSSEUS code for GCRs, CALEB for SGHWRs, and JOSHUA for LWRs.

Basis for comparison

The thermal reactor results refer to utilisation of selected plutonium mixtures on a 'once-through' basis with typical U235-cycle fuel management schemes (Table I) replaced by Pu schemes which achieve the same irradiation. Loadings containing both U235 and Pu, and adjustment of irradiations and fuel management to optimise reactor performance, may show benefits not considered here.

On-load fuelling in gas cooled reactors (and 60 hour refuelling shutdowns each 120 full-power days for SGHWRs) enables power flattening by use of two zone enrichments. Annual refuelling, with a radial fuel move into a chequerboard inner zone, was assumed for the PWR. The Mark III GCR allows adjustment of the fuel to moderator ratio to permit Pu burning. Four plutonium stockpile isotopic compositions have been chosen, the first representing Magnox plutonium.

Plutonium utilisation - thermal reactors

The Pu fed and rejected has been computed (Table II), together with net fissile plutonium consumption and equivalence between initial plutonium and U235 atoms required to give the same reactivity lifetime.

The Mark II GCR is a satisfactory user of Pu, having low fissile consumption and favourable Pu/U235 equivalence, even with higher Pu240 content, suggesting adequacy for once-through or continuous Pu recycle. The SGHWR has a slightly higher fissile consumption and higher Pu/U235 equivalence; nevertheless it should still be satisfactory for Pu recycle.

TABLE I. TYPICAL U235 FEED THERMAL REACTOR DESIGNS

Reactor Type	Refuelling Scheme	Mean Feed Enrichment U235 Wt %	Discharge Irradiation MWd/tU	Thermal Efficiency %
Mk II GCR	On Load	2.36	18,000	42.5
Mk III GCR	On Load	5.10	60,000	42.5
SGHWR	9-batch Off Load	2.09	21,000	32.0
PWR	3-batch Off Load	3.28	33,000	33.0

TABLE II. COMPARISON OF PLUTONIUM UTILISATION IN DIFFERENT TYPES OF THERMAL AND FAST REACTORS

Plutonium Input Quality % weight	Reactor Type	Mean Fuel Burnup MWd/t h.a.[a]	Total Pu Feed to core % h.a.	Total Pu Reject from core % h.a.	Core Pu Output Quality % weight				Fissile Pu Produced in FR Blanket kg/MW(e) y	Net Fissile Pu Burnt in Reactor kg/MW(e) y	Fissile Pu to U235 Equivalence	Breeding Gain Excess Fissile Pu/Fission	
					Pu239	Pu240	Pu241	Pu242				Core	Total
	Mk.II GCR	18,000	1.715	1.181	40.9	41.4	11.7	6.0	-	0.410	0.90	-	-
Pu239 84.2	SGHWR	21,000	1.825	1.356	45.2	36.8	11.7	6.2	-	0.436	1.15	-	-
Pu240 13.6	PWR	33,000	3.605	2.747	47.2	31.7	16.0	5.1	-	0.462	1.21	-	-
Pu241 2.1	Mk.III GCR	120,000[b]	8.865	2.802	20.0	22.8	16.9	40.3	-	0.474	0.88	-	-
Pu242 0.1	Oxide FR	53,000	16.740	17.190	77.9	19.8	2.1	0.2	0.409	-0.213	-	-0.195	0.280
	Carbide FR	40,000	13.640	14.430	80.0	17.8	2.0	0.2	0.451	-0.369	-	-0.065	0.431
	Mk.II GCR	18,000	2.075	1.464	36.3	43.9	12.4	7.4	-	0.400	0.94	-	-
Pu239 69.7	SGHWR	21,000	2.231	1.668	40.8	39.0	12.6	7.6	-	0.423	1.21	-	-
Pu240 24.2	PWR	33,000	4.734	3.715	44.2	33.7	16.6	5.4	-	0.431	1.38	-	-
Pu241 5.1	Mk.III GCR	168,000[b]	17.250	6.593	25.5	23.0	24.5	27.0	-	0.492	1.14	-	-
Pu242 1.0	Oxide FR	53,000	18.250	18.650	66.3	28.1	4.4	1.2	0.409	-0.249	-	-0.178	0.297
	Carbide FR	40,000	14.870	15.630	68.4	26.3	4.2	1.1	0.451	-0.403	-	-0.051	0.445
	Mk.II GCR	18,000	2.093	1.466	33.8	42.2	12.9	11.1	-	0.393	0.92	-	-
Pu239 61.5	SGHWR	21,000	2.319	1.717	38.7	37.6	13.1	10.6	-	0.431	1.22	-	-
Pu240 24.6	PWR	33,000	5.065	3.931	42.4	32.5	17.5	7.6	-	0.444	1.43	-	-
Pu241 11.1	Mk.III GCR	208,000[b]	20.720	6.570	17.1	20.3	20.9	41.7	-	0.517	1.14	-	-
Pu242 2.8	Oxide FR	53,000	18.100	18.450	61.2	27.9	7.8	3.1	0.409	-0.261	-	-0.207	0.268
	Carbide FR	40,000	14.790	15.500	63.1	26.2	7.8	2.9	0.451	-0.411	-	-0.081	0.415
	Mk.II GCR	18,000	2.687	1.954	29.1	44.3	13.0	13.6	-	0.392	1.00	-	-
Pu239 50.2	SGHWR	21,000	3.090	2.342	32.8	40.1	13.5	12.7	-	0.425	1.38	-	-
Pu240 33.0	PWR	33,000	8.558	7.031	39.3	36.4	16.3	8.0	-	0.446	2.04	-	-
Pu241 11.0	Mk.III GCR	397,000[b]	48.700	13.570	10.9	20.9	21.5	46.7	-	0.549	1.04	-	-
Pu242 5.8	Oxide FR	53,000	20.290	20.570	51.4	34.6	8.2	5.8	0.409	-0.296	-	-0.180	0.295
	Carbide FR	55,000	16.580	17.230	53.2	33.1	8.1	5.6	0.451	-0.445	-	-0.055	0.441

[a] h.a. = heavy atoms.

[b] Constant heat output equivalent to 60,000 MWd/t in U235 enriched fuel.

For the PWR, however, even higher values of these parameters make it less satisfactory for recycle, though it is likely to be satisfactory for once-through applications. The fissile equivalence of the Mark III GCR is similar to that of the other thermal reactors, but it uses about two thirds of the Pu fed to it whereas the other designs use only one third; it is therefore more suitable for using Pu when fast reactors are not requiring it.

Operational implications

Fission and absorption cross sections for Pu are higher than for U and will increase new channel power peaking. For a Mark II GCR, fuel management changes and a temporary down-rating of the reactor might be required, but the equilibrium new channel age factor would be only a few % higher than for the U235 case. For an SGHWR, channel to channel peaking factors appear to be improved with Pu-fuelling at equilibrium and the void coefficient becomes slightly more negative; differential enrichment of a fuel bundle could usefully restrict the power peaking factor. In a PWR, pin power peaking may give difficulty where a Pu fuel assembly meets a U235 one but the channel to channel macroscopic effect is reduced, compensating the loss in the departure from nucleate boiling (DNB) ratio; differential enrichments may again help.

A safety review is needed for all Pu fuelled reactors as reactivity coefficients change and shut down rod worths are reduced. For water cooled systems the larger negative coolant density coefficient increases the required shutdown margin. For reactors where excess reactivity is held in control rods (in contrast to chemical shim) it may be necessary to compensate the loss in rod worth with a burnable poison.

Fuel implications

Plutonium fuel irradiated for the fast reactor programme in the Windscale AGR and the Winfrith SGHWR has indicated no special endurance problems.

Two aspects of fuel design influence costs of meeting the same specification as for uranium fuel. The specification must allow for extra Pu manufacturing difficulties e.g. tolerances in fuel density, pellet dimensions and inhomogeneity. The costs are far greater due to the high first cost of the plant and subsequent costs for plant cleaning, inspection, analysis and recycle of plutonium oxide, in all of which special operator protection is needed.

A balance is needed between the extra cost of fabricating several enrichments and the saving from a better core form factor. By restricting plutonium to one enrichment and a specific number of pins, and varying the enrichment level in the uranium pins, a form factor no worse than for uranium fuel might be achieved; if a poorer local form factor is acceptable, concentration of the same plutonium in fewer pins might be economic.

When Pu is recycled in thermal reactors the ratio of U to Pu fuel fabrication, largely because of the effect of scale of production,is about 4:1; the fabrication surcharge for the Pu fuel could be 100% of the uranium fuel fabrication cost. The surcharge could be substantially higher in the early years due to limited requirements or eventually due to the advent of fast reactors terminating Pu recycle.

The existing reprocessing plants at Windscale can, with some modification to operating procedures, reprocess plutonium enriched thermal reactor fuel. The problems are relatively small compared with those of plutonium fuel fabrication.

FAST REACTORS

Methods and data

Important new differential cross-section measurements have emerged from the UK experimental programme. The zero power integral measurement programme has used two types of assembly; one simulating aspects of the Prototype Fast Reactor (PFR) (e.g. Zebra 7 series) and the other for basic physics experiments,(e.g. Zebra 5 Doppler experiments, performed

TABLE III. FAST REACTORS WITH Pu, U235 AND MIXED Pu/U235 FUELLING

	Fissile Feed		Pu with depleted U	Pu with Enriched U 3.8%	Pu with Enriched U 7.2%	Enriched U 25.1% and 19.2%
Equilibrium core	U235	kg	36	388	774	2871
	Pu	kg	2905	2569	2179	204
Feed to core (80% load factor)	U235	kg/y	39	414	824	2876
U235 Reject (2% losses)						
-Total		kg/y	25	271	553	1920
-Core Mixture	U235/U	%	0.24	2.6	5.2	20.1 & 14.2
-Mixture with axial breeder	U235/U	%	0.32	1.5	2.8	8.6
Consumption Rate	U235	kg/y	14	143	271	956
Net Production Rate	Pu239(E)[a]	kg/y	139	241	339	746
Breeding Gain (Excess per fission)	Pu239(E)		0.193	0.168	0.143	0.052
Extra Production	Pu239(E)	kg/y	–	102	200	607
Extra Consumption	U235	kg/y	–	129	257	942
Time to convert to Pu fuelling (Inventory 4138 kg Pu239(E))		y	–	2.0	3.0	5.5

[a] Pu239(E) is the amount of Pu239 having the same reactivity worth as the Pu/U mixture.

in a heated loop with PFR type pins). In the Dounreay Fast Reactor (DFR) emphasis has been on determining alpha for Pu 239 and also for samples of U235, U238, Pu240, Pu241 and Pu242. In 1967 measurements of the Pu239 alpha at energies in the range 0.1 to 10 keV were reported double those previously assumed, and confirmed later at Harwell, in the USA and in Russia. An increase has therefore been made in the alpha value for Pu239 in the latest 35 group cross section set FD4, which raises the spectrum averaged value by about 30%. A second important change is the increase in capture cross section for U238 by about 30% over the range 1 to 20 keV, followed by a reduction of about 20% over the range 75 to 200 keV. The overall result has been an increase in plutonium feed concentration of about 6.5% and a reduction in breeding gain of about 0.08.

Comparison of oxide and carbide fuels

Table II gives Pu fuel cycles for 1200 MW(e) sodium cooled fast reactors with solid oxide and solid carbide pin fuels. A once-through continuous charge/discharge fuel cycle has been used to represent practical batch refuelling. Mean burnup was 5.6% (7.5% maximum) for oxide fuel and 4.2% (5.5% maximum) for carbide, giving the same fissions/cm^3; these are not the ultimate burnup for these fuels. The radial breeder was carbide.

The quality of Pu fed to fast reactors has little effect on their breeding gain and so Pu from any source can be used as input with similar effectiveness. The breeding gains imply linear doubling times of 20 to 25 years and 10 to 15 years for the oxide and carbide designs respectively. An economic comparison also depends on other factors, including fuel burnup and fabrication costs. In determining the fraction of the installation that can be fast reactors in the earlier years, the inventory is important; in this respect the two fuels are similar when out-of-pile requirements are allowed for. In later years the exponential doubling time is of greater significance and values of about 15 and 8 years, which are achievable for oxide and carbide respectively, have to be compared with the long term electricity demand growth.

Start up with uranium fuel

In a sodium cooled fast reactor about 20% more neutrons are produced per absorption in Pu239 than in U235, so Pu is the preferred fuel for breeding. However when insufficient plutonium is available to provide initial inventories for fast reactors in the numbers required, the choice will be between more thermal reactors or starting up some fast reactors with U235; plutonium produced in the latter can be used to convert them progressively to the preferred plutonium fuel.

Table III gives fuelling data for 1200 MW(e) reactors with Pu and U235 alternative enrichments. Maximum core fuel burnup of 7.5% heavy atoms has again been assumed and the breeder is carbide. Uranium and plutonium have been taken to be a uniform mixture within both core zones.

Excess plutonium produced with the U235 fuelled reactor is more than five times that from the Pu design but the breeding gain is much reduced and U235 must be continuously fed to the core. To provide the extra 750 kg of Pu in the U235 reactor there is an additional consumption of about 950 kg p.a. of U235 but more fast reactors become feasible; depending on the Pu/U235 starting mixture and the operating arrangements sufficient plutonium will have been produced in less than six years to allow conversion from U235 to entirely Pu fuelling.

Fuel requirements

Plutonium fuel fabrication cost data relies on manufacturing experience with PFR core fuel at Windscale and breeder fuel at Springfields; the latter operations are similar to those for the thermal reactor fuel. There is sufficient capacity to manufacture fuel for the 250 MW(e) prototype and at least 1300 MW(e) of further requirements. Development work is in hand to explore improved flow sheets to give greater flexibility for alternative fuel designs and better economy in larger fabrication plants achieving the economies of scale.

The heavy atom throughput quoted provides a comparison of fabrication and reprocessing expenditure. For the Mk II GCR on a uranium feed in support of fast reactors specific costs are about £40,000/t U but the larger number required to support an all thermal programme leads to a lower figure of £30,000/t U. Plutonium feed involves a surcharge of some £30,000/t U. For the fast reactor typical figures are £140,000/tU + Pu in the core and £40,000/tU in the radial blanket. For the conservative burnup assumed, on this basis, the penalty to the fast reactor is about £7/kW(e).

The fast reactor is debited with an off-load refuelling penalty as the short-fall in electricity generation from the nuclear sector is made good by the lower merit fossil plant. With a fuel price of 1.8p/therm (50c/MBtu) for the most economic fossil fuelled station the penalty would be about £3/kW(e).

PFR fuel is to be reprocessed in the existing Dounreay plant and the first UK commercial fast reactor fuel will probably be processed through existing head end plants at Windscale. Further development is aimed at new reprocessing facilities in the 1980s, for which no particular problems have been identified.

Attention is being given to achieving short plutonium turn round time and minimum losses, a balance being sought between shorter cooling time and increased fuel handling, reprocessing and transport costs. Analysis has shown that reducing the turn round time of say nine months by one month could give a benefit of £1 to £2/kW.

Operational implications

The importance of fuel process costs in fast reactor fuel cycle economics make it desirable to phase the introduction of new fuel types with the construction of fuel process plants rather than reactors. Accordingly the problems of changing from one type of fuel to another in a fast reactor merit consideration.

The transition from oxide to carbide fuel is an early prospect. This should be relatively straightforward as the time between fuel changing operations is only a few months and the fuel sub-assembly wrappers can be used equally well to accommodate either type of fuel. The main alternatives are to use bulk or progressive fuel changeover.

In the bulk changeover plan, at the start of the last full irradiation period of the oxide fuel before changeover, sub-assemblies that have less than half of their irradiation life in hand are replaced by new ones which are then left in the core to proceed to their full irradiation. The fuel removed is stored, without reprocessing, and used as replacement for fully irradiated fuel until the bulk changeover to the carbide fuel, by which time the whole oxide fuel core should have reached a higher terminal burnup and minimum wastage of the fabrication cost will have occurred.

A progressive fuel substitution method would require more careful organisation but need not incur large economic penalties. The main difficulties are those of neutron flux, and consequently sub-assembly power, to which coolant flow requires matching.

COMPARISON OF ALTERNATIVE PLUTONIUM FUEL CYCLE STRATEGIES

ACITIES (GW(e)) IN UK PROGRAMMES BASED ON MK II GCRs, SGHWRs, OR MK III GCRs

...ctor ...d)	SGHWR									Mk.III GCR								
	All Thermal			Fast Reactor (Reference)			Fast Reactor (Delayed)			All Thermal			Fast Reactor (Reference)			Fast Reactor (Delayed)		
FR	Early Mk.II	U-fed SGHWR	Pu-fed SGHWR	Early Mk.II	SGHWR	FR	Early Mk.II	SGHWR	FR	Early Mk.II	U-fed Mk.III	Pu-fed Mk.III	Early Mk.II	Mk.III	FR	Early Mk.II	Mk.III	FR
-	5	-	-	5	-	-	5	-	-	5	-	-	5	-	-	5	-	-
-	11	7	-	11	6	1	11	7	-	16	2	-	15	2	-	16	2	-
1	11	22	8	11	19	11	11	29	1	16	17	8	15	15	11	16	24	1
13	11	44	18	11	33	29	11	48	14	16	37	20	15	31	27	16	43	14
46	11	74	22	11	42	54	11	48	48	16	71	20	15	44	48	16	46	45
75	6	111	28	6	52	87	6	52	87	11	114	20	9	59	77	11	63	71
18	-	155	37	-	57	135	-	58	134	-	166	26	-	76	116	-	82	110
77	-	195	44	-	47	192	-	40	199	-	211	28	-	72	167	-	76	163
4.8	5.8	30.4	8.8	5.8	18.0	21.2	5.8	23.0	16.2	9.1	27.6	8.3	8.2	17.6	19.2	9.1	21.7	14.2
- -	11.3 100	54.8 486	4.2 38	11.2 99	31.8 282	- -	11.2 100	41.6 369	- -	17.7 156	31.0 275	1.3 11	15.9 141	19.3 171	- -	17.6 156	24.3 216	- -
- -	5.6 79	27.6 387	- -	5.6 78	16.0 225	- -	5.6 78	21.0 294	- -	8.8 124	21.4 300	- -	7.9 111	13.3 187	- -	8.8 123	16.9 236	- -
5.7 334	3.1 111	13.0 340	3.6 174	3.1 114	7.5 208	8.2 479	3.1 115	9.8 272	6.2 364	4.8 176	3.3 618	1.1 277	4.4 158	2.0 383	7.5 434	4.8 175	2.6 483	5.5 320

...and radial blanket are 0.35 and 0.26, respectively.

surcharge of £20,000/t U for plutonium feed. The penalty to the fast reactor due to fuel fabrication and reprocessing is thus about £8/kW(e). After allowing penalties for off-load refuelling, assuming no difference in station construction cost between the SGHWR and the fast reactor, the net saving due to the introduction of fast reactors is about £8/kW(e); rather less than for the case in which the thermal reactor is the Mark II GCR. This saving is reduced by about £5/kW(e) for a 5 year delay in introducing fast reactors.

For the case in which the advanced thermal reactor is the Mark III GCR the first 3 stations are assumed to be introduced in 1976, 1978 and 1979 respectively, whilst the remaining stations are commissioned from 1981 onwards. As in the case of the SGHWR, the balance of thermal reactor requirements is supplied by the Mark II GCR and, on the all-thermal programme, plutonium feed stations are introduced in 1982 with installation limited to 10 GW(e) up to 1986. In the all-thermal programme, the proportion of reactors on a plutonium feed is the highest of all 3 strategies by 1990, but thereafter becomes the lowest as the higher plutonium production rates of the Mark I and Mark II GCR stations are replaced by the lower rate from the Mark III GCRs on a uranium feed. For similar reasons, in the fast reactor programme the proportion of fast reactors is the lowest of the 3 strategies considered.

When supported by the Mark III GCR the fast reactor shows a saving of about £7/kW(e) on uranium ore imports and about £6/kW(e) in respect of separative work requirements. For the Mark III GCR the fabrication and reprocessing cost is about £200,000/tU on a uranium feed, with a surcharge of about £70,000/tU on a plutonium feed. In this case therefore the fast reactor shows a saving on fabrication and reprocessing amounting to about £5/kW(e). Allowing a penalty for off-load refuelling and assuming no difference in station construction cost, the fast reactor achieves a net saving over the Mark III GCR with plutonium recycle of about £15/kW(e), more than 50% greater than the savings for the cases in which the thermal reactor is the Mark II GCR or SGHWR. This saving is reduced by about £6/kW(e) for a 5 year delay in introducing fast reactors.

It appears that the fast reactor is able to show an economic advantage even in the event of adverse differences in station construction cost and with the low burnup assumed. An increase of 1% in burnup increases the benefit from fast reactors by about £5/kW(e). Furthermore, although this advantage to the fast reactor can be materially reduced by delay in its introduction, there is the possibility during the delay of feeding plutonium to thermal reactors for a limited period. In this way it appears possible to reduce the penalty of delaying fast reactors by some 20-25%.

Values have been obtained for the minimum selling price of 3 tonne of Magnox plutonium at the date at which a plutonium shortage first occurs. This is 1987-88 in the case of the all-thermal reactor programmes and 1986 in the case of the fast reactor programmes. For the all-thermal programmes, the minimum selling price works out at about £2.5/g. In the case of the fast reactor programmes, however, the price is upwards of £5/g, falling to zero when plutonium becomes surplus to the requirements of the system in about 40 years. At times before the first occurrence of Pu shortage the price falls by the appropriate discounting factor.

CONCLUSIONS

Good progress has been made in improving experimental data and theoretical techniques required for assessing the use of plutonium in thermal reactors. It has been demonstrated that attention needs to be paid to a number of detailed topics such as self-shielding effects in plutonium resonances and the presence of the americium isotopes for reliable results to be obtained.

From a logistic point of view the Mark II GCR, SGHWR and the LWR are suitable for operation with plutonium discharged from U235 fuelled reactors. The first two seem suitable for continued recycling of plutonium, but this creates increasing difficulty for the particular LWR studied and further investigation is required before a definite conclusion on this fuel cycle could be made.

The helium cooled Mark III GCR with coated particle fuel operates satisfactorily with a plutonium feed if the fuel-to-moderator ratio is adjusted to be lower than in the U235 fuel cycle. Its consumption of plutonium is higher than for the other thermal reactors studied.

While there will be additional operational problems in converting thermal reactors to plutonium fuelling, which require detailed study for specific designs, these are unlikely to have a major impact on the economics of the reactor. The major problem will be in establishing fuel fabrication costs that will be competitive with U235 fuelling. The fabrication cost surcharge with plutonium will depend strongly on the scale and utilisation of the plant in matching reactor programmes and on the fuel specification, but is unlikely to be less than 100% above the U235 fuel costs in practical situations. On this basis the value of Pu is of the order of £2.5/g during the late 1970s.

Improved fuel cycle calculations for sodium cooled fast reactors confirm that their breeding characteristics are unaffected by a wide range of input plutonium isotopic compositions and thus that no problems will arise in using thermal reactor by-product plutonium or in continuous recycling. They can achieve total inventory exponential doubling-times of 15 years with oxide fuel and less than 10 years with carbide fuel – the preferred fuel will be determined by achievable burnup and fabrication costs. Thus the introduction of fast reactors should reduce the need for thermal reactors on logistic grounds. For initiating a fast breeder reactor programme the value of plutonium in the UK in the late 1970s is assessed to be upwards of twice the thermal burning value.

Sodium cooled fast reactors can be initiated with U235 and their breeding characteristics would permit conversion to complete plutonium fuelling within 6 years should overall economics justify this course.

Strategies which involve the use of plutonium depend on a number of specific factors in addition to basic reactor characteristics. From a study based on a typical UK programme it appears that it is preferable to store plutonium for use in fast reactors. The savings of uranium ore and separative work compared with the use of plutonium in thermal reactors more than offsets slightly higher capital and fuel fabrication cost differences even at very modest fast reactor fuel burnup providing the incentive for early large scale exploitation of fast reactors.

In the event of delay in installing fast reactors, economies result from using plutonium in thermal reactors and the penalty incurred can thereby be appreciably reduced.

PLUTONIUM UTILIZATION IN LIGHT-WATER REACTORS

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Abstract–Résumé–Аннотация–Resumen

PLUTONIUM UTILIZATION IN LIGHT-WATER REACTORS.

By 1974 a rapidly rising surplus of plutonium for commercial use will be available from US light-water reactors. It is not expected that the commercial LMFBR will be introduced early enough and in sufficient quantities to warrant storage of the plutonium. This paper describes programs conducted in the United States which will provide the technological basis for safe and economic recycle of the plutonium in light-water reactors. In recent years the USAEC plutonium program has emphasized reactor neutronics, fuel cycle analysis and fuel behaviour in light-water reactors. A large number of experiments have been conducted in critical facilities, PRTR, EBWR and other test reactors. Analysis of these experiments, correlation with theory and application to commercial power plants indicate substantial neutronic differences between uranium and plutonium fuels, but no significant design limitations are seen. Also, irradiations of significant quantities of UO_2 -PuO_2 fuels show performance equal to that of UO_2 fuels. Industry has initiated studies on the technical and economic parameters influencing the commercial utilization of plutonium which include fuel fabrication development, physics methods development, fuel cycle analyses, nuclear design and irradiation experiments. The Westinghouse/USAEC Saxton plutonium experiment will produce peak fuel exposures of 50 000 MWd/MTM by early 1972. The Edison Electric Institute has programs with General Electric and Westinghouse to demonstrate plutonium recycle in a large BWR and PWR. As part of these programs, General Electric and Westinghouse have inserted UO_2-PuO_2 fuels in Big Rock Point BWR and San Onofre PWR, respectively. In a co-operative program between United Nuclear Corporation and Commonwealth Edison, eleven prototype pluronium assemblies have been loaded in the Dresden-1 BWR. As part of each of the above demonstration programs, selected rods are removed at various stages of exposures for careful examination and evaluation. Results to date of the United States programs indicate that UO_2-PuO_2 fuel can be designed to function interchangeably with uranium reload fuel, with flexibility to accommodate varying amounts of plutonium.

UTILISATION DU PLUTONIUM DANS LES REACTEURS A EAU LEGERE.

Un excédent rapidement croissant de plutonium, provenant des réacteurs à eau légère, sera disponible sur le marché américain en 1974. On ne croit pas que les réacteurs surgénérateurs rapides refroidis aux métaux liquides seront mis en service assez tôt et en nombre suffisant, à des fins commerciales, pour justifier le stockage du plutonium. Le mémoire décrit donc les programmes en cours aux Etats-Unis portant sur l'étude de la technologie du recyclage du plutonium pour les réacteurs à eau légère, dans de bonnes conditions d'économie et de sécurité. Pendant ces dernières années, le programme de l'USAEC consacré au plutonium a porté principalement sur la neutronique des réacteurs, l'analyse du cycle du combustible et le comportement

du plutonium comme combustible dans les réacteurs à eau légère. On a fait un grand nombre d'expériences avec des assemblages critiques, le PRTR (réacteur expérimental pour le recyclage du plutonium), le EBWR (réacteur expérimental à eau bouillante) et autres réacteurs expérimentaux. L'analyse de ces expériences, leur confrontation avec la théorie et leur application aux centrales commerciales indiquent qu'il y a de grandes différences en ce qui concerne les neutrons entre les combustibles à l'uranium et les combustibles au plutonium, mais que celles-ci n'impliquent pas de limitations importantes quant aux plans des réacteurs. Les essais d'irradiation de grandes quantités de combustibles à UO_2-PuO_2 ont également indiqué que leur tenue est comparable à celle des combustibles à UO_2. L'industrie a commencé l'étude des paramètres techniques et économiques qui influent sur l'utilisation commerciale du plutonium, en particulier le développement de la fabrication de combustibles, la mise au point des méthodes physiques, l'analyse du cycle du combustible, l'étude technologique et les expériences d'irradiation. Le réacteur expérimental au plutonium Saxton construit en commun par la Compagnie Westinghouse et l'USAEC fournira des expositions maximales de 50 000 MWj/t vers le début de 1972. L'Edison Electric Institute travaille en collaboration avec General Electric et Westinghouse sur des programmes visant à démontrer les possibilités de recyclage du plutonium dans des réacteurs de grande taille à eau bouillante et à eau sous pression. Dans le cadre de ces programmes, General Electric et Westinghouse ont introduit des combustibles à UO_2-PuO_2 dans le réacteur à eau bouillante de Big Rock Point et le réacteur à eau sous pression de San Onofre, respectivement. Le réacteur à eau bouillante Dresden-1 a été chargé de 11 assemblages prototypes au plutonium, suivant le programme de coopération entre United Nuclear Corporation et Commonwealth Edison. Dans le cadre de chacun de ces programmes de démonstration, on enlève des barres déterminées une fois atteints les différents niveaux d'irradiation désirés et on les soumet à un examen approfondi. Les résultats des programmes américains obtenus jusqu'à présent indiquent qu'il est possible de donner aux combustibles à UO_2-PuO_2 des caractéristiques telles qu'ils soient interchangeables avec le combustible à l'uranium aux fins de rechargement, avec une souplesse de fonctionnement qui permet d'utiliser des quantités variables de plutonium.

ИСПОЛЬЗОВАНИЕ ПЛУТОНИЯ В РЕАКТОРАХ, ОХЛАЖДАЕМЫХ ОБЫЧНОЙ ВОДОЙ.

К 1974 году в США будет наблюдаться быстрый рост избытка плутония, производимого в реакторах на обычной воде и пригодного для коммерческого использования. Не ожидается, что коммерческие реакторы-размножители на быстрых нейтронах с жидкометаллическим теплоносителем будут введены достаточно рано и что оправдано хранение значительных количеств плутония. В данном докладе описываются программы исследований, осуществляемых в США, которые обеспечат технологическую основу для безопасного и экономичного использования плутония в реакторах на обычной воде. За последние годы программа исследований плутония КАЭ США предусматривала, в основном, исследования в области нейтронной физики реактора, анализ топливного цикла и изучение поведения топлива в реакторах на обычной воде. Проводилось большое число экспериментов с использованием критических сборок, реактора для испытания плутониевого цикла, экспериментального кипящего водяного реактора и других опытных реакторов. Анализ результатов этих экспериментов и корреляция теории применительно к коммерческим энергетическим реакторам показали существенное различие физики нейтронов для уранового и плутониевого топлива, однако это различие не приводит к существенным проектным ограничениям. Облучение большого количества топлива из двуокисей плутония и урана показало, что характеристики этого топлива аналогичны характеристикам топлива из двуокиси урана. Промышленность начала исследования технических и экономических характеристик, влияющих на коммерческое использование плутония, включая изготовление топлива, физические методы, анализ топливного цикла, конструкции узлов ядерных установок и эксперименты по облучению. К началу 1972 года максимальное выгорание топлива на Сакстонском плутониевом экспериментальном реакторе фирмы "Вестингауз" и КАЭ США достигнет 50 000 МВт·сутки/т. Институт фирмы "Эдисон электрик" совместно с фирмами "Дженерал электрик" и "Вестингауз" разработал программу для демонстрации плутониевого цикла с использованием кипящего водяного реактора и реактора с водой под давлением. В соответствии с этой программой фирмы "Дженерал электрик" и "Вестингауз" облучают топливо из двуокисей урана и плутония в кипящем водяном реакторе на Биг-Рок-Пойнтской атомной электростанции и реакторе с водой под давлением на атомной электростанции в Сан-Онофре. В соответствии с совместной программой фирмы "Юнайтед ньюклеар" и "Коммонуэлс Эдисон" в Дрезденский кипящий реактор №1 загрузили 11 прототипных сборок с элементами из плутония. В соответствии с вышеуказанными программами отобранные элементы вынимались из реактора после достижения определенной глубины выгорания и затем тщательно проверялись и оценивались. Результаты, полученные в рамках программ США, показывают, что топливо из двуокисей урана и плутония может быть заменено при повторной загрузке урановым топливом, содержащим различные количества плутония.

UTILIZACION DEL PLUTONIO EN LOS REACTORES DE AGUA LIGERA.

En 1974 se dispondrá de un excedente rápidamente creciente de plutonio para uso comercial, procedente de los reactores de agua ligera de los Estados Unidos. No es de esperar que los reactores reproductores rápidos de metal líquido comerciales se introduzcan en fecha suficientemente temprana ni en número bastante para justificar el almacenamiento del plutonio. Esta memoria describe los programas llevados a cabo en los Estados Unidos que proporcionarán las bases tecnológicas para el seguro y económico recirculado del plutonio en los reactores de agua ligera. En años recientes el programa de producción de plutonio de la USAEC ha subrayado la importancia de la física neutrónica con relación a los reactores, el análisis del ciclo del combustible, y el comportamiento del combustible en los reactores de agua ligera. Se han realizado gran número de experimentos en instalaciones críticas, el reactor de pruebas de recirculado de plutonio (Plutonium Recycle Test Reactor), el reactor de agua hirviente experimental (Experimental Boiling Water Reactor) y otros reactores de ensayo. Los análisis de esos experimentos, su correlación con la teoría, y la aplicación a las plantas generadoras comerciales indican diferencias neutrónicas sustanciales entre los combustibles de uranio y plutonio pero no se han observado significativas limitaciones de diseño. Del mismo modo, las irradiaciones de cantidades significativas de combustible de UO_2-PuO_2 muestran igual comportamiento que los combustibles de UO_2. La industria ha iniciado estudios sobre los parámetros técnicos y económicos que influyen en la utilización comercial del plutonio y que incluyen el desarrollo de la fabricación de combustible, el desarrollo de métodos físicos, el análisis del ciclo del combustible, el diseño nuclear y los experimentos de irradiación. El experimento sobre el plutonio Westinghouse-USAEC Saxton producirá máximos de irradiación de combustible de 50 000 MWd/t min para principios de 1972. El Edison Electric Institute desarrolla programas conjuntos con General Electric y Westinghouse para comprobar el recirculado del plutonio en un gran reactor de agua hirviente y en un reactor de agua presurizada. Como parte de esos programas, la General Electric y Westinghouse han introducido combustibles de UO_2-PuO_2 en el reactor de agua hirviente de Big Rock Point y en el reactor de agua presurizada de San Onofre, respectivamente. De acuerdo con un programa cooperativo entre United Nuclear Corporation y Commonwealth Edison, se han cargado once conjuntos prototipo de plutonio en el reactor de agua hirviente Drésden-1. Como parte de cada uno de los anteriores programas de comprobación, se han extraído en diversas fases de las irradiaciones barras seleccionadas para su cuidadoso examen y evaluación. Hasta la fecha, los resultados de los programas de los Estados Unidos indican que el combustible UO_2-PuO_2 puede utilizarse para funcionar intercambiablemente con el combustible de uranio recargado, sin límites estrictos respecto a la cantidad de plutonio.

1.0 INTRODUCTION

By 1974 a rapidly rising surplus of plutonium for commercial use will be available from U.S. light water reactors. Although plutonium is projected to have a higher value in fast reactors than water reactors, commercial LMFBR's will not be introduced early enough and in sufficient quantities to warrant or require storage of the plutonium. The USAEC discontinued its plutonium buyback practice as of December, 1970. Because plutonium credit represents a substantial part of the fuel costs, of the order of 0.25 mill/kWh, it is imperative that the U.S. nuclear industry have in a timely manner the necessary demonstrated capability to utilize the projected available plutonium in order to obtain the plutonium credit.

The USAEC has recognized the need for developing the technology for safe and economic recycle of plutonium in thermal reactors and has conducted a major program in this area at its Pacific Northwest Laboratory since 1956 and has provided partial support to Westinghouse for conducting the plutonium experiment in the Saxton reactor. Significant irradiation demonstration programs have been initiated in the Big Rock Point and San Onofre reactors by Edison Electric Institute (EEI) in conjunction with General Electric Company and Westinghouse Electric Corporation and in Dresden Unit 1 as a cooperative effort between Commonwealth Edison Company and United Nuclear Corporation. This paper is based on the results of these programs.

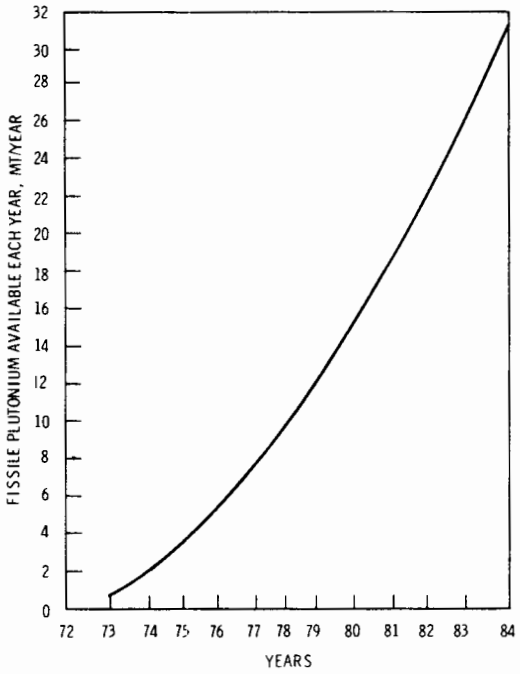

FIG. 1. Plutonium available for thermal recyle each year.

2.0 PLUTONIUM AVAILABILITY AND UTILIZATION STRATEGIES

At the end of 1970, the U.S. electric power industry had in operation or planned nuclear power plants totaling more than 95,000 MWe. The USAEC has projected that the installed capacity will be 150,000 MWe by 1980.[1] Assuming an 80% load factor and equilibrium cycle conditions, these reactors will discharge of the order of 180 kg of fissile plutonium per year for each 1000 MWe of installed capacity. Figure 1 shows the estimated fissile plutonium annual availability versus time for recycle purposes. Annual allocations for fast reactors have been subtracted from the total plutonium discharged.

Because the first commercial fast breeder reactors will not likely be in operation until the mid-1980's, plutonium inventories aimed at this use will not become a significant factor in the plutonium market before the early 1990's. Some short term storage may be required for LMFBR inventories, depending on the rapidity of their introduction. However, analysis of storage and inventory costs indicatesthat it is uneconomic and in fact not required over much of the period before the LMFBR is introduced, even if Pu value in fast reactors is as high as $15/gram. Therefore, plutonium recycle in LWR's is an economic necessity in the U.S.

Utilities are considering two approaches to plutonium recycle: (1) recycling the Pu back into the reactor from which it came, called "self-generation"; and (2) selling the plutonium on the open market. In the "self-generation" case the plutonium recycle fuel in a given core will be no more than one-third of the total fuel. Under these conditions,

it is likely that the fuel subassembly mechanical design will be identical to the uranium assemblies. In the "open market" case, the possibility exists where recycle plutonium could make up the entire core. With an all plutonium core, the incentives would be greater for modifying the assembly lattice design for plutonium so that the plutonium value would be optimized. Whether the cost associated with this optimization can be justified will depend on the Pu value enhancement and the timing of fast reactor introduction.

3.0 PLUTONIUM RECYCLE TECHNOLOGY

3.1 Nuclear Characteristics

The problems encountered in calculating the nuclear performance of plutonium fueled light-water cores have been described in papers presented at other conferences[2,3,4]. Inherently, the problems stem from the complex behavior of the cross sections from zero to 3 eV for the plutonium and transplutonium isotopes, in the transmutation chain, and the degree of nonhomogeneity of the plutonium fuel (i.e., particles of PuO_2 in UO_2). Rather than develop new methods for handling the complexities, calculational methods which were developed for uranium systems have been adopted and modifications made to these methods to make them applicable to plutonium systems. While different sets of calculational methods are used by different organizations, they are similar. The methods used by Battelle-Northwest and which are the bases of the following theory-experiment correlation consist of the Hanford Revised Gam (HRG)[5] for slowing down calculations, the Battelle-Revised Thermos (BRT)[6] for cell spectra calculations, and HFN[7] for calculating the system multiplication factor, k_{eff}.

TABLE I. SUMMARY OF MEAN VALUES OF CALCULATED k_{eff} FOR URANIUM AND PLUTONIUM CRITICALS

Fuel	Number of experiments	Mean value of k_{eff}
2.73 wt% UO_2-H_2O lattices	6	1.000 ± 0.003
Varying set of slightly enriched UO_2-H_2O lattices	6	0.998 ± 0.007
Uranyl nitrate solution in spheres	5	0.995 ± 0.001
1.8 wt% Pu-Al-H_2O lattices	5	1.008 ± 0.002
2.0 wt% Pu-Al-H_2O lattices	5	1.023 ± 0.003
5.0 wt% Pu-Al-H_2O lattices	8	1.009 ± 0.004
Plutonium nitrate solution in spheres	2	1.013 ± 0.001
1.5 wt% PuO_2-UO_2-H_2O lattices	6	1.000 ± 0.002
2.0 wt% PuO_2-UO_2-H_2O lattices		
(8% ^{240}Pu)	7	0.999 ± 0.005
(16% ^{240}Pu)	5	1.003 ± 0.004
(24% ^{240}Pu)	6	1.002 ± 0.003
4.0 wt% PuO_2-UO_2-H_2O lattices	7	0.997 ± 0.006
6.6 wt% PuO_2-UO_2-H_2O lattices	5	1.008 ± 0.007

TABLE II. COMPARISON OF MEASURED AND CALCULATED k_{eff}'s AND POWER DISTRIBUTIONS

Fuel	Array	k_{eff}		Power distributions Std. deviation, σ [a]
		Meas.	Calc.	
2.35 wt% UO_2	Uniform	1.0032	1.0051	1.18
"	H_2O Hole	1.0025	1.0046	0.98
"	H_2O Slot	1.0018	1.0044	1.48
"	H_2O Cross	1.0010	1.0039	1.37
"	7×7 Bundle	1.0010	1.0044	1.72
"	9×9 Bundle	1.0027	1.0010	2.67
UO_2-2 wt% PuO_2 (8% ^{240}Pu)	Uniform	1.0006	1.0080	1.86
	H_2O Hole	1.0020	1.0096	1.78
	H_2O Slot	1.0068	1.0182	1.38
	H_2O Cross	1.0054	1.0231	1.40
	7×7 Bundle	1.0038	1.0220	1.69
	9×9 Bundle	1.0078	1.0245	1.90

[a] $$\sigma = \sqrt{\sum_{i}^{N} (\delta - \delta_i)^2/(N-1)}$$

where N is the number of rods measured,

$$\delta_i = \frac{P_{calc.} - P_{meas.}}{P_{meas.}}$$ for the ith rod,

and

$$\delta = \sum_{i}^{N} \delta_i / N$$

The most reliable and extensive data available for testing the methods are from "clean" lattice experiments.[8] Some reactivity-burnup data are available from the PRTR,[9] EBWR[10] and Saxton[11] reactor experiments. In Table I, the mean of the calculated values of k_{eff} are shown for each series of "clean" critical experiments studied[12]. The results show that with the exception of the 2 wt% Pu-Al and plutonium nitrate series, the agreement between calculation and experiment is within ± 1%. For the UO_2 and UO_2-PuO_2 fueled lattices, the agreement is within ± 0.5%. The reasonably good results shown here may be somewhat fortuitous since other detailed studies show significant discrepancies in various aspects of the calculation such as the thermal disadvantage factor, neutron leakage, core-reflector spectrum, etc.

Table II presents a comparison of calculated and measured multiplication values and power distributions of experiments designed to mockup various aspects of fuel subassemblies and control rods.[13] In these cases, the calculated k_{eff}'s are somewhat higher than the experimentally determined values. Also shown is the standard deviation of power distributions.

Limited reactor data exist for evaluating burnup calculations. Based on a partially completed batch core experiment in PRTR using UO_2- 2 wt% PuO_2 fuel, the calculated exposure life of the core using methods described

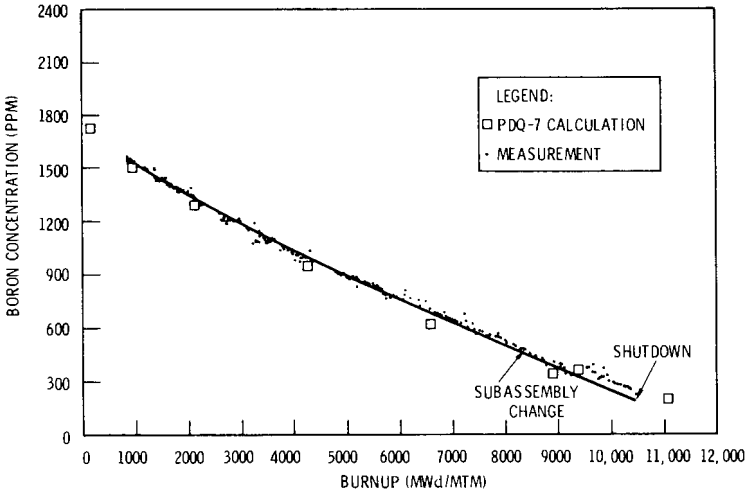

FIG. 2. Saxton core II: A comparison of the calculated and measured critical boron concentration as a function of burnup.

above was 17,400 MWd/MTM[1] compared with the extrapolated measured lifetime of 14,100 MWd/MTM[14]. Today, methods normalization is required in order to obtain desired burnup predictions. Westinghouse, using their code system appropriately modified for plutonium fuels and modifying the cross sections to improve correlation with lattice experiments and the Saxton burnup data, has obtained good agreement between calculation and experiment throughout the life of Core II and during the first year of Core III of the Saxton plutonium experiment.[11] An example of this is shown in Figure 2.

Because of the variation with energy of the capture and fission cross sections in the thermal and near thermal region and the large ^{240}Pu capture resonance at 1 eV, the moderator temperature, void and doppler reactivity coefficients are generally more negative for plutonium than for uranium fueled reactors. These coefficients all represent reactivity feedback in a transient and affect the design shutdown margin. A qualitative summary of the basic differences between nuclear design characteristics of large (1000 MWe) plutonium fueled reactors and uranium fueled ones is shown in Table III. The most significant difference is the increased number of required control rods for the plutonium fueled core which is brought about because of the larger doppler and moderator coefficients and the reduced control rod worth. However, by using reload strategies, plutonium loading in control rod positions can be avoided, thus minimizing the reduction in control rod worth. Also, limiting the plutonium in the core to the self-generated case greatly reduces the various reactivity differences.

3.2 Fuel Characteristics

Pellet fuels, because of the large amount of satisfactory UO_2 experience, is the reference design for plutonium utilization in LWR's. Packed particle fuel (Vipac) is considered to be the most promising alternate form. Under the operating conditions presently employed or projected for

[1] MTM = metric tonnes metal.

TABLE III. CAPSULE COMPARISON OF URANIUM AND PLUTONIUM NUCLEAR DESIGN CHARACTERISTICS

Parameter	Plutonium core	Reason for difference	Consequence
Moderator temperature coefficient	More negative	Increased resonance absorption and spectrum shift	Improved stability and transient characteristics except for steam break
Void coefficient	More negative	Increased resonance absorption and spectrum shift	Improved stability and transient characteristics except for steam break
Doppler coefficient	More negative	^{240}Pu resonances	Improved transient characteristics
Cold-to-hot reactivity swing	Increased	Larger moderator temperature coefficient	Increased boron (PWR) or control rod (BWR) requirements
Installed reactivity	Reduced	Reduced depletion rate-reactivity saturates	None
Control rod requirement	Increased	Larger moderator and Doppler coefficients	Possible increase in number of rods
Control rod worth	Reduced	Thermal flux reduced	Possible increase in number of rods
Boron worth	Reduced	Thermal flux reduced	None
Xenon worth	Reduced	Thermal flux reduced	Improved stability
Fission product poisons	Increased	Increased yields - increased resonance absorptions	Reactivity penalty
Local power peaking	Increased	Increased water worth	Fuel management action required
Delayed neutron fraction	Reduced	$\beta_{Pu} < \beta_u$	Rod ejection accident

Qualifications: 1. Effects can be modified by changes in design H/F.
2. Successive recycles influence the parameters.

commercial LWR's, neither fuel type appears to exhibit inherent performance limitations. The satisfactory irradiation performance of mixed-oxide fuels in thermal reactors has been demonstrated through the success of numerous experimental and demonstration programs (Table IV).

Experimental evidence indicates that small PuO_2 additions to UO_2 for the LWR fuel enrichment do not significantly affect fuel thermal conductivity[15], fission product migration[16], fission gas release[17][11], and fuel clad interactions in either pellet or Vipac fuels.

Although the small PuO_2 additions to UO_2 for thermal reactor fuel has an insignificant effect on most performance characteristics, there are considerations related to performance that are unique with plutonium enriched fuels. Mixed-oxide fuel prepared by coprecipitation is essentially uniform $(U, Pu)O_2$ solid solution[18]. However, the more commonly used preparation method of mechanical mixing UO_2 and PuO_2 powders prior to pressing and sintering results in the formation of localized regions of high PuO_2 concentration[19]. Uniform solid solution forms rapidly in regions of mechanically mixed mixed-oxide fuel operating above columnar

TABLE IV. SIGNIFICANT MIXED-OXIDE IRRADIATIONS

Reactor	No. of rods	Fuel type [a]	Peak burnup (MWd/MTM) [b]	Peak rod power (kW/ft)	Peak heat flux (Btu/h-ft^2)
PRTR	~1860	Swage	17,000	17	390,000
PRTR	~700	Vipac	18,500	16	370,000
PRTR (high power density)	~1730	Pellet (cold-press-sinter & hot press) Vipac, Swage	13,000	21.5	500,000
EBWR	1296	Vipac	3,000	8	200,000
Saxton-Core II	638	Pellet-Vipac	29,000	16	530,000
Saxton-Core III	~250	Pellet	44,000	19	630,000
Dresden-I	4	Hot-press pellet	13,000	10	230,000
Dresden-I	99	Pellet	14,800	14.9	345,000
San Onofre	720	Pellet	8,500	13	460,000
Big Rock Point	32	Pellet-solid dished & annular	19,500	14	325,000
Big Rock Point	204	Pellet-annular	14,600	16	370,000
Garigliano	96	Vipac(12), hot-press pellet(24) Cold-press-sinter Pellet(60)	13,800	15	333,000
Vermont-Yankee	48	Pellet-solid & annular	-	17.3	403,000

[a] Pellets are cold pressed and sintered unless otherwise noted.
[b] Projected - September, 1971

grain growth temperatures (~1700°C) by a vaporization-condensation process; whereas, homogenization occurs more slowly in the equiaxed grain growth regions (1400-1700°C) by solid state diffusion. Essentially no further homogenization occurs during irradiation at fuel temperatures below 1400°C. Therefore, localized regions of high plutonium concentration persist in a significant volume of the fuel in a rod operating under normal conditions. The high fission density associated with localized regions of high plutonium concentration due to either migration or large particles could affect fuel swelling or fission gas release although there is no definitive supportive evidence of this for fuel operating under power reactor conditions. In any event, PuO_2 particles in mechanically mixed fuel can be controlled to acceptable levels.

A significant change in radial plutonium concentration can occur rapidly in mixed-oxide fuel rods during irradiation[16]. For mixed-oxide fuel operating above columnar grain growth temperatures, radial plutonium redistribution is coincident with the fuel restructuring phenomena and occurs as a result of the preferential evaporation of gross amounts of uranium oxide from the central hot region of the fuel to the cooler peripheral regions near the cladding. Thermal diffusion can also cause plutonium enrichment in the high temperature region of the fuel[20].

The behavior of oxide fuels when subjected to accidental high energy, short duration, power excursions is an important safety consideration in both thermal and fast reactors. The results of comparative transient experiments performed with unirradiated pellet and particle fuel pins show that:[21,22]

(i) The cladding failure threshold energy of approximately 270 cal/g fuel is essentially the same for both pellet and particle fuels.

(ii) Both pellet and particle fuels fail by clad melting, however, because of higher fuel temperatures in particle fuels at comparable energy depositions, more fuel is expelled from the pin.

(iii) The extent of Zircaloy-water reaction is comparable for both pellet and particle fuels and increases with increasing total energy deposition.

The results of transient tests conducted to investigate the possible effect of large (550 μm dia.) PuO_2 particles, which could be present in fabricated mixed-oxide fuel, show that the cladding failure threshold energy was reduced slightly from the range of 225 to 274 cal/g fuel to the range of 200 to 213 cal/g fuel[23]. There were no indications of the effects of prompt fuel dispersal caused by the expulsion of the PuO_2 particles into the surrounding water when tested at these energy levels. Based upon the results of these experiments, UO_2-PuO_2 product specifications which limit the maximum PuO_2 particle size below 550 μm diameter do not appear to be warranted from the standpoint of transient fuel performance considerations.

Tests on purposely defected Zircaloy-clad mixed-oxide fuel rods indicated that the defect performance of both pellet and Vipac fuels is excellent under normal operating conditions.[24] From these tests, defect behavior was observed to be most sensitive to fuel type, fuel temperature, and power history, with activity release greater for Vipac fuel under comparable heat ratings than for pellet fuel.[25,26]

3.3 Fuel Fabrication

Mixed-oxide pellet fuels can be prepared by sintering either coprecipitated $(U, Pu)O_2$ powder or a mechanical mixture of UO_2 and PuO_2. Compacting and hot-pressing the blended powder has also been investigated. Variations in the basic pellet fuel design such as controlled density, controlled diametral gap, dishing and coring are utilized to satisfy various performance criteria. Packed particle fuel is an alternate to pellet fuels and may be of the vibrationally compacted or swage-compacted concepts, utilizing various feed materials such as sintered and crushed, fused, electrodeposited, pneumatically impacted, or Sol-Gel[21]. Cost studies indicate approximately equal fabrication costs for pellet and Vipac mixed-oxide fuels.[27]

Fuel fabrication costs are very sensitive to plant throughput.[27] When comparing fabrication plants of equal size ranging from 1/4 to 1 ton/day throughput, plutonium fuel fabrication costs are approximately 18% to 25% higher than for UO_2. However, the more likely situation in the early years of plutonium recycle is that the throughput for plutonium plants will be 1/4 ton/day or less and that of uranium throughput will be one ton/day or more. When compared on this basis, the plutonium fuel fabrication penalty will be at least 70% to 80%. Such a penalty would

reduce the plutonium value by approximately two dollars per gram, depending on enrichment and actual fabrication cost differences.

In addition to the usual safeguards related to criticality and alpha contamination, attention must be given to the significant gamma and neutron radiation in fabricating and handling plutonium.[28,29,30] The dose rate from the plutonium is a function of the reactor type, fuel exposure, and time since processing.

Initially, the gamma dose rate from unshielded high exposure plutonium is due mostly to ^{238}Pu. With successive increases in shielding thickness, radiation from ^{241}Pu and then ^{239}Pu become dominant. Activity associated with the daughter products of ^{236}Pu and ^{241}Pu causes a substantial increase of dose rate with time since reprocessing. With successive increases in shielding the dominant isotopes in aged plutonium are ^{238}Pu, ^{241}Am, ^{237}U, ^{241}Pu, ^{239}Pu, ^{241}Am(again) and ^{208}Tl (a daughter of ^{236}U).

Neutron dose measurements from plutonium are not well characterized. The isotopic composition of plutonium has a significant effect on dose rate due to spontaneous fission and (α, n) reaction with oxygen and possible impurities. Because of its relative high concentration, ^{240}Pu usually contributes the largest percent of neutrons emanating from high exposure plutonium.

Surface dose rates from high exposure plutonium may be of the order of several rem/hr. But because much of the radiation consists of low energy gammas, plutonium fuels can be fabricated successfully in glove box facilities with only small amounts of shielding. Studies have shown that the incremental fuel fabrication cost factor for high exposure UO_2-PuO_2 (^{239}Pu <80%) fuels is about 4% higher than for fuel containing low exposure plutonium ($\sim$93% ^{239}Pu).[27]

4.0 UTILIZATION IN BOILING WATER REACTORS (BWR)[31]

4.1 General Characteristics of the Mixed Oxide Fuel Design

The BWR core consists of modules of 4 fuel bundles surrounding a cruciform control rod (see Figure 3). The control rod water gap causes a non-uniform neutron flux distribution across the fuel bundles. To achieve minimum power peaking within the bundles, enrichment is varied among individual rods. Generally, the central rods contain higher enrichment than the peripheral rods. Another important feature of the BWR fuel is the use of several rods of gadolinium oxide (Gd_2O_3) in UO_2 as a supplementary burnable poison.

Plutonium fuel is accommodated in the same mechanical design by substituting mixed oxide rods for a portion of the uranium rods. Figure 3 also shows a typical configuration where ten UO_2-PuO_2 rods replace the central region of highest uranium enrichment, yielding the highest Pu value. This configuration will accommodate approximately 1.1 to 1.3 times the self-generated plutonium. The mixed oxide island is displaced away from the control blade corner to minimize the effect of the large Pu absorption on the movable control rod worth. Gadolinia-urania rods are located within the plutonium island to provide supplementary control.

This type of bundle can be designed to function interchangeably with uranium fuel and due to the control flexibility provided by the Gd_2O_3-UO_2 can accommodate a variable number of mixed oxide rods. Although

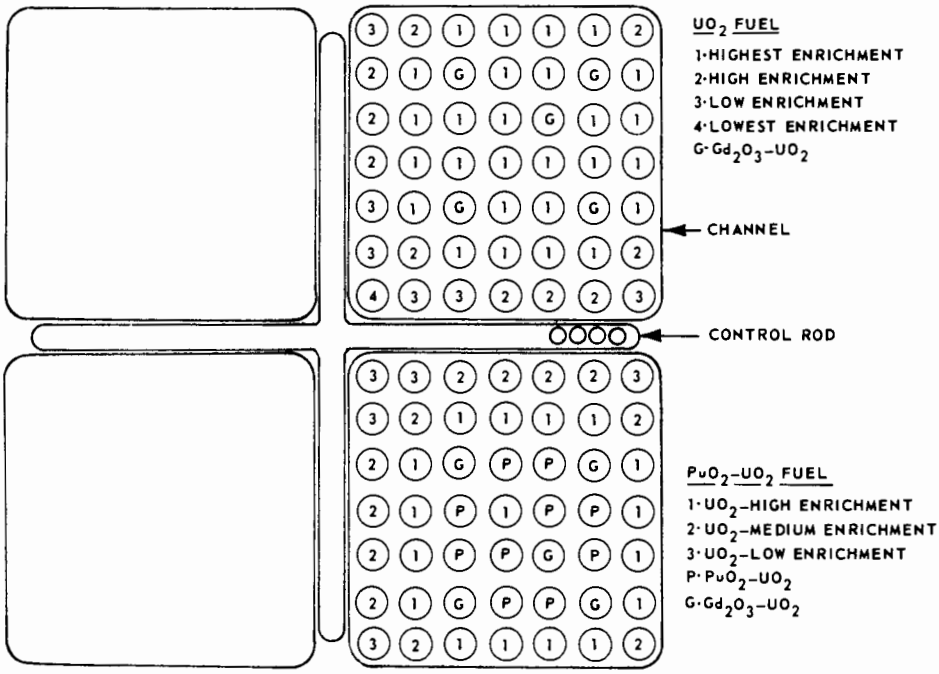

FIG. 3. Typical BWR fuel module with control rod.

TABLE V. NUCLEAR CHARACTERISTICS OF COMPARABLE URANIUM AND PLUTONIUM RECYCLE BWR RELOAD FUEL

	Plutonium	Uranium
Exposure, MWd/MTM [a]	28,100	27,500
k_∞, No xenon, 0 exposure	1.23	1.24
Exposure slope, $\%\Delta k_\infty$/1000 MWd/MTM	1.11	1.12
Fissile atom depletion per 1000 MWd/MTM	0.00045	0.00045
Fissile fuel/total fuel	0.0278	0.027
Number Gd_2O_3-UO_2 rods	5 - 6	5
$\Delta k_\infty/k_\infty$ cold control	0.12	0.14
Δk_∞ cold shutdown - hot operating	0.171	0.160
P/A local power	1.20	1.23
Void response, $\Delta k_\infty/k_\infty/\Delta$ void	-.10	-.09
β_{eff} at 17,500 MWd/MTM	0.0051	0.0054

[a] For equivalent energy per bundle.

bundle designs with larger number of mixed oxide rods are feasible, the design complexity is increased and the economic value of Pu is somewhat reduced relative to the 10-rod bundle. Because of the virtual interchangeability of bundle designs, a reload batch may be made up of any combination of mixed oxide island bundles and uranium rod bundles.

4.2 Detailed Characteristics of 10 Rod Mixed Oxide Fuel Design

The following sections describe the characteristics of a typical 10 rod mixed-oxide fuel assembly designed to be interchangeable with uranium fuel for an equilibrium core exposure capability of 27,500 MWd/MTM (Refer to Table V.)

These characteristics are based upon use of annular mixed-oxide pellets which increase the H to Pu ratio in the Pu island and produce more favorable nuclear characteristics.

Power Peaking

In this design the peak uranium rod has a relative power of 1.20 times bundle average and the peak mixed-oxide rod is 1.18. The peak mixed-oxide rod remains consistently below the peak uranium rod during burnup.

Reactivity

The initial reactivity of the mixed-oxide bundle is slightly lower but it has a lower slope with fuel exposure yielding slightly higher reactivity at high exposure.

Transient Characteristics

The delayed neutron fraction (β_{eff}) of the mixed-oxide fuel is 5-10% lower and the doppler reactivity coefficient is 5-10% more negative. These differences are sufficiently small to produce only minor effects on over-all reactor operational and transient characteristics and safeguards requirements.

4.3 Development Programs

4.3.1 EEI-General Electric Plutonium Recycle Demonstration Program[32,33]

The principal experimental activities of this program are:

1. Thirty-two (32) removable UO_2-PuO_2 rods of various pellet fuel designs in 16 bundles of uranium reload fuel and 3 fuel bundles of UO_2-PuO_2 rods in the Big Rock Point Reactor of Consumer Power of Michigan. The removable rods have successfully completed 2 cycles and the bundles 1 cycle. Several specimen rods have been thoroughly examined in the radioactive materials laboratory with favorable results.

2. Four (4) bundles in the initial core of the Vermont Yankee reactor. These bundles include a 12 rod island of UO_2-PuO_2 and are directly interchangeable with initial core fuel. These bundles are being fabricated and will be installed in the initial core. Two bundles contain solid pellets and 2 contain annular pellets of UO_2-PuO_2.

4.3.2 Commonwealth Edison - United Nuclear Demonstration Program[34]

A cooperative demonstration program has been initiated by Commonwealth Edison and United Nuclear Corporation to develop the information needed for supplying a full plutonium recycle reload batch. In this program, eleven prototype plutonium assemblies have been designed and fabricated. These assemblies will operate in the Dresden-1 reactor for four cycles with periodic inspections at each refueling period. As of September 1, 1971, the average exposure for all plutonium recycle assemblies is expected to be 8,300 MWd/MTM with the highest power assembly having an average exposure of 9,000 MWd/MTM. The highest power PuO_2-UO_2 rod will have obtained an average exposure of 10,800 MWd/MTM and the highest power pellet an irradiation of 14,800 MWd/MTM.

5.0 UTILIZATION IN PRESSURIZED WATER REACTORS (PWR)

5.1 Fuel Reload Strategies and Design Considerations

The results and conclusions presented in this section are based on several programs on plutonium recycle in pressurized water reactors conducted by Westinghouse Electric Corporation. Results to date from these programs indicate that commercial plutonium recycle fuel for pressurized water reactors will have the following characteristics:

(i) The reload fuel will have the same mechanical design for both the plutonium recycle and enriched uranium assemblies.

(ii) The plutonium recycle assemblies will consist entirely of mixed-oxide fuel rods.

(iii) There will be a minimum of three different plutonium enrichments in each plutonium recycle assembly.

Core power distribution studies show that recycle can be accomplished using these characteristics without incurring a penalty on either core lifetime or power capability. This is not always the case when both mixed-oxide and uranium oxide fuel rods are installed in the same fuel assembly. Different plutonium enrichments are required in each assembly to control local power peaking at the uranium-plutonium interfaces and at the control rod thimbles (water holes). This is illustrated in Figure 4 for the assemblies which are currently operating in the San Onofre reactor.

Another advantage of the assembly design containing all mixed-oxide rods is that it minimizes the number of Pu recycle assemblies in the core. This, in turn, minimizes the reduction of control rod worth that occurs when plutonium is installed in the core by being able to avoid control rod positions. No significant licensing or safety problems are anticipated for PWR plutonium recycle. For example, plutonium particle sizes can be controlled to avoid undesirable steady-state or transient performance characteristics.

5.2 Demonstration Programs

5.2.1 Saxton Plutonium Program

The first demonstration of plutonium loading in PWR's was the Westinghouse conducted Saxton Plutonium Program. It has been a joint

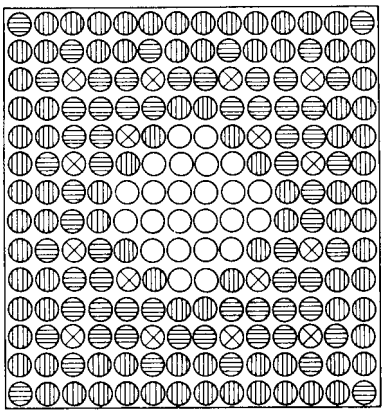

FIG.4. Enrichment pattern for the EEI-Westinghouse demonstration assemblies.

effort with the AEC and the Saxton Nuclear Experimental Corporation. The program has these objectives.

(a) Demonstrate performance of mixed-oxide fuel at linear power and burnup levels consistent with modern PWR technology; and

(b) Obtain design information on depletion and transuranic isotope generation characteristics of plutonium fuel at higher burnup.

The Core II post-irradiation program was completed in early 1970. The peak burnup evaluated was 29,000 megawatt days per tonne (MWd/MTM) on pellet and Vipac mixed-oxide fuel. Seven open-lattice assemblies consisting of approximately 250 rods from Core II have continued to operate satisfactory in Core III since December, 1969. Since then, a significant number of plutonium rods have operated at 19 kW/ft. Peak burnup for the mixed-oxide fuel is about 44,000 MWd/MTM. By early 1972, the Saxton plutonium fuel should achieve 50,000 MWd/MTM peak burnup and will have demonstrated the peak power and burnup levels required for modern PWR's.

5.2.2 EEI-Westinghouse Plutonium Recycle Demonstration Program[35]

In early 1968 the EEI-Westinghouse Plutonium Utilization Program was completed. This program was primarily an analytical feasibility study and constituted the necessary preliminary step toward actual irradiation of plutonium in a commercial PWR. The remaining step is to demonstrate the concept in a large pressurized water reactor of today's design. To accomplish this, the EEI-Westinghouse Plutonium Recycle Demonstration Program has been undertaken. The purpose of this program is to license, operate and evaluate a representative number of plutonium fuel rods in the San Onofre reactor. This demonstration experience under actual utility

operating conditions will complement the material and design recycle information being generated in the Saxton test reactor. In order to achieve a meaningful demonstration, Westinghouse first determined the characteristics of future commercial recycle fuel, and then selected a prototypic recycle configuration for the demonstration fuel. As a result, each of the four demonstration assemblies in San Onofre consist entirely of mixed-oxide fuel rods and have three different plutonium enrichments.

In early 1970 fabrication was completed on the 720 plutonium-bearing, Zircaloy-clad fuel rods required for the EEI Demonstration Assemblies. The four San Onofre Demonstration Assemblies contain 45 kilograms of plutonium, five times the current Saxton loading. These assemblies were loaded in the San Onofre reactor during its first refueling shutdown and achieved full power operation in early November 1970. They will be operated during three reactor cycles. Periodic measurements and tests will be made as a check on analytical predictions. At the end of each cycle selected plutonium fuel rods will be removed, examined and returned to the core. Some of the 104 removable rods will be replaced and then shipped for post-irradiation examinations. By September, 1971, the Demonstration Assemblies will have achieved about 5,000 MWd/MTM burnup, and the next San Onofre refueling is scheduled for January 1972.

ACKNOWLEDGEMENTS

The authors express their appreciation to all of those who assisted in preparing this paper and particularly to Messrs. M. D. Freshley, R. C. Liikala, and H. H. Van Tuyl for their contributions.

REFERENCES

[1] *The Nuclear Industry - 1970,* U.S. Atomic Energy Commission, November 18, 1970.

[2] *Commercial Plutonium Fuels Conference,* Washington D.C., pp. 205-249, March 1-2, 1966, USAEC Report CONF-660308.

[3] *Plutonium as a Reactor Fuel,* Proceedings of a Symposium, Brussels, March 13-17, 1967, Published by International Atomic Energy Agency.

[4] International Atomic Energy Agency Panel on Plutonium Utilization, Vienna, Austria, September 2-6, 1968, IAEA-112.

[5] Carter, J.L., *HRG-3: A Code for Calculating the Slowing-Down Spectrum in the P_1 or B_1 Approximation,* BNWL-1432 (1970).

[6] Bennett, C. L., and W. L. Purcell, *BRT-1: Battelle-Revised-THERMOS,* BNWL-1434 (1970).

[7] Lilley, J. R., Jr., *Computer Code HFN-Multigroup, Multiregion Neutron Diffusion Theory in One Space Dimension,* USAEC Report HW-71545 (1961).

[8] Schmid, L. C., et. al., *Reactor Physics Data for the Utilization of Plutonium in Thermal Reactors,* BNWL-801, May 1968.

[9] Smith, R. I., et. al., *Critical Experiments with the UO_2 - 2 wt.% PuO_2 Batch Core in the PRTR,* BNWL-1553, March 1971.

[10] Dawson, F. G., et. al., *The EBWR Plutonium Demonstration Experiment,* Trans. Am. Nucl. Soc., 10, 500 (1967).

[11] Smalley, W., *Saxton Core II Fuel Performance Evaluation,* WCAP-3385-56, December, 1970.

[12] Liikala, R. C., et. al., *Uncertainties in the Analysis of Plutonium Fuel Assemblies,* Trans. Am. Nucl. Soc. 1, 227 (1969).

[13] Loizzo, P., et. al., *Experimental and Calculated Results for UO_2 - and UO_2-PuO_2 Fueled - H_2O Moderated Loadings,* BNWL-1379, August 1970.

[14] Prezbindowski, D. L., Private Communication, Battelle, Pacific Northwest Laboratories.

[15] Gibby, R. L., *The Effect of Plutonium Content on the Thermal Conductivity of (U, Pu)O_2 Solid Solutions*, J. Nucl. Materials, Vol. 38, 163-177, February, 1971.
[16] Bahl, J. K., and M. D. Freshley, *Plutonium and Fission Product Migration in Mixed-Oxide Fuels during Irradiation*, Trans. Am. Nucl. Soc., Vol. 13 (2), 1970.
[17] Freshley, M. D., and T. B. Burley, *Vibrationally Compacted Ceramic Fuels*, BNWL-SA-2412, February 1969.
[18] Caldwell, C. S., and K. H. Puechl, *Plutonium-Uranium Dioxide Powder and Pellet Manufacture*, AIME Symposium on Plutonium Fuels Technology, Scottsdale, Arizona, October 1967.
[19] *Plutonium Utilization Program Technical Activities Quarterly Report - March, April, May 1970*, BNWL-1442, pp. 3.18-3.24, June 1970.
[20] Bober, M., C. Sari and G. Schumacher, *Plutonium Migration in a Thermal Gradient in Mixed (U, Pu) Oxide Fuels*, Trans. Am. Nucl. Soc., Vol. 12 (2), 1969.
[21] Freshley, M. D., *A Comparison of Pellet and Vipac Nuclear Fuels*, BNWL-SA-3466, July 1970.
[22] Gulley, R. L., L. J. Harrison, and D. R. Armstrong, *Transient Irradiation Experiments with Vibrationally Compacted UO_2 Fuel Rods in TREAT*, Trans. Am. Nucl. Soc., Vol. 11 (1) 1968.
[23] Freshely, M. D., E. A. Aitken, D. C. Wadekamper, R. L. Johnson and W. G. Lussie, *Behavior of Discrete PuO_2 Particles in Mixed-Oxide Fuel During Rapid Power Transients*, Trans. Am. Nucl. Soc., Vol. 13 (2) 1970.
[24] Freshley, M. D., and F. E. Panisko, *The Irradiation Behavior of UO_2-PuO_2 Fuels in PRTR*, BNWL-366, March 1967.
[25] Freshley, M. D., R. G. Wheeler, J. M. Batch and G. M. Hesson, *The Combined Failure of a Pressure Tube and Defected Fuel Rod in PRTR*, Trans. Am. Nucl. Soc., Vol. 9 (2) 1966.
[26] Locke, D. H., *Defected Zircaloy Fuel*, Nuclear Engineering International, August 1969.
[27] Burnham, J. B., L. G. Merker, and D. E. Deonigi, *Comparative Cost of Oxide Fuel Elements*, Vol. 1, 2, and 3, BNWL-273, July 1966.
[28] Merker, L. G., L. G. Faust, W. J. Bailey and Staff, *Battelle-Northwest Fuel Fabrication Experience with High Exposure Plutonium*, BNWL-27, January 1965.
[29] Van Tuyl, H. H., *Calculation of Gamma Dose Rates at the Surface of Plutonium Oxide Sources*, BNWL-1259, January 1970.
[30] Smith, R. C., L. G. Faust, and H. H. Van Tuyl, *The Effect of Radiation Levels from Plutonium on Fuel Fabrication Process Design*, BNWL-SA-3139. May 1970.
[31] Fischer, D. L., et. al., *The Utilization of Plutonium as an Alternative Fuel in Boiling Water Reactors*, Nuclex '69 Technical Meeting No. 2/6.
[32] *Plutonium Utilization in BWR Phase II*, General Electric Report NEDC-12152, Semiannual Report of work performed for the Edison Electric Institute Project No. RP72, January-June 1970.
[33] *Plutonium Utilization in BWR Phase II*, General Electric Report NEDC-12111, 1969 Annual Report of work performed for the Edison Electric Institute Project No. RP 72.
[34] Tulenko, J. S., G. T. Hamilton, *A Demonstration Program for Plutonium Utilization in a Boiling Water Reactor*, NED-681, June 1970.
[35] Haley, J., *EEI-Westinghouse Plutonium Recycle Demonstration Program Progress Report for the Period Ending December, 1970*, WCAP 4167-2, April 1971.

PLUTONIUM RECYCLING IN LIGHT-WATER REACTORS

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Abstract–Résumé–Аннотация–Resumen

PLUTONIUM RECYCLING IN LIGHT-WATER REACTORS.

The Belgian plutonium recycling program, started in 1958, was gradually increased to cover most aspects of plutonium utilization in LWRs. Prospective studies dealt with the prerequisite plutonium availability, manufacturing costs and cost of interim storage to benefit from large-scale facilities. The incentives to use plutonium in first generation PWRs (stainless-steel cladding, cruciform rods, flexibility for progressive conversion to plutonium fuelling) and in BWRs (separate fuel element channels, multiple enrichment elements), and the simultaneous recycling in both reactor types of the enriched uranium recovered from reprocessed fuel were examined in detail. The nuclear PANTHER code chain was tested on results of about 130 critical experiments in the VENUS facility, from simple lattices to mock-ups of recycle configurations, in which almost 3000 pelleted or vibrated oxide fuel rods were used. The thermohydraulic codes were calibrated for special situations encountered in plutonium fuel environments. COMETHE and CRASH, fuel rod design codes, were checked against many irradiation experiments. The fuel development program included the irradiation of over 80 specimens in BR2 and DIORIT, covering heat ratings up to 1300 W/cm, cladding temperatures up to 600°C and burn-ups up to 50 000 MWd/t. Both pelletizing and vibrocompaction manufacturing techniques were developed from the laboratory to the pilot stage; a large plant is now under construction. These activities were complemented by a diversified assessment and demonstration program. BWR plutonium recycle assemblies were designed for Kahl and Dodewaard, fuel was fabricated for Garigliano and Dodewaard and there was close collaboration with ENEL's experimental program in Garigliano. For PWRs, various recycle schemes, covering four reactor cycles, were studied for SENA and TRINO type plants; shutdown margins and control-rod worths were investigated. Plutonium fuel was gradually introduced in the BR3 from 1963 on; at the end of 1970, a peak burn-up of 40 000 MWd/t was reached in a UO_2 - PuO_2 assembly.

RECYCLAGE DU PLUTONIUM DANS LES REACTEURS A EAU LEGERE.

Depuis 1958, le programme belge d'étude du recyclage du plutonium s'est graduellement étendu à la plupart des aspects de l'utilisation de cet élément dans les réacteurs à eau légère. Les études prospectives ont porté sur les quantités nécessaires de plutonium, les coûts de fabrication et de stockage temporaire permettant de profiter des installations de grande capacité. Les auteurs étudient les avantages de l'emploi du plutonium dans les réacteurs à eau sous pression (gainage en acier inoxydable, barres cruciformes, facilité de conversion progressive au plutonium) et les réacteurs à eau bouillante (canaux d'éléments combustibles séparés, éléments à enrichissement multiple), ainsi que l'intérêt de recycler simultanément, dans les deux filières, l'uranium enrichi provenant du combustible retraité. La chaîne de code nucléaire PANTHER a été éprouvée sur la base d'environ 130 expériences critiques réalisées dans l'installation VENUS, allant des simples réseaux aux maquettes de configuration de recyclage; environ 3000 barres de combustible d'oxyde vibro-compacté ou pastillé ont été utilisées à cette fin. Les codes thermohydrauliques ont été étalonnés pour des situations typiques de recyclage. COMETHE et CRASH, codes de calcul des barres combustibles, ont été vérifiés à l'aide de nombreuses expériences d'irradiation. Le programme d'étude du combustible a porté sur plus de 80 échantillons irradiés dans BR2 et DIORIT à des puissances linéiques atteignant 1300 W/cm, des températures de gaine atteignant 600°C et des taux de combustion jusqu'à 50 000 MWj/t. Des techniques de fabrication par vibro-compactage ou pastillage ont été mises au point, depuis les études en laboratoire jusqu'au stade pilote; une grande usine est en construction. Ces activités ont été complétées par un programme diversifié de démonstration et d'évaluation: pour les réacteurs à eau bouillante, on a étudié des assemblages de Pu recyclé pour KAHL et DODEWAARD, fabriqué du combustible pour GARIGLIANO et DODEWAARD, collaboré au

programme expérimental ENEL sur GARIGLIANO; pour les réacteurs à eau sous pression on a étudié divers schémas de recyclage, couvrant quatre cycles de réacteurs, pour les filières SENA et TRINO, y compris les calculs des marges d'arrêt et l'antiréactivité des barres de commande. Depuis 1963, du combustible au plutonium a été graduellement introduit dans BR3; à la fin de 1970, un taux de combustion atteignant 40 000 MWj/t y avait été obtenu dans un assemblage de UO_2-PuO_2.

ПОВТОРНОЕ ИСПОЛЬЗОВАНИЕ ПЛУТОНИЯ В РЕАКТОРАХ, ОХЛАЖДАЕМЫХ ОБЫЧНОЙ ВОДОЙ.

Бельгийская программа исследований повторного использования плутония, которую начали осуществлять в 1958 году, постепенно расширялась и охватывает в настоящее время большинство вопросов, касающихся использования плутония в реакторах, охлаждаемых обычной водой. В перспективных исследованиях рассматривались необходимая, как условие, доступность плутония, стоимость изготовления тепловыделяющих элементов и стоимость временного хранения плутония, которое экономически выгодно для крупных заводов. Были детально изучены мотивы для использования плутония в первых энергетических реакторах с водой под давлением (оболочки из нержавеющей стали, стержни крестообразного профиля, возможность постепенного перехода к плутониевой топливной загрузке) и в кипящих реакторах (отдельные топливные каналы, элементы с многократным обогащением), а также возможность одновременного использования в реакторах обоих типов обогащенного урана, восстановленного после извлечения из переработанного топлива. Кодовая цепочка PANTHER, разработанная для ядерных расчетов, проверялась по результатам примерно 130 критических экспериментов в установке VENUS, включающих как простые решетки, так и макеты реактора, воспроизводящие повторную топливную конфигурацию. В этих экспериментах использовалось до 3000 тепловыделяющих элементов со спеченным или виброуплотненным окисным топливом. Термогидравлические расчеты использовались для специальных ситуаций, встречающихся в реакторах с плутониевым топливом. Программы COMETHE и CRASH, касающиеся проектирования топливных стержней, проверялись по результатам многих экспериментов по облучению. Программа разработок топливных материалов включала облучение более 80 образцов в реакторах BR-2 и DIORIT при линейной мощности до 1300 Вт/см, при температурах оболочек до 600°С и глубинах выгорания до 50 000 МВт·сутки/т. Разработаны методы производства в лабораторном и полупромышленном масштабах таблетизированного и виброуплотненного топлива. В настоящее время сооружается крупный завод. В ходе разработок делались различные оценки и проводились испытания. Для реакторов в Кале и Додеваарде были спроектированы сборки, предназначенные для изучения повторного использования плутония в кипящих реакторах; изготовлено топливо для Гарильянской и Додеваардской атомных электростанций; обеспечено тесное сотрудничество с фирмой ENEL по выполнению экспериментальной программы на Гарильянской атомной электростанции. Для реакторов с водой под давлением изучались различные схемы топливного цикла, охватывающие четыре повторных цикла для установок фирмы SENA и атомной электростанции в Трино-Верчелезе; исследовались границы выключения реакторов и вес регулирующих стержней. С 1963 года плутониевое топливо постепенно вводилось в реактор BR-3; к концу 1970 года максимальная глубина выгорания в сборке с топливом UO_2-PuO_2 достигла 40 000 МВт·сутки/т.

RECICLADO DE PLUTONIO EN LOS REACTORES DE AGUA LIGERA.

El programa belga para el estudio del reciclado del plutonio, iniciado ya en 1958, ha cubierto de forma progresiva la mayor parte de los aspectos de la utilización del plutonio en los reactores de agua ligera. Los estudios económicos se refieren a la disponibilidad de plutonio, a los costes de fabricación y a los debidos al almacenamiento temporal, necesario para beneficiarse con el empleo de grandes instalaciones de fabricación. Se examinan detalladamente las ventajas de usar plutonio en los reactores PWR de la primera generación (vaina de acero inoxidable, barras cruciformes, flexibilidad para una posible conversión en combustible de plutonio) y en los reactores BWR (canales independientes para los elementos combustibles, elementos con enriquecimiento múltiple). Asimismo se estudia, para ambos tipos de reactor, el reciclado del uranio enriquecido obtenido por reelaboración del combustible. La serie de programas de cálculos nucleares PANTHER se ha comprobado sobre los resultados de unos 130 experimentos críticos realizados en la instalación VENUS, empleándose más de 3000 barras de combustible, pastillado o compactado por vibración, fabricadas para este objetivo. Los programas de cálculo termohidráulicos se han calibrado para las condiciones especiales cercanas en la región próxima a los elementos combustibles de plutonio. Los programas de cálculo COMETHE y CRASH, para el proyecto de elementos combustibles, se han comprobado con un número elevado de experimentos de irradiación. El programa de desarrollo del combustible ha comprendido más de 80 probetas irradiadas en los reactores BR2 y DIORIT, empleándose potencias lineales que alcanzaron los 1300 W/cm, temperaturas de vaina de hasta 600°C y grados de quemado de 50 000 MWd/t. Ambas técnicas de fabricación, pastillado y compactación vibratoria, se han desarrollado desde la escala de laboratorio a la de planta piloto.

Está en construcción una gran fábrica. Estas actividades se han complementado con un amplio programa de evaluación y demostración. En el tipo BWR, se han diseñado conjuntos para el reciclado del plutonio, para los reactores de KAHL y DODEWAARD, se ha fabricado combustible para GARIGLIANO y DODEWAARD y hay una íntima colaboración con el programa experimental de ENEL en GARIGLIANO. En el PWR, se han estudiado diversos esquemas de reciclado, que han cubierto cuatro tipos de ciclo para los reactores SENA y TRINO. En estos programas está comprendido el cálculo de los márgenes de apagado y valores para las barras de control. Gradualmente, desde 1963 hasta fin de 1970, se ha ido introduciendo combustible de plutonio en el reactor BR3, alcanzándose un máximo en el grado de quemado de 40 000 MWd/t, en el conjunto combustible UO_2-PuO_2.

1. THE BELGIAN PROGRAM

Since 1959, Belgonucléaire and SCK/CEN have been associated in a 1000 man-year program on all aspects of plutonium recycling in light-water reactors (Fig. 1). The initial phase was partially sponsored by EURATOM.

2. PROSPECTS

Plutonium availability and incentives to recycle have recently been discussed at length [1, 2]. Figure 2 presents revised prospective data on availability, manufacturing cost and market prices. The penalty of direct utilization of plutonium can be calculated on purely economic grounds. Figure 3 results from the assumptions indicated by heavy lines in Fig. 2; recycling is advisable if plutonium sale is not guaranteed within two years in 1973, within one year in 1976 and immediately in the eighties. The decision to recycle or to stockpile until manufacturing costs decrease will also be influenced by the tendency of utilities and fuel manufacturers to minimize risks through realization of prototypes at progressively increasing scales.

3. RECYCLE SCHEMES

The compatibility of fresh plutonium fuel with partially burnt uranium fuel remaining in the reactor is the prime requirement, leading either to an economic penalty or to the adoption of transition cycles.

In equilibrium recycle cores, the water/fuel ratio should be increased to achieve better plutonium utilization and to compensate for control-rod worth decrease; this increase is limited by the associated decrease in temperature coefficients. In existing reactors, the cooling limitations practically impose the coolant cross-section and the fuel rod outer diameter, but the fuel density can be decreased. This improves the technological behaviour of fuel to higher burn-ups. The economic incentive is to increase exposure beyond the enriched uranium optimum, due to the effect of the higher fabrication costs and the constant price of fissile material whatever the plutonium content.

The first generation PWR plants are attractive for plutonium recycle. Their cruciform control rod gaps tend to isolate the new assemblies from their neighbours, allowing a progressive shift from a uranium burner to a plutonium burner or vice-versa (if justified by a plutonium price increase). The so-called 'plutonium island' fuel assembly type (plutonium zone amid enriched uranium surroundings) is recommended, since the shutdown

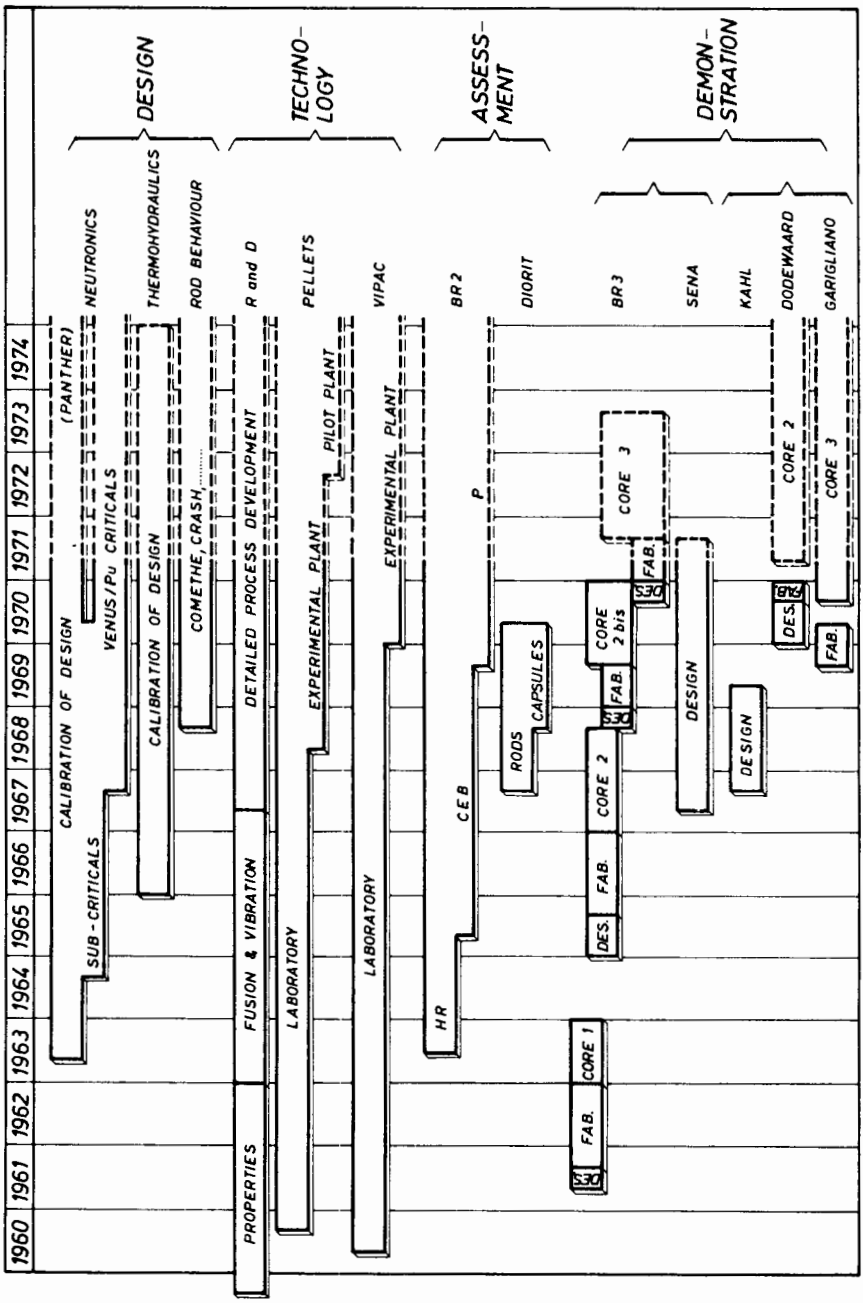

FIG.1. Belgian plutonium recycle program.

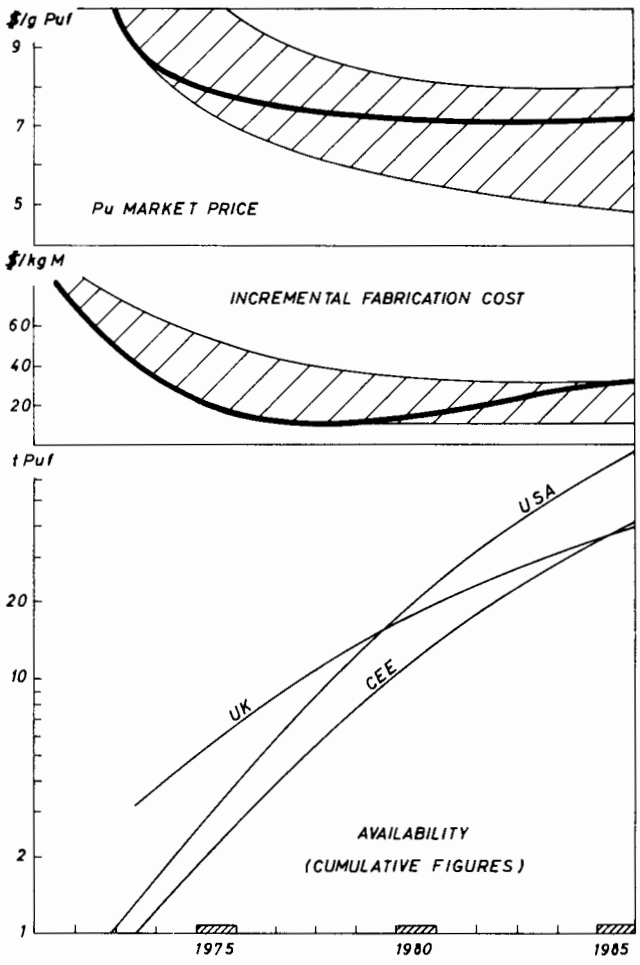

FIG. 2. Prospective data on plutonium.

margins remain practically unaffected; in the SENA case (see section 5.1.2), the control rod worth is decreased by less than 2% relative [3]. Moreover, the relatively high enrichment of stainless steel clad PWRs favours plutonium recycle in these reactors (Fig. 4).

Plutonium utilization in BWRs appears economically less attractive than in PWRs (Fig. 4), but several BWR characteristics favour progressive conversion into plutonium burners:

relatively independent behaviour of the fuel assemblies inside individual shrouds separated by large water gaps (flexibility to adapt the moderation ratio);
practice of power distribution flattening by control rod movements;

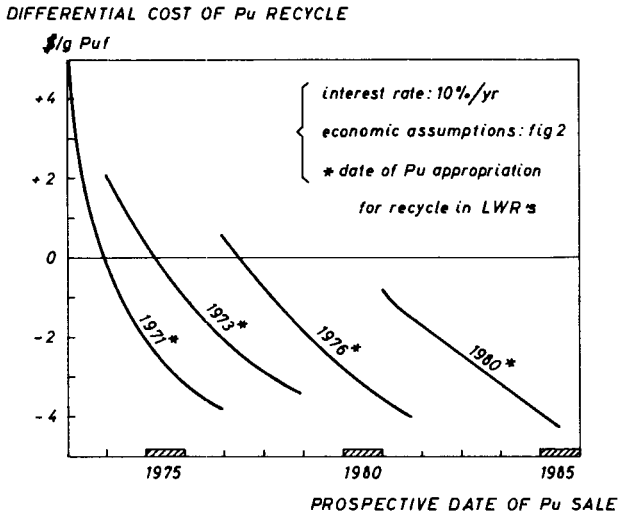

FIG.3. Economic penalty of plutonium recycle versus plutonium stockpiling.

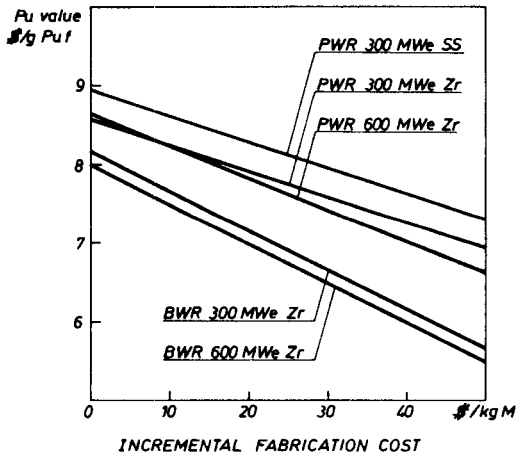

FIG.4. Effect of the fabrication cost on the plutonium value for various power plants.

routine utilization of multiple enrichments within the fuel assemblies, which means a lower relative penalty for plutonium fuels.

Finally, the interest of using uranium recovered from reprocessed first generation PWR fuel has to be stressed for PWRs such as SENA [3]. For BWRs, the peripheral rods of the assemblies can consist of reprocessed uranium from PWRs; for the central rods, it is of interest to blend the plutonium with uranium from gas-cooled reactors or PWRs, if the plutonium price is, respectively, below $12 or $8/g Puf.

4. TECHNICAL BACKGROUND

4.1. Core design

4.1.1. Neutronics

The cell calculation is performed by PANTHER, a proprietary code based on CADILHAC's thermal model, with relatively short computation times [4]. The cross-sections are introduced in one- (PANOPLIE, PENDULE, SQUIRREL, ...), two- (SQUID, SQUID-EREBUS, CONDOR 3, ...) or three-dimensional (TRITON, ...) codes to calculate UO_2 - PuO_2 lattices with the same precision as for UO_2 reactors: 0.5 to 1% on k_{eff}, ±4 to 5% on power distribution, ±10% on control rod reactivity worth. These values have been reached by introducing some modifications in the original calculation procedures. However, for very complicated recycle assemblies, a check on a mock-up case is to be recommended (see section 5.1.1.).

This confidence is based not only on numerous checks on published data and on the BR 3 power reactor experience, but mainly on a large program of neutronic experiments set up at Mol to ensure a systematic design code calibration. The VENUS critical facility, previously utilized as a BR 3/VULCAIN mock-up, was transformed for this purpose. Almost 3000 fuel rods were fabricated, of which more than 1000 contain mixed oxide fuel (Table I); tight fabrication tolerances were imposed to obtain accurate physics data.

The experimental program is summarized in Table I. In a first phase, criticality and substitution tests, as well as boundary and perturbation studies, were carried out in different types of configurations with an unpoisoned H_2O moderator; both integral and fine-structure measurements were included. A second phase was devoted to configurations with a H_2O moderator, poisoned at various boron concentrations. For the high concentrations, a sub-critical technique was used, based on appropriate measurement principles [5] and data analysis methods [6]. The third phase of the program consists of mock-ups of proposed plutonium recycle assemblies for various LWRs (Fig. 5).

4.1.2. Thermohydraulics

Design problems (e.g. burn-out margins) are treated with formalisms appropriate to PWRs and BWRs. The codes were checked on experimental results, including channels with a non-uniform cross-sectional power distribution, typical for an interface between uranium and plutonium fuel.

4.2. Fuel design

4.2.1. Mechanical design

For evaluation of the fuel behaviour and calculation of temperatures, stresses and strains in the various assembly components, two codes were developed: COMETHE for the mechanical and thermal behaviour of fuel under irradiation [7] and CRASH for the calculation of stresses and strains in the cladding [8]. They were checked on many irradiation results, representative of PWRs, BWRs and fast reactors [9].

TABLE I. CRITICAL EXPERIMENTS IN VENUS

Available fuel

Type	^{235}U enrichm.	$\frac{PuO_2}{UO_2 + PuO_2}$	Pu isotopic composition 239/240/241/242	Rods available	
				Stand.	Dismount
4/0	4%	0	-	1760	36
3/1	3%	1.08%	92.08/7.29/0.6/0.03	430	27
2/2.7	2%	2.7%	79.37/17.14/3.05/0.44	470	40
0.7/5	Nat. U	5.04%	79.37/17.14/3.05/0.44	60	5
0.7/4	Nat. U	4.37%	95.65/4.10/0.24/0.001	4	23

A. Basic experimental program

Type of study	Configurations	Characteristics investigated
Criticality tests	Square and/or circular geometry 1 and/or 2 zones 3 pitches 4/0, 3/1, 2/2.7 fuels	Critical mass Axial buckling Radial buckling
Substitution tests	Different Pu fuels in centre of 4/0 (pitch 13 mm) Different fuels in centre of 3/1 (3 pitches)	Reactivity effects
Boundary study	4/0 slab next to 3/1 and 2/2.7 slabs 2/2.7, 0.7/5 and 0.7/4 central square zones in 4/0	Power distribution Power sharing $^{239}Pu_f/^{235}U_f$ and non-fissile detector spectrum indices
Local perturbation study	B_4C, Al, H_2O perturbations in 4/0, 3/1, 2/2.7 ID. at 4/0 - 3/1 boundary	Reactivity effects Power distribution
H_3BO_3 poisoned moderator	4/0 square geometries From 0 to 766 ppm B 462 ppm B: without perturbation with perturbations with central Pu zone From 766 to 2025 ppm B	 Critical mass; B^2 Power distribution Reactivity effects k_{eff} (subcrit. meas.)

B. Mock-up configuration experiments: Fig. 5

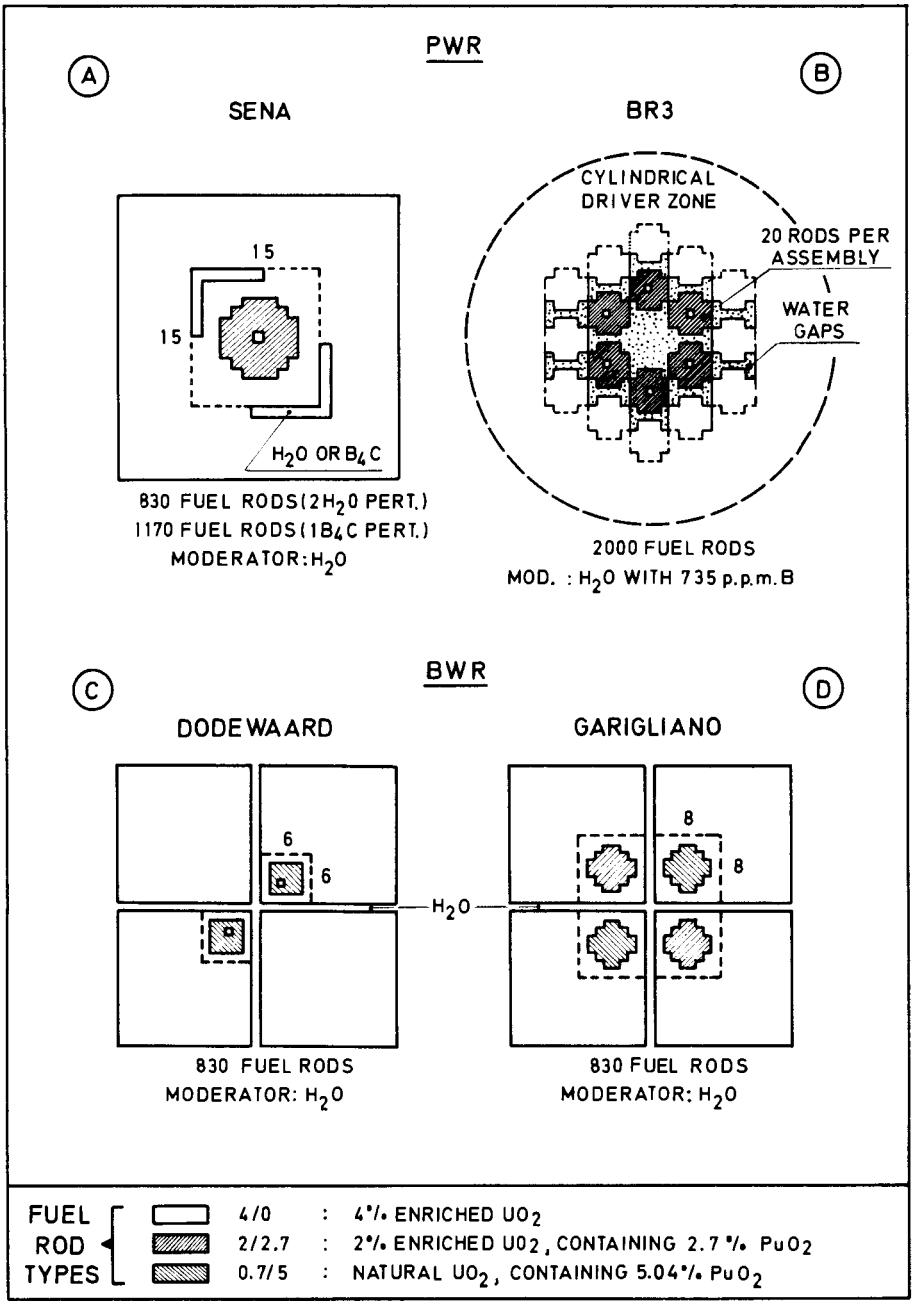

FIG. 5. Plutonium assembly mock-up configurations.

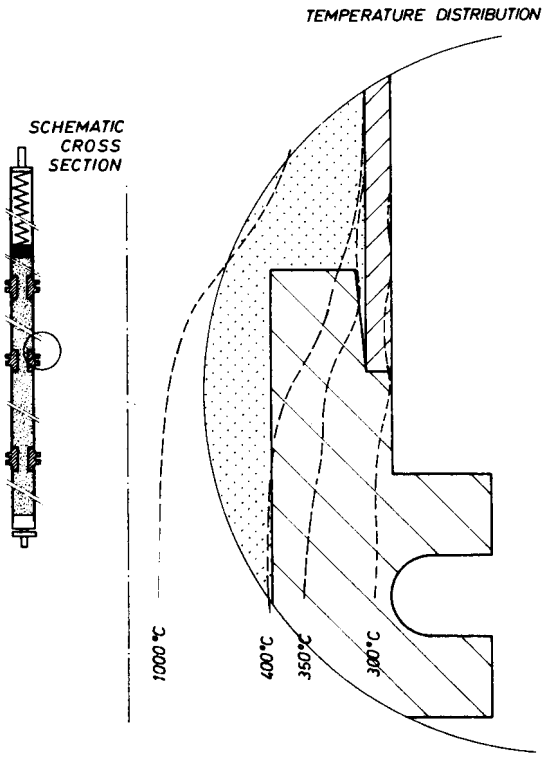

FIG. 6. Plutonium spacer capturing rod.

4.2.2. Particular fuel rods

Some fuel designs developed for enriched uranium fuel were adapted whenever the plutonium manufacturing problems did not allow the use of standard solutions. For BWRs, for example, the spacer capturing rod for positioning the spacer grids has been redesigned to permit its fabrication outside the glove box before filling it with plutonium fuel. A vibro-compacted version is represented in Fig. 6 together with the temperature distribution around the connector plug under unfavourable conditions.

4.2.3. Irradiation experiments

The fuel design has been assessed experimentally through a large-scale irradiation program, mainly performed in the BR 2 testing reactor at Mol. Table II summarizes these experiments, partially reported elsewhere.

The fission cross-section being higher for plutonium than for uranium, the peak ratings are shifted towards the beginning of life, so that the initial thermal fuel behaviour under high ratings is an important characteristic. It has been checked in hydraulic rabbit irradiations (HR) to screen the influence of the fuel specifications and the power history [10, 11].

Burn-up behaviour at normal cladding temperatures is being investigated in boiling water capsules (CEB), which enables the irradiation history to be traced [12, 13]. This is supplemented by basket irradiations (P), with dimensional and visual checks at regular burn-up intervals. Gas release under irradiation has been measured by EIR in the DIORIT reactor, utilizing capsules with continuous pressure monitors.

All these experiments have covered a burn-up range up to 40 000 MWd/t; the program is being pursued for both Zircaloy and stainless-steel claddings up to 60 000 MWd/t.

4.3. Fuel technology

Pellet and vibrocompaction fuel manufacturing techniques are discussed in detail in a companion paper in these Proceedings [14].

The investigation on the sinterability of mixed oxides with low plutonium content was started at the beginning of the program and was concluded in 1963. The work on pellets was resumed in 1967 with the commissioning of a pilot fabrication facility to fulfil the demands for pelleted fuels on a routine basis. A fabrication plant is being constructed to cover industrial scale fabrications from 1973 onwards.

For vibrocompacted fuel, the problem is the manufacturing of high density UO_2-PuO_2 granular material. A special melting technique was developed and applied in an experimental facility over the last five years, proving its ability to reach high yields of powders in correct sieve size ranges. The irradiation of these fuels at high ratings has shown their superiority to pelleted fuel, with regard to fission gas release, swelling and ridging of the cladding [14]. A special vibrocompaction technique, consisting of the introduction of the plutonium in the fine sieve size only (or in the medium and fine sieve sizes), leads to a preferential concentration of the plutonium at the rod periphery and hence to a substantial reduction of the central temperature for a given heat rating. This technique is attractive but requires extensive testing.

5. ASSESSMENT AND DEMONSTRATION PROGRAM

In parallel with the R and D work, gradually expanding design and fabrication activity has been devoted to prototype plutonium assemblies for both PWRs and BWRs.

5.1. PWRs

5.1.1. BR 3

After its successful operation as a PWR (core 1) and a VULCAIN prototype (core 2) [15], the 11.5-MW(e) Belgian BR 3 plant has been turned into a tool for testing new fuel designs and fabrications in power reactor conditions, mainly for the plutonium program (Table III).

The plutonium fuel in core 1 was the first in the world to be loaded into a LWR power plant [16]; this was a purely technological demonstration, calculated with large safety margins.

TABLE II. EXPERIMENTAL IRRADIATIONS ON Pu FUEL MANUFACTURED AT MOL

Facility	BR 2									Diorit	
	HR		P			CEB				Rod	Capsule
Cladding	SS	Zr2	SS		Zr2	Incoloy	SS		Zr2	SS	
O.D. (mm)	16	14.4	9	9.8	15.1	7	7	9.8	15.1	20	16
Thickness (mm)	0.5	0.9	0.5	0.38	0.9	0.5	0.5	0.38	0.9	1.0	0.5
Total length (cm)	15		16-25			15-25				230	29
Number of tests	9	10	2		1	2	5		1	2	5
Number of specimens:											
pellets	-	6	1		2	-	1		3	-	-
Vipac homog.	9	4	1		1	2	4		-	-	6
Vipac heterog.	30	8	4		-	2	7		-	4	4
Cladding temp. (°C)	200		170			400-500	390-600		400	150	210
Heat rating (W/cm)	130-1300	400-800	500-800		500-700	290-400	280-700		500	600-800	700-1000
Burn-up (max.) (GWd/t)	0.06		32		6	16	50		3.4	6	9

TABLE III. IRRADIATIONS OF Pu FUEL IN POWER REACTORS

Reactor	BR 3							Dodewaard	Garigliano
Cladding	SS	SS		Zr4			Zr4	Zr2	Zr2
O.D. (mm)	8.7	8.5		8.7			10.7	13.4	15.1
Thickness (mm)	0.5	0.5		0.6			0.7	0.8	0.9
Total length (m)	1.5	1.2		1.2			1.1	1.8	3.0
Number of Pu rods									
pellets	-	18 [b]		30		240	nd	27	204 (+48 [c])
vibrated	12	19		6		-	nd	3	-
Number of Pu assemblies	1	1		2		8	12	2	4
Core	1	2	2 bis		3			2	
Period	63-64	66-68	69-70		72-73			71-75	70-75
Peak rating (W/cm)	130	210	250	310	350 [a]	400 [a]	500 [a]	500 [a]	400
Burn-up									
average (GWd/tM)	3.4	17	25	13	32 [a]	20 [a]	21 [a]	25 [a]	22 [a]
peak (GWd/tM)	6	31	40	20	50 [a]	38 [a]	36 [a]	30 [a]	30 [a]

[a] Target values.
[b] Manufactured by UKAEA within the Vulcain agreement.
[c] Manufactured by ALKEM.

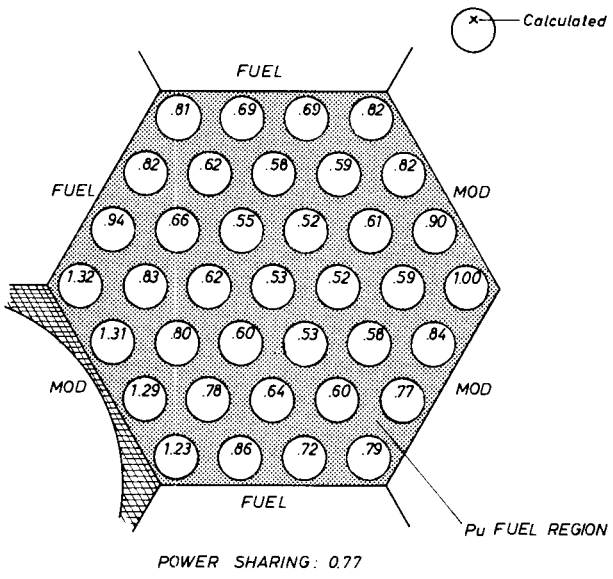

FIG. 7. Power distribution in the plutonium assembly at start BR 3/2.

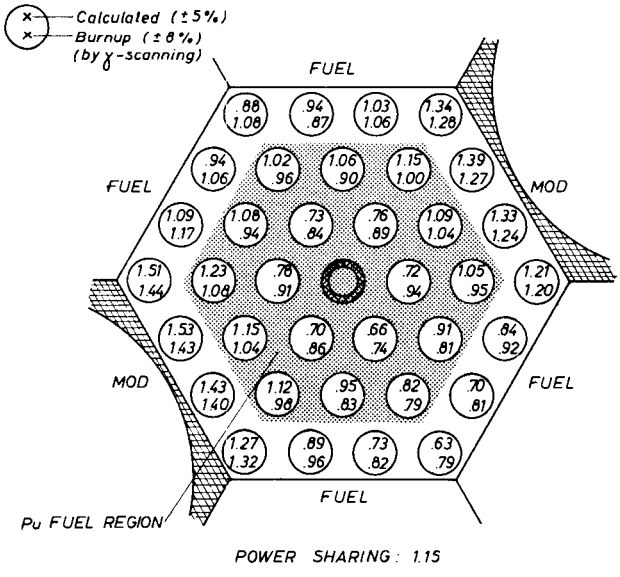

FIG. 8. Power distribution in the 2 new plutonium assemblies in BR 3/2 bis.

The VULCAIN plutonium assembly was also designed with a technological aim, namely, testing the plutonium rod behaviour under high power gradients (Fig. 7), typical of initial recycle situations. The comprehensive thermal and nuclear designs were backed by a full size nuclear simulation in the VENUS facility [17]. Dry-sipping and visual examination indicated good fuel behaviour after a power cycling campaign at the end of core 2 [15], and after core 2 bis, with peak burn-ups typical of the SENA reloads.

The two assemblies introduced into core 2 bis simulate the "plutonium island" concept. Figure 8 compares the burn-up distribution after core 2 bis, measured by gamma-scanning on ^{95}Zr-^{95}Nb, and the calculated power distribution, averaged between start (800 ppm B) and end of life (60 ppm B); it indicates the reliability of the nuclear design codes, even for complicated configurations.

Core 3 will be loaded with fresh hexagonal plutonium assemblies and with new assemblies incorporating 20 large diameter plutonium rods on a square lattice; this complex situation was mocked-up in the VENUS facility (Fig. 5). The core, scheduled to operate by early 1972, will then be about one third composed of plutonium assemblies.

5.1.2. SENA-type reactors

Detailed recycle studies were performed for fist generation PWRs of the SENA and TRINO types, adopting basic assumptions about the plutonium source (the reactor itself), the equilibrium cycle duration (to remain unaffected), the form factors (in respect of DNB margins), and the UO_2 fuel rod enrichment (identical to standard reload fuel)[3]. These studies were made for four cycles (5 to 8), two cladding materials (stainless steel and Zircaloy) and two different reloading schemes: with natural uranium in the mixed oxide rods or slightly enriched uranium reprocessed from unloaded fuel.

With the selection of a "plutonium island" design, it is possible to have adequate form factors with only one plutonium enrichment per reload batch. Figure 9 illustrates, for one of the cases considered (stainless steel cladding,

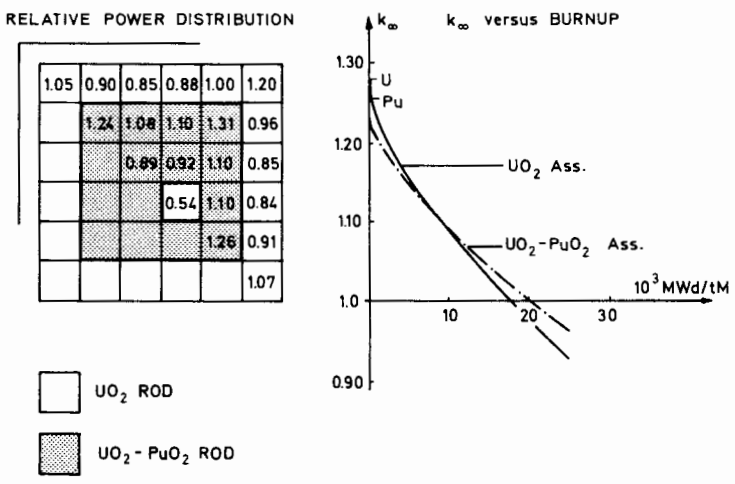

FIG. 9. Power distribution in the SENA reactor during cycle 6.

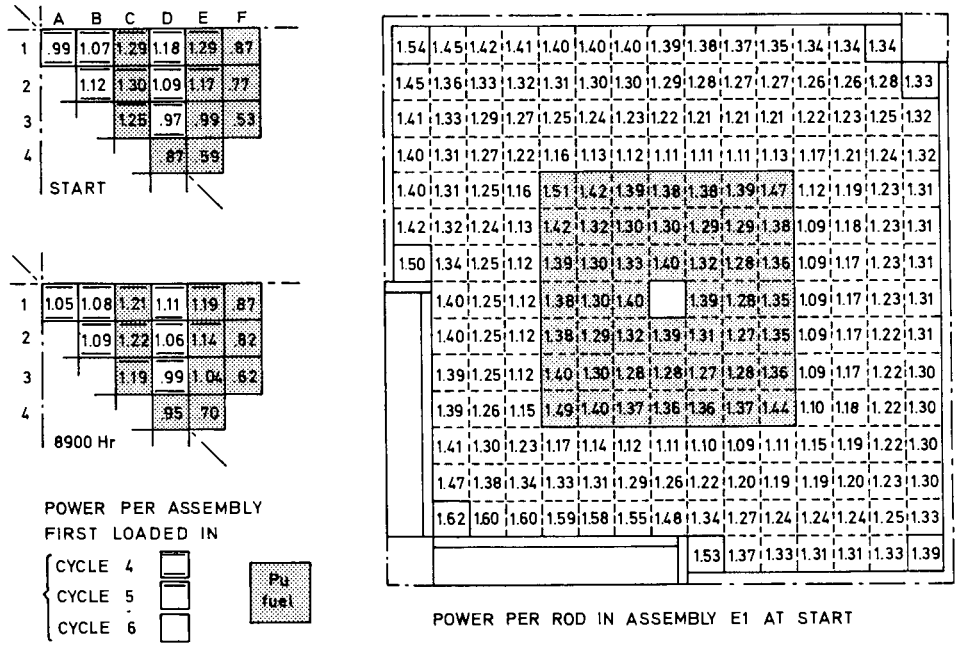

FIG.10. Dodewaard plutonium assembly.

reloading of reprocessed UO_2 of the reactor itself), the power distribution in the core as well as inside the hottest plutonium assembly at the start of the sixth cycle.

Confidence in the calculation tools was acquired by studying a mock-up of a SENA recycle assembly in VENUS (Fig. 5): the power distribution in the case of the two H_2O L-shaped perturbations was calculated within ±4.5% and the reactivity effect calculation of the insertion of B_4C rods showed a satisfactory agreement with the measurement.

5.1.3. RCC reactors

In RCC reactors, the "plutonium island" assembly type loses most of its advantages. The reshuffling schemes must be adapted to minimize burn-up penalties and form factors, during the running-in cycles as a plutonium burner, at the expense of the plutonium value compared with cruciform control rod reactors.

5.2. BWRs

5.2.1. Kahl and Dodewaard

On the basis of the Kahl reactor characteristics, a high performance BWR fuel assembly was designed according to the "plutonium island" principle. The same recycle scheme has been adopted for two assemblies loaded in the Dodewaard reactor in April 1971 (Table III). Figure 10 represents the initial relative power distribution and the reactivity evolution

of an assembly with 15 plutonium pelleted rods and a uranium spacer capturing rod. The second assembly incorporates vibrocompacted rods to reduce the power peaks. To check the design, a mock-up configuration was studied in VENUS (Fig. 5), and cold criticals with the assemblies themselves were performed in the Dodewaard core.

5.2.2. Garigliano

The Joint ALKEM-Belgonucléaire Association manufactured 252 UO_2-PuO_2 fuel rods, assembled in four Garigliano bundles; irradiation started in September 1970 (Table III).

Two Belgonucléaire-SCK/CEN representatives participated in ENEL's plutonium demonstration program in Garigliano on core performance assessments, critical experiments and intermediate examination of irradiated assemblies. Some design activity is under way in Belgium (Fig. 5).

6. ECONOMIC CONSIDERATIONS

The data summarized in sections 4 and 5 show that, technologically, everything is ready for starting the recycle of plutonium when it becomes available in large quantities.

Economically, the present trend towards a large decrease of the plutonium price [18] alone is insufficient to justify recycle, as the incremental fabrication costs are still much too high due to the small plant outputs. However, recycle is strongly favoured by the plutonium storing cost and invested capital charges and by the increase of enriched uranium prices; therefore, the forthcoming availability of relatively large capacity manufacturing plants will finally result in an economic advantage for plutonium recycle.

In the meantime, the construction of increasingly large-scale prototypes of both types of light water reactor seems highly advisable in preparing for the commercial plutonium recycle era; this will minimize the penalty associated with transition cycles and the risks inherent in a complete change to new fuel.

ACKNOWLEDGEMENTS

The authors wish to acknowledge the participation of all those who have enabled this program to be carried out over the last twelve years: the managements of Belgonucléaire and SCK/CEN; the Belgian Government and EURATOM, for their financial contribution and the continuous interest of their representatives; Belgian and Foreign Utilities, for their interest in our work; the general services of our two Organizations, especially our Computer Services and the LMA, Radiation Control and Safety Departments of the SCK/CEN.

The contributions of many colleagues were decisive: for core design and economics – J. Basselier, L. Bindler, H. Bonnet, A. Charlier, P. Deramaix, F. de Waegh, J. Dewandeleer, E. Fossoul, M. Guyette, P. Haubert, N. Hoppe, C. Vandenberg, J. Verraver; for experimental reactor physics – J. Debrue, L. Leenders, F. Motte, W. Rotter,

P. Vandeplas; for fuel development and fabrication – H. Andriessen, A.J. Flipot, I. Lafontaine, J.M. Leblanc, A. Lhost, N. Mostin, P. Papeleux, G. Stroo, E. Vanden Bemden, G. Vanhellemont, B. van Outryve d'Ydewalle; and their technical teams.

REFERENCES

[1] Technical Meeting No.2, International Nuclear Industries Fair, Basle, October 6-11 (1969).
[2] FOSSOUL, E., VANDEN BEMDEN, E., Paper presented at the FORATOM Congress, Stockholm, September 21-23 (1970).
[3] DEBRUE, J., DERAMAIX, P., DE WAEGH, F., Nucl. Appl. Technol. 9 (1970) 516.
[4] BINDLER, L., LEENDERS, L., VAN DEN BROECK, H., Rep. BLG 440/BN 7006-03/EUR 4034 (1970).
[5] VANDEPLAS, P., "Analyse des techniques de mesure de la réactivité d'ensembles multiplicateurs fortement sous-critiques", Fast Reactor Physics (Proc. Symp. Karlsruhe, 1967) 1, IAEA, Vienna (1968) 493.
[6] ROTTER, W., J. nucl. Energ. 23 (1969) 289.
[7] GODESAR, R., GUYETTE, M., HOPPE, N., Nucl. Appl. Technol. 9 (1970) 205.
[8] GUYETTE, M., Nucl. Appl. Technol. 9 (1970) 60.
[9] DEWANDELEER, J. et al., Paper presented at the meeting on Fast Reactor Fuel and Fuel Elements, Karlsruhe, September 28-30 (1970).
[10] LHOST, A., VAN OUTRYVE D'YDEWALLE, B., Rep. BN 6911-02 (1969).
[11] LHOST, A., VAN OUTRYVE D'YDEWALLE, B., Rep. BN 6912-01 (1969).
[12] LHOST, A., VAN OUTRYVE D'YDEWALLE, B., Rep. BN 6902-02 (1969).
[13] LHOST, A., VAN OUTRYVE D'YDEWALLE, B., Rep. BN 6911-01 (1969).
[14] DEJONGHE, P., "Progress in Belgium in nuclear fuel technology", these Proceedings, Paper 275, Vol.4.
[15] POTEMANS, M., "A critical review of operating experience with the BR-3 plant", Performance of Nuclear Power Reactor Components (Proc. Symp. Prague, 1969), IAEA, Vienna (1970) 453.
[16] LHOST, A., BAIRIOT, H., Rep. BN 6806-02/EUR 4004 (1968).
[17] DE WAEGH, F. et al., Plutonium as a Reactor Fuel (Proc. Symp. Brussels, 1967) 1, IAEA, Vienna (1967) 69.
[18] Nucleonics Week 12 3 (1971).

FUEL MANAGEMENT STUDIES FOR PRISMATIC-TYPE HIGH-TEMPERATURE REACTORS

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Abstract–Résumé–Аннотация–Resumen

FUEL MANAGEMENT STUDIES FOR PRISMATIC-TYPE HIGH-TEMPERATURE REACTORS.

The basis of these studies is a realistic prismatic core design proposed by the OECD High Temperature Reactor DRAGON Project for a station of 640 MW(e) with helium cooling and a secondary steam cycle. The core is made up of 264 fuel element columns each consisting of 5 hexagonal fuel blocks. Each block contains compacted coated particle fuel in inside and outside cooled fuel pins. The fuel columns are replaced on-load. The fuel management for the approach to equilibrium and for the equilibrium itself is investigated in this paper. The use of zero-, one- and two-dimensional studies is discussed and the conclusion drawn to which degree of costly core representation one has to go to achieve satisfactory predictions. Typical utility requirements, e.g. electricity demand curves, were the basis of the xenon override capacities and safe shut-down margins incorporated in these studies. The investigations concentrate on low enriched fuel cycles because these are the ones which are considered for the first prismatic HTR power station in Europe. They include fuel cycle cost estimates which assist in deciding which of the investigated running-in approaches should be chosen for real operation. The paper reports the particular points and restrictions which have to be heeded in working out fuel management schemes for HTRs. These include limiting features like fast neutron dose for structural graphite and graphite consolidated fuel compacts, burn-up of coated particle fuel and local power peaks.

ETUDES DE GESTION DU COMBUSTIBLE POUR LES REACTEURS A HAUTE TEMPERATURE DU TYPE PRISMATIQUE.

Les études sont basées sur une conception réaliste d'un cœur de réacteur proposée par l'équipe du projet DRAGON (réacteur à haute température) de l'OCDE, pour une centrale de 640 MW(e) refroidie par hélium avec circuit secondaire de vapeur. Le cœur est fait de 264 colonnes d'éléments combustibles, chacune consistant en un empilement de 5 blocs hexagonaux de combustible. Chaque bloc contient des aiguilles de combustible formées de particules compactées et enrobées, refroidies intérieurement et extérieurement. Le chargement des colonnes de combustible a lieu en marche. Le mémoire étudie l'organisation du cycle du combustible pour la progression vers l'équilibre et l'équilibre lui-même. Des études à zéro, une et deux dimensions sont analysées et on évalue le budget à consacrer à la représentation du cœur pour obtenir une prévision satisfaisante. Le surempoisonnement xénon et les marges sûres d'antiréactivité à l'arrêt, faisant partie de ces études, ont été fondés sur les conditions typiques d'exploitation, telles que les courbes de consommation d'électricité. Les recherches ont été axées sur les cycles des combustibles à faible enrichissement, dont on envisage l'utilisation pour la première centrale de la filière prismatique à haute température à construire en Europe. Ces études comprennent différentes estimations du coût du cycle du combustible qui doivent aider à décider quelle est, parmi les formules de rodage étudiées, celle qu'il faudrait choisir pour l'exploitation réelle. Le mémoire fait état des points particuliers et des restrictions dont il faudra tenir compte pour élaborer les diverses méthodes de gestion du combustible des réacteurs à haute température. On envisage notamment des facteurs limitatifs tels que l'incidence de la dose de neutrons rapides sur le graphite de structure et le combustible compacté au graphite, le taux de combustion des particules de combustible enrobées et les crêtes locales de puissance.

ИССЛЕДОВАНИЯ ПО ОБРАЩЕНИЮ С ТОПЛИВОМ ДЛЯ ВЫСОКОТЕМПЕРАТУРНЫХ РЕАКТОРОВ ПРИЗМАТИЧЕСКОГО ТИПА.

Основой этих исследований является реальная призматическая конструкция активной зоны, предложенная в проекте ОЭСР по высокотемпературному реактору "DRAGON" для станции мощностью 640 МВт(эл) с гелиевым теплоносителем и вторичным паровым циклом.

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Активная зона состоит из 264 каналов топливных элементов, каждый из которых составляется из 5 шестиугольных топливных блоков. Каждый блок содержит охлаждаемые изнутри и снаружи топливные стержни из покрытых и уплотненных частичек топлива. Топливные каналы перегружаются без остановки реактора. В докладе рассматриваются вопросы обращения с топливом для достижения равновесия и саморавновесия. Сравниваются 0, 1- и 2-х мерные исследования, показывающие, каким должно быть в стоимостном отношении представление активной зоны, чтобы достигнуть определенной степени точности. Эти исследования включают также определение типичных потребностей в энергии, например, в соответствии с кривыми спроса, возможности пренебречь образованием ксенона, надежности остановки реактора. Исследования сосредотачиваются на циклах с малообогащенным топливом, которые предусматриваются для первой электростанции в Европе с высокотемпературным призматическим реактором. Они включают стоимостные оценки топливного цикла, используемые при решении вопроса о его выборе в период перехода к нормальной эксплуатации. В докладе подчеркиваются отдельные моменты и ограничения, на которые должно быть обращено внимание при разработке схем обращения с топливом для высокотемпературных реакторов. Сюда относятся и ограничительные особенности, такие, как доза быстрых нейтронов для конструкционного графита и топливных прессовок на графите, выгорание топлива в виде частичек с покрытием и местные энергетические пики.

ESTUDIOS DE GESTION DEL COMBUSTIBLE PARA LOS REACTORES DE ALTA TEMPERATURA Y TIPO PRISMATICO.

Estos estudios se basan en una concepción realista de un núcleo, formado por bloques prismáticos, propuesta dentro del marco del proyecto del reactor de alta temperatura DRAGON, de la OCDE, para el reactor de una central de 600 MW(e), refrigerado por helio y un circuito secundario de vapor. El núcleo está constituido por 264 columnas de elementos combustibles, cada una de las cuales está formada por cinco bloques de combustible hexagonales. A su vez, cada bloque contiene partículas compactadas de combustible, revestidas formando varillas, refrigeradas por dentro y por fuera. La recarga de las columnas de combustible se efectúa con el reactor en funcionamiento. En esta memoria se estudia la organización del ciclo de combustible para alcanzar el equilibrio y el propio equilibrio. Se comparan estudios en cero, una y dos dimensiones y se pone de manifiesto el coste que supone la representación del núcleo si se pretende alcanzar un cierto grado de precisión. De los estudios forma parte un examen de los requisitos típicos de los explotadores, por ejemplo, el seguimiento de la demanda, la capacidad de compensación del xenón y el margen de seguridad después de la caída de barras. Las investigaciones se concentran sobre los ciclos de combustible de bajo enriquecimiento, puesto que éstos son los del tipo considerado para la primera central HTR con elementos prismáticos en Europa. En dichos estudios se presentan diferentes estimaciones del coste del ciclo de combustible, que se utilizan para deducir cuál de los períodos iniciales, hasta la operación normal, examinados debe elegirse para la operación real. Esta memoria subraya los puntos particulares y las restricciones que hay que tener en cuenta para llevar a cabo la gestión del combustible en el caso de los HTR. Entre éstas figuran las limitaciones relativas, por ejemplo, a la dosis de neutrones rápidos en las estructuras de grafito y en el combustible compactado a base de grafito, el grado de quemado de las partículas de combustible revestidas y los máximos locales de potencia.

1. INTRODUCTION

Fuel management in prismatic-type High-Temperature Reactors (HTR's) presents special features, which in the presented design are connected to the on-load refuelling of a core where graphite serves both as moderator and structural material.

The design chosen after optimisation studies [1] is laid out for a power station of 640 MW(e) with downward helium cooling flow and a secondary steam cycle. The fuel cycles aim at an average burnup of 60 GWd/t and foresee ^{235}U enrichments between 4% and 7%.

2. LAYOUT OF FUEL ELEMENT AND CORE DESIGN

The layout of the core is given in Fig. 1. Between the core and the radial fixed graphite reflector structure is a region of removable radial reflector blocks which can be replaced through the outer standpipes.

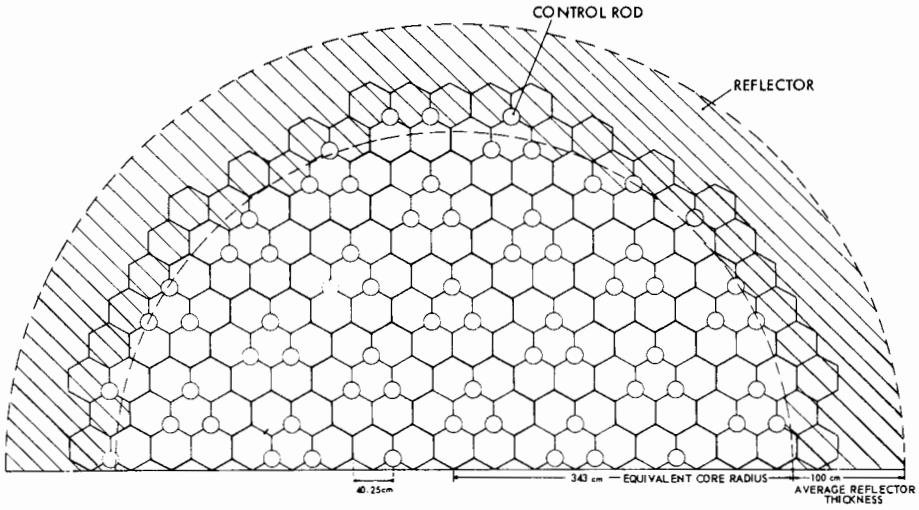

FIG. 1. Core layout.

The main fuel and fuel element design data as well as the reactor data are given in Table I. The fuel consists of coated particles embedded in a graphite matrix compacted into pencil-like fuel sticks. These fuel compacts are inserted into the holes of teledial fuel pins shown in Fig. 2. Each hexagonal fuel block contains, centred around a central handling hole, 17 or 15 fuel channels in which the teledial pins are situated. The pins are internally and externally cooled. Five fuel blocks stacked one on top of the other make a fuel column. Above each fuel column is a top and below a bottom reflector block, the latter resting on the core support grid. The fuel blocks are refuelled by a charge machine which enters the upper plenum chamber through standpipes above the core. Each of the inner standpipes carries a set of three control rods, one black, one grey, and one representing the secondary shut-down system. All control rods are inserted in between the fuel columns.

The fuel blocks for the initial charge contain burnable poison sticks which are inserted in holes near the block edges as shown in Fig. 2.

3. MATERIAL AND ENGINEERING CHARACTERISTICS FOR DRAGON TYPE HTR's

The material limits in Table II refer to the present state of knowledge. Since extensive fuel production programmes and material testing programmes are under way, it can be expected that future results will bring a relaxation in the given limits.

Characteristic for prismatic HTR designs is their gagging of the cooling flow through the various fuel columns to smooth the temperature distribution across the core. In the present design a fixed gagging is installed in the bottom reflector blocks to account for the radial power form factor while variable gagging in the top reflector takes care of the changes of power with burnup.

TABLE I. DESIGN DATA

Total core power	1535 MW
Plant efficiency	41.7%
Net electrical output	640 MW
Core inlet temperature	305°C
Core outlet temperature	785°C
Mixed reactor outlet temperature	750°C
Pressure in primary circuit	56 atm
Core pressure drop	≈0.28 kgf/cm^2
Steam temperature at HP turbine inlet	535°C
Pressure at HP turbine inlet	174 atm
Core height	5.00 m
Representative core diameter	6.83 m
Average core power density	8.29 MW/m^3
Number of fuel columns per core	264
Number of fuel blocks per column	5
Number of standpipes	54
Length of fuel block	100 cm
Fuelled length of fuel block	94 cm
Across flat dimension of hexagonal block	400 mm
Fuel channel diameter	75.8 mm
Coated particle/kernel diameter	1180 μm/800 μm
Kernel material/density	UO_2/8.77 g/cm^3
Burnable poison material	Gd_2O_3/Al_2O_3
Standard heavy metal density in fuel sticks	0.8 g/cm^3
Average carbon to uranium ratio in core	272
Amount of heavy metal in the core	16.9 t

4. ASSUMPTIONS FOR THE FUEL MANAGEMENT INVESTIGATIONS

A catalogue of assumptions is given in Table II. The assumed load factor and xenon override correspond to demand curves of medium or large sized utilities in Europe. The table also contains some cost assumptions which reflect latest estimates.

It is assumed that all fuel blocks of a fuel column are replaced at the same reload and are not reshuffled. The replacement of the initial charge should not start before half a year and later than one year after start-up. The initial reactivity is controlled by a burnable poison.

In all the investigations a 2-zone core layout was chosen to limit local power peaks.

5. INVESTIGATION OF EQUILIBRIUM FUEL MANAGEMENT

5.1. Comparison of zero-, one- and two-dimensional calculations on a selected case

In short, O-D calculations gave a mean feed enrichment which was 1% too high compared with the result from 1-D calculations. For this reason and because of the importance of the radial power distribution in the calculation of the hot channel factor which will be discussed in the next chapter one could obviously conclude that O-D calculations of equilibrium cores at this stage of investigation are unsatisfactory.

Comparing the highest local power peaks in a lattice of fuel elements of various burnup calculated as product of 1-D radial power distributions times the age factors (the ratio of maximum/irradiation average macroscopic fission cross section) with the results from 2-D hexagonal calculations of the whole core, Fig. 3, we found that the power peaks had been underestimated by factors of up to 1.2. The application of correction factors deduced from such comparative calculations, make it possible, however, to return to 1-D calculations and investigate certain features in a rather cheap way.

5.2. Radial power distributions

The equilibrium cores were laid out for equal power bump in inner and outer core regions. Taking this power bump as the product of radial power peak, age factor and correction factor defined above, we studied means of radial power flattening by 1-D methods.

The result of this investigation was the recommendation of an equilibrium cycle with a feed enrichment of 5% and 6% ^{235}U in the inner and outer core zones, respectively, and a constant heavy metal loading of 0.8 g/cm^3 in all fuel sticks throughout the core. The average burnup in both zones was 60 GWd/t. This fuel cycle scheme is referred to as Reference Case 1 and was the target of all approach to equilibrium studies reported in this paper.

Since the core is undermoderated high power peaks and power gradients occur at the core/reflector interface. For the above reference case, temperature calculations accounting for random uncertainties in a 2σ mode showed that the peak fuel temperatures at the core edge would exceed 1400°C. The power gradients would also be rather high (8% over the outer 5 cm of the outermost fuel blocks).

Further studies led to a new reference equilibrium cycle for 60 GWd/t, Reference Case 2, with the same heavy metal loading of 0.8 g/cm^3 in the inner but only 0.64 g/cm^3 in the outer zone and a constant feed enrichment of 5.32% over the core (Fig.3). In this way the edge power peak was reduced by an increase in the moderating ratio of the outer core zone. The hottest channel runs at a power of 1.52 times the average channel (hot channel factor = 1.52) corresponding to a satisfactory peak fuel temperature of less than 1350°C. The power gradient was reduced to 5% over the outer 5 cm of the edge fuel blocks.

5.3. Axial power distributions

The investigations of axial power distributions have first of all shown that it is very important to take the temperature dependence of the cross sections properly into account. For the case with all control rods

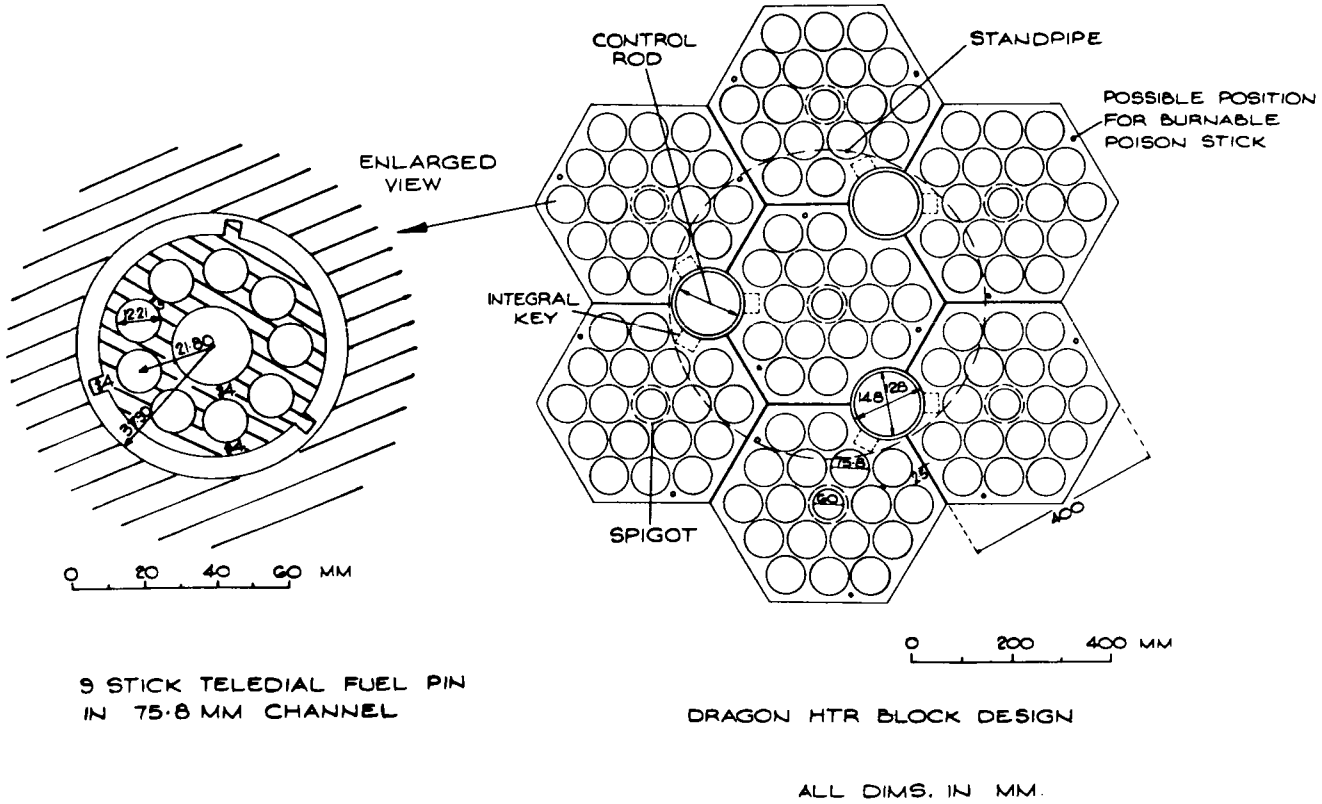

FIG. 2. Fuel pin and block layout.

TABLE II. ASSUMPTIONS

Materials	
Long-term limit on fuel temperature	1350°C
Long-term limit on cooling channel surface temperature	1050°C
Burnup limit	80 GWd/t (≈8.4% FIMA)[a]
Limit on fast neutron dose in fuel	4×10^{21} EDN[b]
Limit on fast neutron dose in reflector blocks	6×10^{21} EDN[b]
Fuel Management	
Average burnup of fuel under equilibrium conditions	60 GWd/t
Xenon override capacity	100%–20% load
Extra reactivity requirement for fuel management	1%
Safe shutdown margin	5% Δk/k
Costs	
Uranium ore cost	8 $/lb U_3O_8
Enrichment service	32 $/kg HM[c]
Fuel element fabrication cost	commercial
Reprocessing cost	130 $/kg HM[c]
Pu credit	10 $/g fissile Pu
Amortisation time	25 years
Interest rate	10% p.a.
Load factor	0.7

[a] FIMA = Fissions per Initial heavy Metal Atom.

[b] EDN = Effective DIDO Nickel Dose (n/cm^2), conventional unit referring to DIDO reactor spectrum.

[c] HM = Heavy Metal.

withdrawn this will result in a power peaking in the cold upper part of the core. With control rods partly inserted the axial power shape will take on a more symmetric appearance.

Fig. 4 shows axial nominal temperature profiles in a central fuel column for half of the grey control rods half inserted. In cases with equal enrichment and heavy metal density in all blocks of a fuel column the limiting fuel temperature peak occurs near the bottom reflector.

Two methods have been studied for lowering this temperature peak: decreasing the heavy metal density and degrading the enrichment in the lower blocks of the core. The adjustments, Table III, were made to equal height of axial temperature peaks (see the curves for 2 enrichments in Fig. 4).

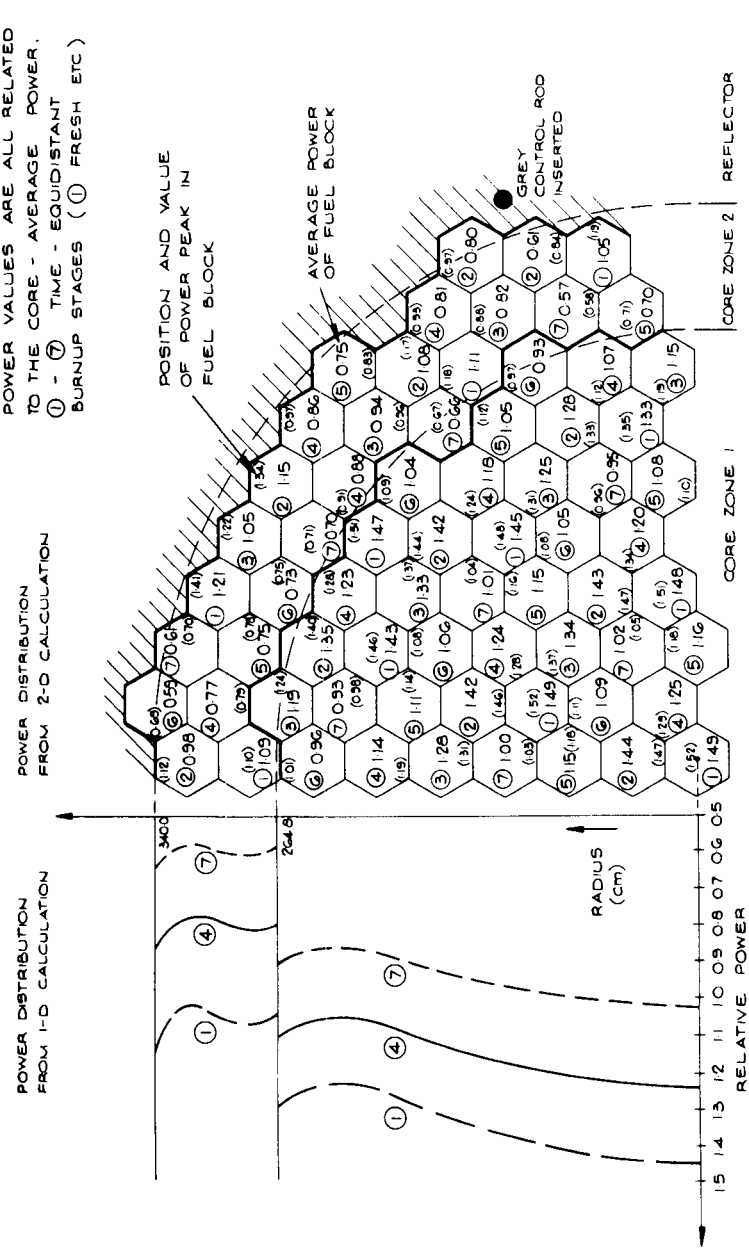

FIG.3. Comparison of calculated 1-D and 2-D power distributions for the equilibrium core (Reference Case 2).

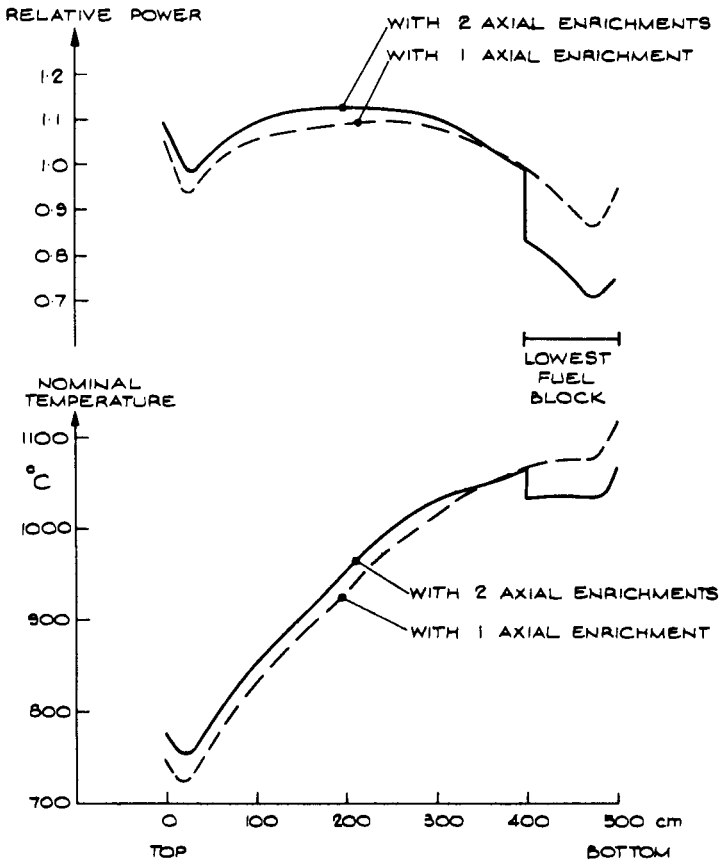

FIG. 4. Axial power and temperature distributions for a fresh central fuel column in the equilibrium core.

The results suggest that the degrading of the enrichment in the lowest block is the most favourable solution, since it brings the nominal peak temperature down by 45°C and at the same time increases the axial peak/average burnup by only 3%.

5.4. Control considerations

Means of controlling the power distribution under normal operation were investigated. It was found that the reactivity step and the local increase in power due to a fuel column change could be amply controlled by one grey control rod in the neighbourhood of the refuelled column (see also Fig. 3).

Calculations on the cold reference cores without xenon showed that the design is safe.

TABLE III. VARIOUS METHODS FOR REDUCING THE PEAK NOMINAL TEMPERATURE (T) AND THEIR EFFECT ON THE AXIAL PEAK/ AVERAGE BURNUP RATIO (Bu) IN A FUEL COLUMN

Case	T (°C)	Bu
All blocks of equal feed composition	1115	1.09
HM-density decreased by 15% in lowest block	1105	1.08
Enrichment decreased by 0.7% in lowest block	1070	1.12
Enrichment decreased by 1.1% in 2 lowest blocks	1065	1.15
Enrichment decreased by 1.8% in 3 lowest blocks	1060	1.27

6. APPROACH TO EQUILIBRIUM FUEL MANAGEMENT

6.1. Discussion of methods

Due to the importance of radial and axial effects it was found necessary to use a 2-D fuel management model in R-Z geometry.

The actual refuelling procedure foresees that each of 264 fuel columns is replaced one at a time. In the fuel management model the simplifying assumption was made that in the central part of the core four columns and in the outer part eight columns at a time were replaced. The fuelling sequence of various core regions was chosen to achieve an evenly spread loading pattern throughout the core. The control rods were represented by homogeneously distributed control poison. In a separate investigation a burnable poison stick was laid out to fit the reactivity requirements of the initial charge. It was found that the control poison simulating the control rods also represents the spectrum effect of the burnable poison.

In these studies axially graded enrichment was not taken into account.

The xenon override capacity was continuously evaluated during the reactor operation as the requirements are very different in the initial core and the equilibrium.

6.2. Layout of the initial core

Economic constraints demand the use of the lowest possible number of different fuel elements. Three initial fuel enrichments and two feed enrichments were regarded as a minimum.

General considerations are to obtain sufficient excess reactivity during the initial, non-refuelling period and to achieve an overall power flattening by matching the enrichments in two radial zones. The fuel columns belonging to one standpipe were made up of blocks with 2-3 different enrichments, the spread of which was chosen according to discharge sequence to give approximately the same utilisation of the fissile material in all elements. Limits were the initial hot channel factor and the maximum burnup.

TABLE IV. FUEL MANAGEMENT DATA FOR DIFFERENT APPROACHES TO EQUILIBRIUM

		Case 1A	Case 1B	Case 1C
Initial Charge:				
Enrichments inner core zone	(%)	3/4/4.5	3/4/4.5	1.5/3/4
Enrichments outer core zone	(%)	4/4/4.5	4/4/4.5	3/3/4
Relative costs of initial charge[a]	(Mio $)	0	0	-2.0
Length of non-refuelling period	(d)	305	308	196
Replacement of First $\frac{1}{3}$ of Initial Charge:				
Feed enrichment inner/ outer core zone	(%)	5/6	4.5/5	4/5
Reloading frequence	(columns/week)	variable	3.2	4
All Following Replacements:				
Feed enrichment inner/ outer core zone	(%)	5/6	5/6	5/6
Reloading frequence	(columns/week)	variable	2	2
Relative fuel cycle costs[a]	(mills/kWh)	0	0.011	0.034

[a] The costs are quoted as differences relative to Case 1A.

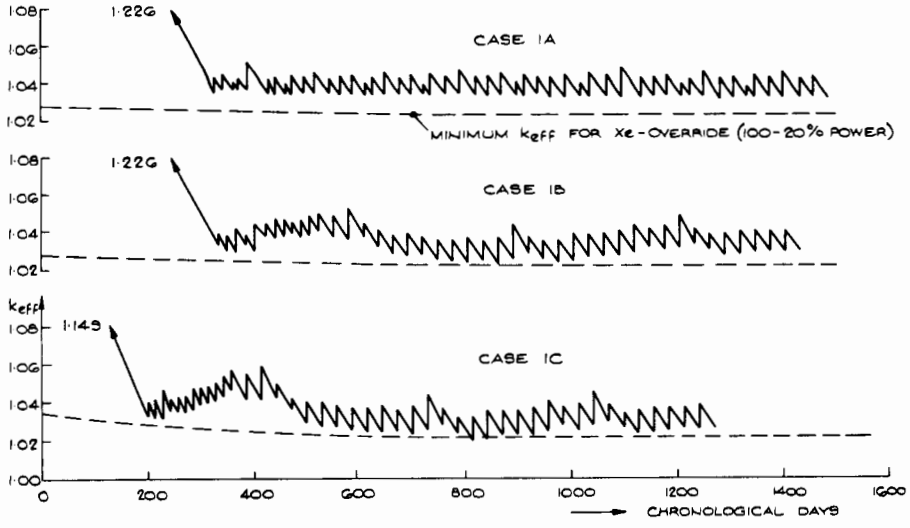

FIG. 5. k_{eff} curves for different approaches to equilibrium.

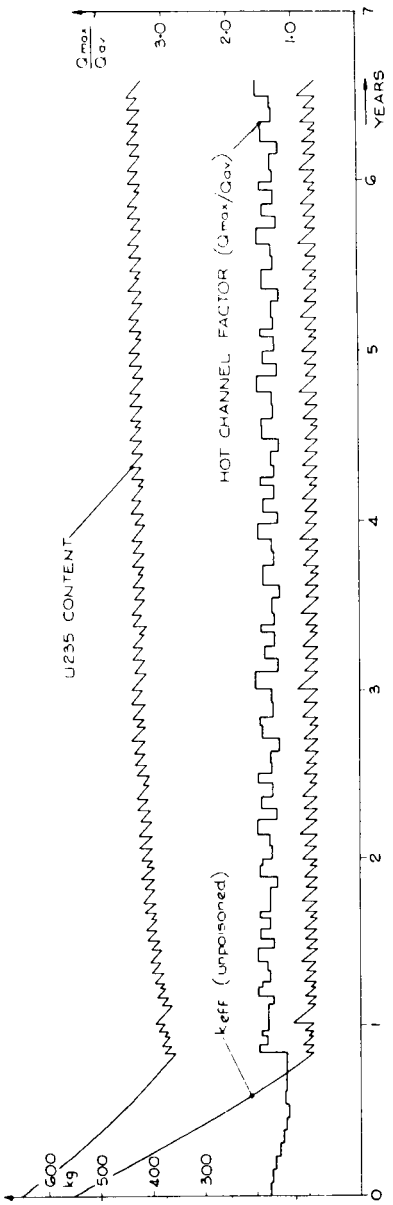

FIG. 6(a). Keff, hot channel factor, and ^{235}U content during the approach to equilibrium (Case 1A).

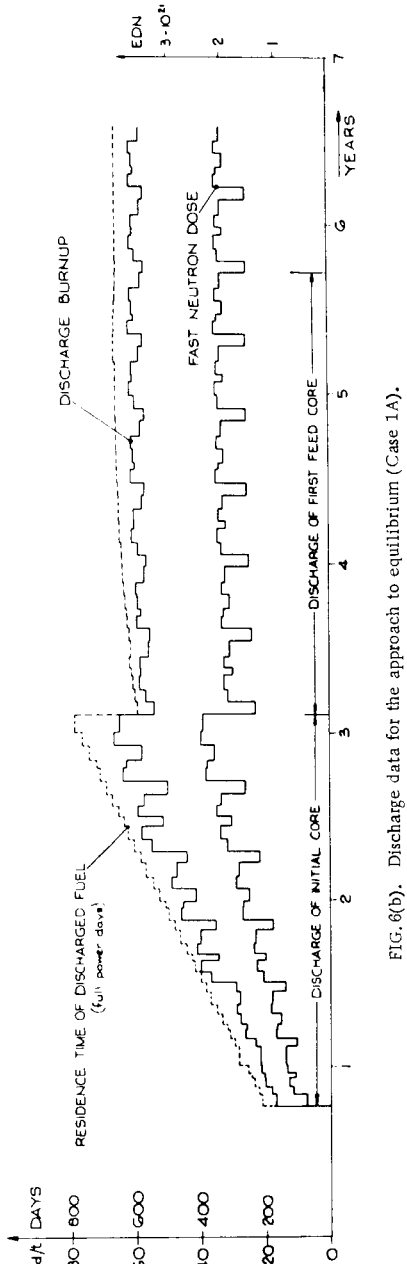

FIG. 6(b). Discharge data for the approach to equilibrium (Case 1A).

Burnable poison sticks containing 0.7 g/cm^3 gadolinium with a diameter of 7 mm were found to control the initial excess reactivity of cases with an average initial enrichment of 4%.

6.3. Approaches to equilibrium

The characteristic data and results for some of the cases investigated are given in Table IV and Fig. 5. The simplest fuel management scheme is 1A where equilibrium feed fuel is already used in the first replacements at the end of the non-refuelling period. The fuelling intervals are automatically adjusted to suit the part-load requirements. Cases 1B and 1C are refuelled at constant intervals using lower than equilibrium enriched intermediate fuel in replacing the first third of the initial charge.

Intermediate fuel lowers the local age factors and leads to a somewhat longer running-in period. Present high fabrication costs outbalance the lower enrichment expenses so that intermediate feed fuel becomes more expensive due to the larger number of blocks needed.

The scheme with the lower initial enrichment, Case 1C, gives higher local age factors during the first refuellings. Furthermore, the softer spectrum and the higher thermal flux require a larger xenon override capacity. Savings due to lower capital investments in the initial charge are wiped out by the increased costs of the necessarily higher replacement frequence in the intermediate period.

Scheme 1A was chosen as a reference case. Some typical characteristics of the approach to equilibrium are shown in Figs. 6(a) and 6(b). The refuelling starts after an initial period of 305 chronological days. During the following 825 days the initial core is replaced by equilibrium feed fuel. This is then followed by a period of 940 days during which the first feed core is replaced by the second one.

The equilibrium phase is reached after the complete discharge of the first core with equilibrium feed fuel. This is an important general outcome of all our investigations. Dependent upon the residence time of the fuel elements the running-in phase will take $\frac{1}{4}$ to $\frac{1}{3}$ of the total reactor lifetime.

The local age factors are higher during the approach than in equilibrium. The radial form factors for the first refuelling intervals, however, are lower due to burnup-flattening during the non-refuelling period. As a combined effect, the hot channel factors during the approach are not higher than in equilibrium. The discharge burnup and the fast neutron dose vary according to the irradiation period and the position of the element in the core. Fig. 6(b) gives the average values for the fuel columns. The peak values are approximately 20% higher. The elements exposed to the limit of their endurance are the ones last discharged from the initial charge. The removable radial reflector blocks have to be replaced every 12 years.

For economic reasons one is tempted to raise the average burnup of the first charge, which in the reference case amounted to 44 GWd/t only. This would require a longer non-refuelling period, a shorter discharge period for the initial core, and, consequently, an intermediate fuel with lower fissile content that can be loaded at a higher frequence without a severe rise in reactivity. Though the initial gain in burnup will tend to be absorbed by the higher fabrication costs per kWh, we regard this as a line for further investigations.

By doing a proper running-in study one arrives at fuel cycle costs which are less than 0.1 mills/kWh higher than if equilibrium was assumed over the whole reactor amortisation period.

7. SUMMARY AND CONCLUSIONS

7.1. It was found that multi-dimensional calculations are essential for proper fuel management studies for HTR's.

7.2. Characteristic power peaks occur at the core/reflector interfaces due to undermoderation. These can be satisfactorily reduced by decreasing the heavy metal density in the fuel columns adjacent to the radial reflector and lowering the feed enrichment in the fuel blocks near the hot axial reflector.

7.3. Burnable poisons are required in the fuel blocks of the initial charge. None, however, are required in the feed fuel.

7.4. It is possible to find a proper fuel management scheme where the technological problems during the approach are not greater than in equilibrium.

7.5. Equilibrium is reached when the first feed core of equilibrium feed fuel composition is completely discharged.

7.6. Evaluating in detail the costs for all refuelling intervals during the approach to equilibrium we found that for a well chosen fuel management scheme the fuel cycle costs are less than 0.1 mills/kWh higher than ones worked out under equilibrium condition. This justifies optimisation studies based on simple fuel cycle assumptions.

REFERENCE

[1] GUTMANN, H., et al., "Parametric Survey on Fuel Cycles and Total Generating Costs for HTR's with Hollow Rod, Teledial, and Tubular Interacting Fuel Elements", Proceedings of the Gas-Cooled Reactor Information Meeting at ORNL, April 1970, USAEC-CONF-700401, p.726.

THEORETICAL METHODS FOR DETERMINATION OF CORE PARAMETERS IN URANIUM-PLUTONIUM LATTICES

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Abstract–Résumé–Аннотация–Resumen

THEORETICAL METHODS FOR DETERMINATION OF CORE PARAMETERS IN URANIUM-PLUTONIUM LATTICES.

The prediction of plutonium production in power reactors depends essentially on how the change of neutron energy spectra in a reactor cell during burn-up is determined. In the epithermal region, where the build-up of plutonium occurs, the slowing down effects are particularly important, whereas, on the other hand, the thermal neutron spectrum is strongly influenced by the low-lying plutonium resonances. For accurate analysis, multi-group numerical methods are required, which, applied to burn-up prediction, are extremely laborious and time consuming even for large computers. This paper contains a comprehensive review of the methods of core parameter determination in the uranium-plutonium lattices developed in Yugoslavia during the last few years. Faced with the problem of using small computers, the authors had to find new approaches combining physical evidence and mathematical elegance. The main feature of these approaches is the tendency to proceed with analytical treatment as far as possible and then to include suitable numerical improvements. With this philosophy, which is generally overlooked when using large computers, fast and reasonably accurate methods were developed. The methods include original means for adequate treatment of neutron spectra and cell geometry effects, especially suitable for U-Pu systems. In particular, procedures based on the energy dependent boundary conditions, the discrete energy representation, the improved collision probabilities and the Green function slowing down solutions were developed and applied. Results obtained with these methods are presented and compared with those of the experiments and those obtained with other methods.

METHODES THEORIQUES DE DETERMINATION DES PARAMETRES DE CŒUR DE RESEAUX URANIUM-PLUTONIUM.

La prévision théorique de la production de plutonium dans un réacteur de puissance est essentiellement fonction de la façon dont on détermine la variation du spectre de neutrons dans la cellule de réacteur au cours de l'irradiation du combustible. Dans la région des neutrons épithermiques, où le plutonium est produit, les effets de ralentissement sont particulièrement importants, tandis que les résonances basses du plutonium influent fortement sur le spectre des neutrons thermiques. Pour une analyse précise, on doit faire appel aux méthodes numériques multi-groupes qui deviennent extrêmement laborieuses et prennent beaucoup de temps lorsqu'on calcule le taux d'irradiation, même à l'aide de grands ordinateurs. Le mémoire étudie de manière approfondie les méthodes élaborées en Yougoslavie au cours de ces dernières années pour le calcul des paramètres de cœur d'un réseau uranium-plutonium. Ne disposant que de petits ordinateurs, les auteurs ont cherché à trouver de nouvelles méthodes combinant les données physiques disponibles et des moyens mathématiques appropriés. La caractéristique principale de ces méthodes est de pousser le traitement analytique le plus loin possible et d'y inclure des améliorations numériques pertinentes. Grâce à cette tactique, généralement négligée par les utilisateurs des grands ordinateurs, des méthodes rapides et suffisamment précises ont pu être élaborées. Celles-ci, particulièrement adaptées aux systèmes U-Pu, contiennent des techniques originales pour le traitement de l'influence du spectre de neutrons et de la géométrie des cellules. Les méthodes élaborées portent notamment sur les conditions aux limites variant avec l'énergie, la représentation discontinue de l'énergie, les probabilités de choc améliorées et le calcul du ralentissement à l'aide des fonctions de Green. Les résultats obtenus par ces méthodes sont présentés et comparés avec l'expérience et avec les résultats obtenus à l'aide d'autres méthodes.

ТЕОРЕТИЧЕСКИЕ МЕТОДЫ ОПРЕДЕЛЕНИЯ ПАРАМЕТРОВ АКТИВНОЙ ЗОНЫ, СОСТОЯЩЕЙ ИЗ УРАН-ПЛУТОНИЕВЫХ РЕШЕТОК.

Расчет производства плутония в энергетических реакторах существенно зависит от того, как изменяется энергетический спектр нейтронов в ячейке реактора в процессе выго-

рания. Накопление плутония идет, в основном, в эпитепловой области энергий, где особенно важны эффекты замедления, тогда как, с другой стороны, резонансы плутония в области низких энергий оказывают заметное влияние на тепловой спектр. Для точного анализа требуется использовать многогрупповые численные методы расчетов, которые в применении к расчетам выгорания особенно трудоемки и занимают много времени даже при работе на крупных электронно-вычислительных машинах. Доклад содержит всеобъемлющий обзор методов определения параметров активной зоны, состоящей из уран-плутониевых решеток. Указанные методы были разработаны в Югославии за последние несколько лет. Оказавшись перед фактом использования небольших вычислительных машин, авторы должны были искать новые пути решения, сочетающие физическую наглядность и математическое изящество. Основная особенность этих решений – аналитическая обработка результатов с использованием соответствующих усовершенствований численных методов расчета. В результате были разработаны сравнительно нетрудоемкие методы, обеспечивающие разумную точность расчета. Разработаны оригинальные методы обработки нейтронных спектров и эффектов геометрии ячейки, в основном применительно к уран-плутониевым системам. В частности, были разработаны и применены методики с использованием энергетической зависимости граничных условий; дискретного энергетического представления, решения функции Грина для процесса замедления. Приводятся результаты, полученные с использованием разработанных методов. Эти результаты сравниваются с экспериментальными данными и результатами других методов.

METODOS TEORICOS PARA LA DETERMINACION DE LOS PARAMETROS DEL NUCLEO EN RETICULOS DE URANIO-PLUTONIO.

La predicción de la producción de plutonio en los reactores de potencia depende esencialmente de cómo se determine la variación del espectro energético en una celda del mismo durante la irradiación del combustible. En la región epitérmica en la que tiene lugar la acumulación del plutonio, son particularmente importantes los efectos de la moderación, mientras que por otra parte el espectro neutrónico térmico se ve considerablemente influido por las resonancias bajas del plutonio. Para un análisis preciso se necesitan métodos numéricos en multigrupos, los cuales, cuando se aplican a la predicción del grado de quemado, son extremadamente laboriosos y consumen mucho tiempo incluso para máquinas calculadoras grandes. Esta memoria contiene una amplia reseña de los métodos desarrollados en Yugoslavia durante los recientes últimos años para la determinación de parámetros del núcleo en retículos de uranio-plutonio. Enfrentados con el problema que supone contar únicamente con máquinas calculadoras pequeñas, fue preciso que los autores encontraran nuevos métodos en los que la evidencia física se combina con la elegancia matemática. La característica principal de estos métodos es la tendencia a seguir un procedimiento analítico hasta donde sea posible para introducir entonces adecuados recursos numéricos. Siguiendo esta línea de pensamiento, que generalmente se pasa por alto cuando se utilizan máquinas calculadoras grandes, se desarrollaron métodos rápidos y de precisión razonable. Los métodos utilizados comprenden técnicas originales para tratar convenientemente los espectros neutrónicos y efectos de la geometría de la celda, y son especialmente idóneos para sistemas U-Pu. En particular fueron desarrollados y aplicados procedimientos basados en las condiciones de contorno dependientes de la energía, la representación discreta de la energía, las probabilidades de colisión mejoradas y el cálculo de la moderación mediante funciones de Green. Se presentan los resultados obtenidos con estos métodos y se comparan con los de la experimentación y con los que resultan de aplicar otros métodos.

1. INTRODUCTION

The prediction of plutonium production in power reactors depends essentially on how the change of neutron energy spectra in a reactor cell during burn-up is determined. In the epithermal region, where the build-up of plutonium occurs, the slowing down effects are particularly important, while, on the other hand, the thermal-neutron spectrum is strongly influenced by the low-lying plutonium resonances. For accurate analysis, multi-group numerical methods are required, which, applied to burn-up prediction, are extremely laborious and time-consuming even for large computers.

There are several plutonium recycling modes, and the decision on which is best for the specific reactor in which plutonium is to be used depends primarily on the detailed nuclear design studies. However, it is usually the case that calculational schemes, used successfully for core parameter determination in UO_2 or metallic uranium lattices, result in systematic discrepancies, when extended and applied to U-Pu lattices, so that semi-empirical nuclear data adjustments are necessary. These discrepancies are especially sensitive to the changes in ^{240}Pu concentration and the lattice pitch, i.e. to thermalization and transport effects.

As is known, plutonium cores, compared with uranium cores, are expected to show improved stability and transient characteristics, because they have more negative Doppler and moderator temperature coefficients due to plutonium resonances, increased resonance absorption and spectrum shift. On the other hand, the control-rod efficiency is reduced as a result of thermal flux reduction, which necessitates an increased number of control rods. Moreover, there is a reactivity penalty caused by intensified resonance effects and yield of fission product poisons. Because of all these effects, more elaborate methods are required for U-Pu lattice calculation, especially for prediction of adequate reactivity and power distribution control.

This paper contains a comprehensive review of the methods for core parameter determination in U-Pu lattices developed in Yugoslavia during the last few years. Faced with the problem of using small computers, the authors had to look for new approaches combining physical evidence and mathematical elegance. The main feature of these approaches is the tendency to proceed with analytical treatment as far as possible and then to include suitable numerical improvements. With this philosophy, which is often overlooked when using large computers, fast and reasonably accurate methods have been obtained. They contain original developments for adequate treatment of neutron spectra, cell geometry and core composition effects, especially suitable for U-Pu systems.

2. LATTICE CELLS

2.1. Thermal-neutron region

Thermal-neutron spectra in plutonium-containing reactor lattice cells are strongly influenced by the 0.297-eV resonance of ^{239}Pu and the 1.055-eV resonance of ^{240}Pu. To treat adequately both thermalization and resonance effects, detailed multi-group calculation is usually required. In the course of Swedish-Yugoslav co-operation, a new idea was developed [1] to replace the standard multi-group method by discrete representation of the neutron spectra according to the Gaussian quadrature scheme. The accuracy of the plutonium reaction rate integration was improved by introducing different integration variables in different parts of the thermal-energy region and clustering the integration points around the plutonium resonance [2]. Furthermore, the convergence of the method, as regards the number of integration points, was improved by splitting the flux under the energy transfer integral into a Maxwellian part and a hardening part with an energy dependent amplitude [3]. When the preliminary guess of the hardening function is good, this approach can be fairly efficient, since only the interpolation in the hardening function amplitude is to be performed.

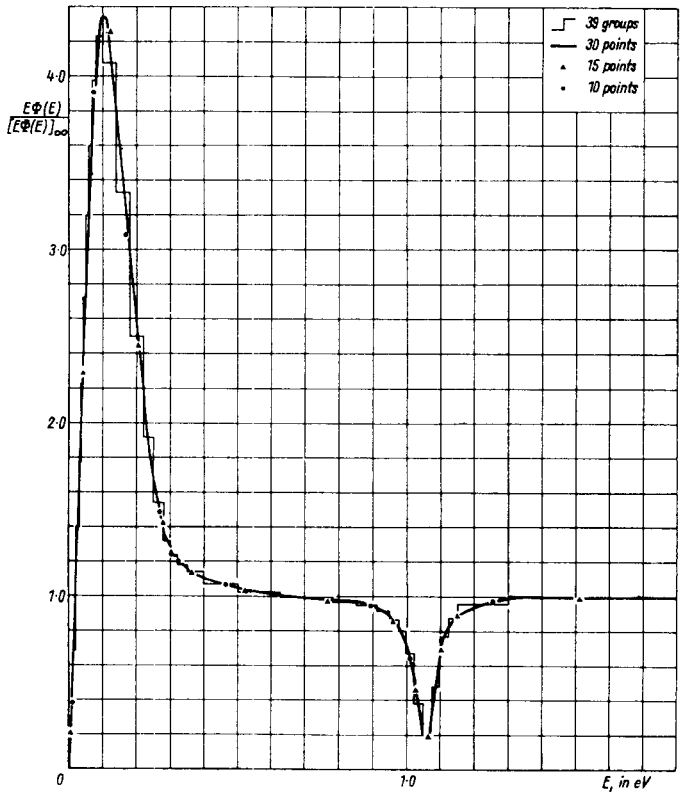

FIG. 1. Thermal-neutron spectrum in fuel of a two-region reactor cell: fuel radius, 0.75 cm; cell radius, 2.25 cm; temperature, 500 °K; ^{239}Pu and ^{240}Pu enrichment, 0.1%.

A system of multi-point equations, formally identical to multi-group equations, was derived [4]. The applicability of the energy point method for heterogeneous systems was examined by combining it with the collision probability technique. The routines PIN and SOMAT [5] were written for calculating thermal neutron spectra and reaction rates in a two-zone Wigner-Seitz cell. Both multi-point and multi-group calculations were performed for a number of different cases, the typical examples being illustrated by Figs 1 and 2. The number of energy points needed to achieve the pre-assigned accuracy of reaction rate integration was shown to be considerably lower than the corresponding number of energy groups.

To study the thermalization effects in a reactor lattice cell, the P_3 approximation of the spherical harmonics method was extensively used and a method was suggested for efficient solution of multi-group or multi-point P_3 equations [6]. A computer program, MULTI, was developed [7] to calculate the space and energy distribution of thermal neutrons in a multi-zone cylindrical lattice cell, using either the multi-group data calculated by program SIGMA or the multi-point data computed by program CAMP [8].

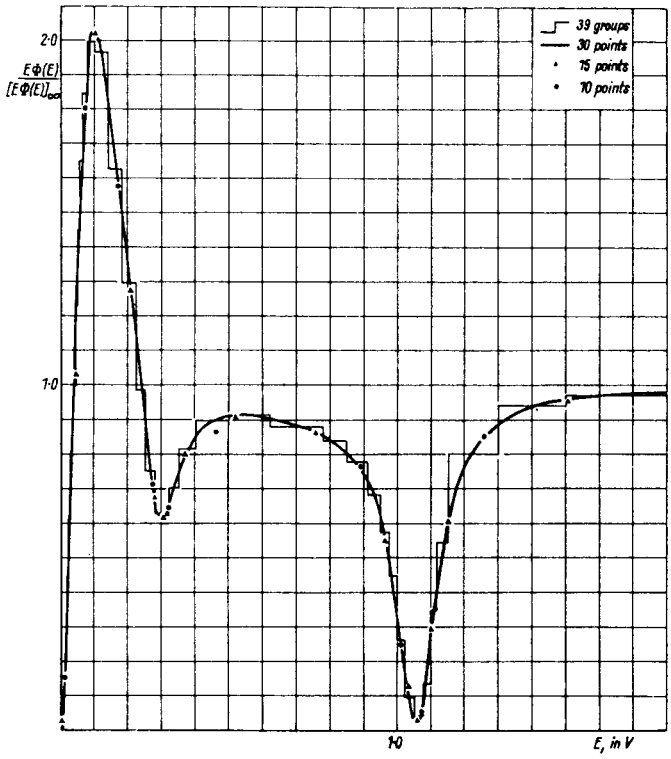

FIG. 2. Thermal-neutron spectrum in fuel of a two-region reactor cell: fuel radius, 0.75 cm; cell radius, 3 cm; temperature, 500 °K; ^{239}Pu and ^{240}Pu enrichment, 0.5%.

In the program CAMP, a procedure was developed for the point scattering matrix construction based on the Lagrange interpolation scheme. Instead of splitting the neutron flux under the energy transfer integral into the Maxwellian part and hardening part, it was approximated by a third degree polynomial with coefficients depending on the flux values at the four neighbouring integration points. The total balance condition is satisfied through the iterative procedure of solving the system of multi-point P_3 equations, which makes this approach much simpler and more straightforward than that suggested in Ref. [1].

Another improvement of the point method was obtained by more exact integration of neutron absorption. Instead of using the Maxwellian amplitude [1] or the iterative procedure applied in Ref. [8], the neutron balance condition was obtained by a more convenient distribution of energy points. The slightly increased thermal energy interval was divided into four subintervals according to the physical properties of U and Pu isotopes and a suitable integration procedure was proposed for each interval. The same integration functions were used for determination of scattering matrices.

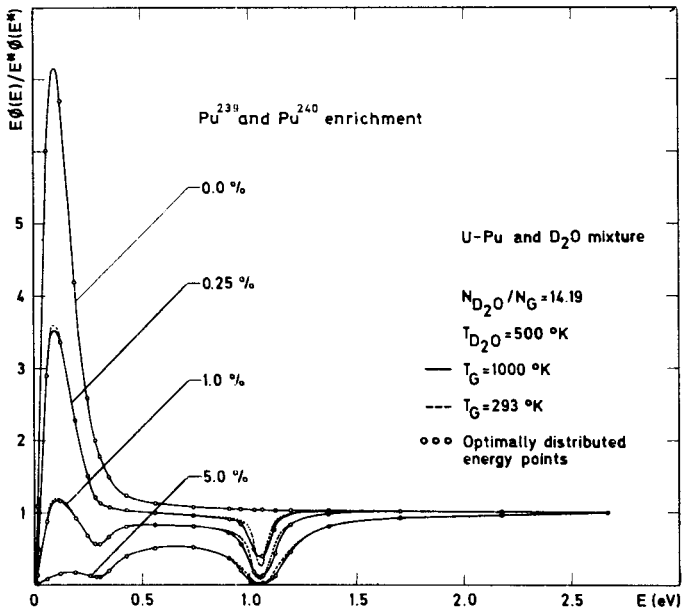

FIG. 3. Neutron energy spectra for different plutonium enrichments.

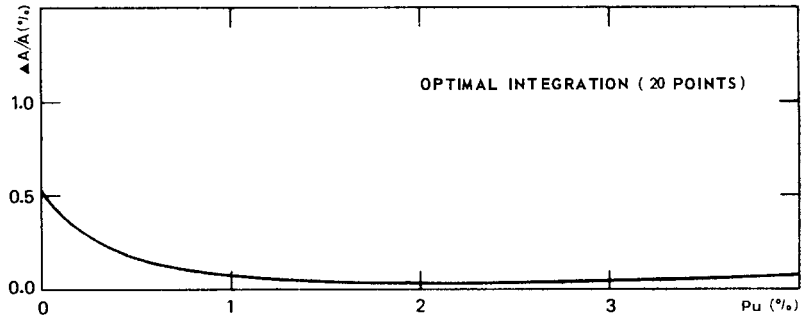

FIG. 4. Accuracy of the reaction rate calculation as function of the plutonium concentration.

The Doppler broadening of Pu resonances was also taken into account. In this procedure, a thermalization program PLUTON was developed and some of the results obtained are presented in Figs 3 and 4.

To take into account the space variation of the thermal flux inside the cell, an investigation was carried out on connecting the collision probability treatment with the energy point method. Starting from a collision probability study for concentric cylindrical zones [9], a corresponding space point method (based on the combination of analytical and numerical approaches) was developed [10]. For every chosen space point, the integral transport equation was written and the flux between the points was interpolated by a

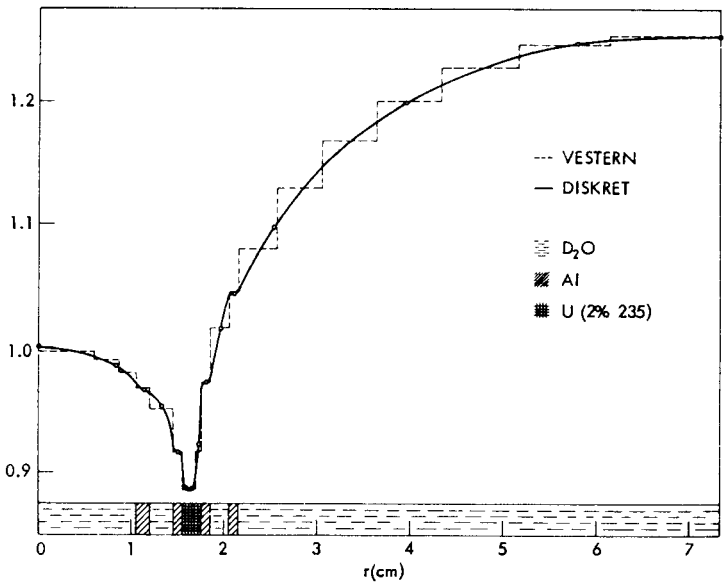

FIG. 5. Thermal-neutron flux distribution in RA reactor cell (VESTERN and DISKRET computer programs).

series in even powers of the radius. The system of linear algebraic equations obtained gives the flux at the desired points. In Fig. 5 some of the results obtained by the method described are compared with the standard collision probability treatment. The application of the method to multi-group calculations is straightforward, as shown in Ref. [11]. The method is also adapted to cluster-type fuel elements [12].

Another rapid and reasonably accurate procedure suitable for Pu lattice calculation is the program ANTER, based on the analytical method for calculating neutron thermalization [13, 14]. In this method, use is made of the energy-dependent boundary conditions combined with the collision probability technique in the fuel and the Laguerre polynominal expansion in the moderator [15]. In many burn-up programs, for instance in the Swedish BOP and the Yugoslav TER [16], the one-velocity ABH procedure [17] is used for disadvantage factor calculation. The accuracy of such programs may be improved by the substitution of ABH by ANTER, since the latter has been shown to give considerably better results [18]. Some comparisons of both methods with the THERMOS procedure and experiments can be seen in Fig. 6.

2.2. Resonance neutron region

In treating the resonance effects in a reactor cell, the usual assumptions are: a spatially flat flux in every cell zone and an asymptotic 1/E spectrum between the resonances. As a result, a number of semi-empirical corrections are necessary. In addition, an adequate connection with the usually

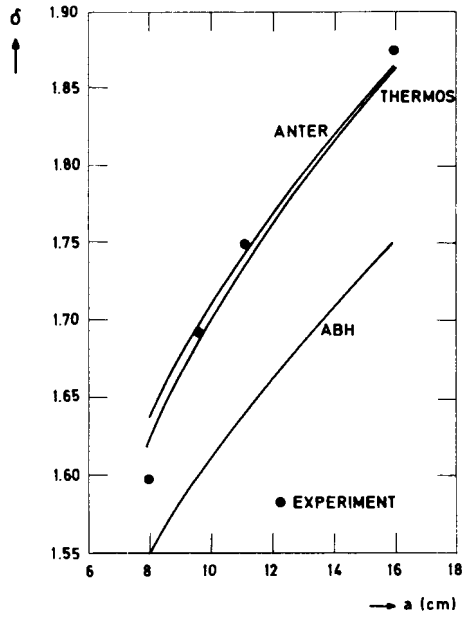

FIG. 6. Cell disadvantage factors for different lattice pitches of RB reactor cell.

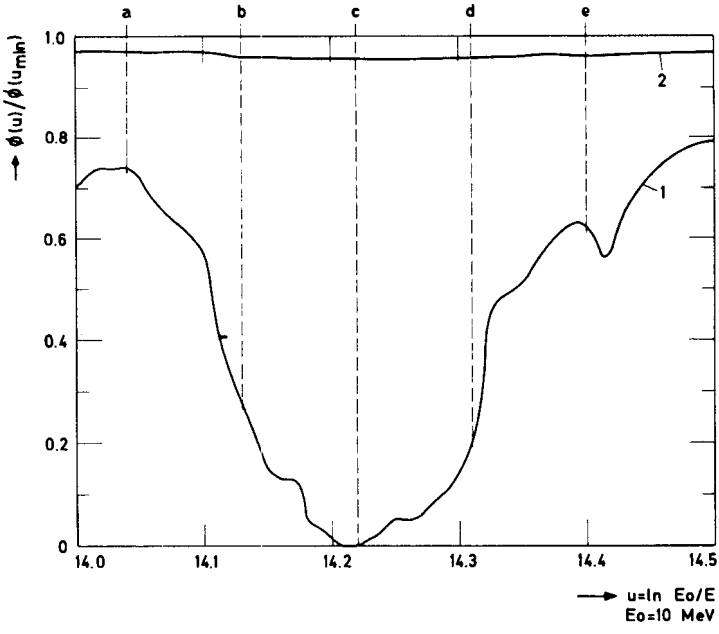

FIG. 7. Lethargy distribution of the average flux in fuel (1) and the average flux in moderator (2) in the vicinity of the 6.7-eV resonance of ^{238}U.

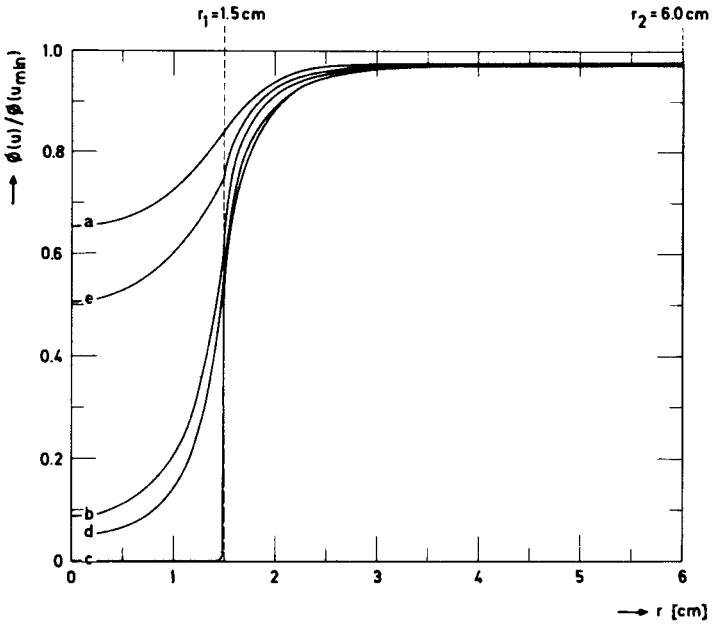

FIG. 8. Space distribution of resonance-neutron flux for the energies denoted by a-e in Fig. 7.

detailed treatment of the thermal neutron region is impossible. Moreover, more rigorous treatment of the resonance region is particularly important for adequate Pu build-up prediction.

To overcome these shortcomings, a new procedure was developed to solve the space-, lethargy- and angle-dependent transport equation for resonance neutrons. In the resonance region, the neutron scattering is isotropic, but, since the values of cross-sections in different zones of a reactor cell are widely different, the neutron flux near the zone boundaries becomes very anisotropic. For this reason, the P_3 approximation of the spherical harmonics method was applied, resulting in a system of integro-differential equations with a difference kernel depending on lethargy and space. Combining the analytical and numerical procedures, the expressions are obtained for the space and energy distributions of resonance neutrons in a multi-zone reactor cell [19]. The accuracy is limited practically only by input cross-section data, and the computing time is relatively short, so that it is convenient for incorporation in burn-up programs. Some results for resonance-neutron flux distributions in the centre of a fuel element and at its surface are shown in Figs 7 and 8. To investigate in detail the effects of ^{238}U resonances (where the build-up of plutonium occurs) the flux distributions near a giant 6.7-eV resonance and the corresponding space and lethargy distribution in a cell are calculated and presented in Figs 9 and 10, respectively.

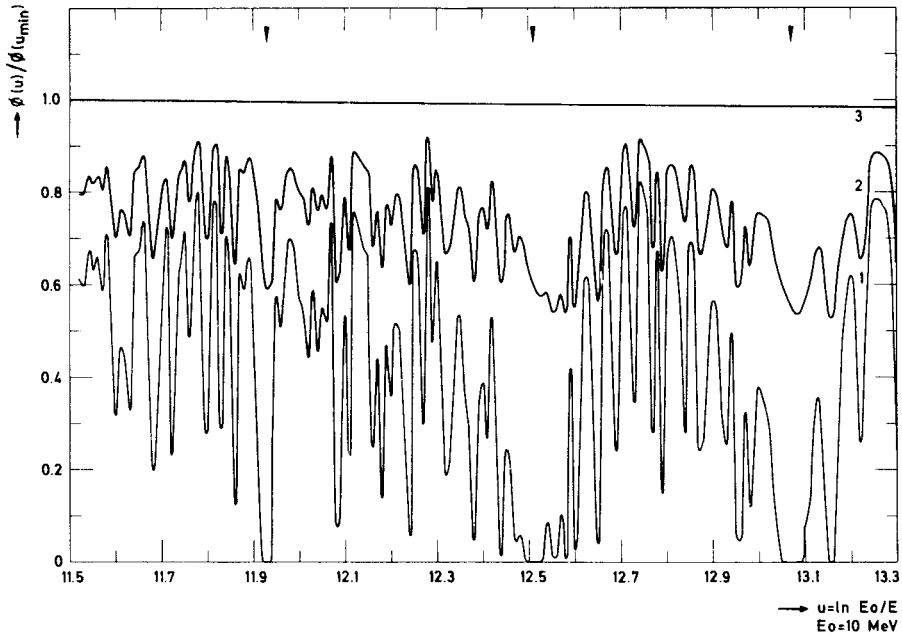

FIG. 9. Lethargy distribution of resonance-neutron flux in the centre of fuel element (1), at the surface of fuel element (2) and at the surface of the cell (3), in the interval 10 eV - 100 eV.

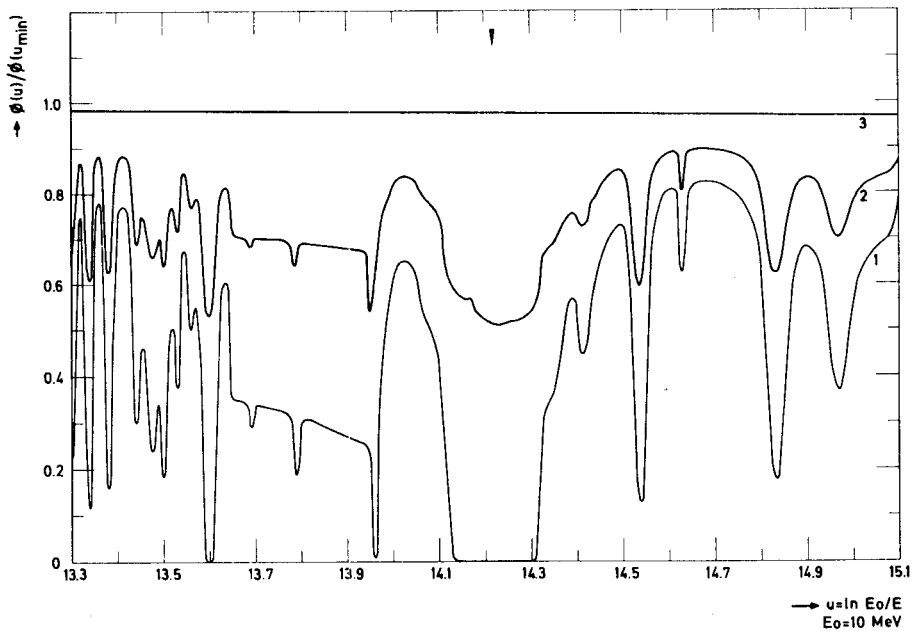

FIG. 10. Lethargy distribution of resonance-neutron flux in the centre of fuel element (1), at the surface of fuel element (2) and at the surface of the cell (3), in the interval 1 eV - 10 eV.

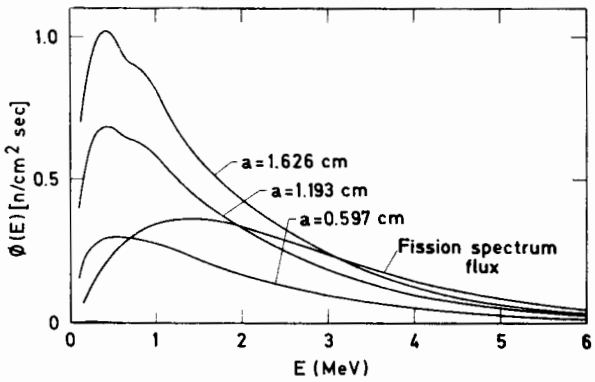

FIG. 11. Energy distribution of the fast-neutron flux in a natural uranium fuel element.

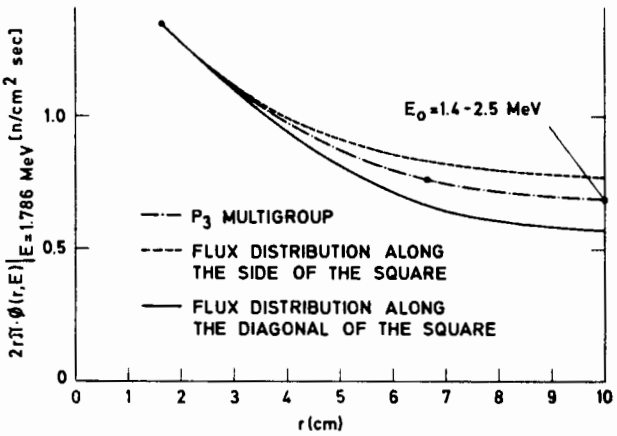

FIG. 12. Space distribution of the fast-neutron flux in the moderator of a natural uranium - D_2O cell, taking into account anisotropic elastic scattering: rod diameter, 1.626 cm; lattice pitch, 17.7 cm.

2.3. Fast-neutron region

The fast region is relevant to the problem of Pu build-up, mostly through supplying adequate sources for resonance neutrons. Here the problem of fast-neutron transport and slowing down becomes important. An exact analytical method for solving the problem of the slowing down of fast neutrons was developed recently [20]. The solution of the slowing down equation was obtained indirectly, by detailed consideration of the neutron slowing-down process. By representing the collision density by a sum of collisions with different prior histories and substituting Green functions by Dirac delta functions, the problem was reduced to determination of neutron scattering functions. These functions were determined by factorization in lethargies (in treating the inelastic scattering) as well as by expansions in cosine of the scattering angle (in treating the elastic scattering). The expression obtained for collision density is valid for arbitrary dependence of cross-sections on energy and angle.

With the Green function technique, the method was applied to a reactor lattice cell [21]. The space energy distribution of fast neutrons in a reactor cell was also determined by the multi-group P_3 procedure with the modified MULTI program [22], as well as by the multi-group collision probability procedure [11]. Some of the results obtained by these methods are displayed in Figs 11 and 12.

3. REACTOR CORE

3.1. Material buckling

U-Pu lattices represent a typical example of the so-called mixed lattices, which contain different types of fuel. In the equilibrium state, by continuous fuel exchange, fuel elements of different uranium and plutonium compositions are uniformly arranged in the reactor lattice. In some cases, e.g. in a reactor with steam superheating, fuel elements of different geometries and compositions are uniformly arranged in the lattice from the very beginning of the operation. To solve the criticality problem, elaborate methods are required, such as the heterogeneous method or the numerical three-dimensional calculation of the reactor core.

When treating the long-term fuel composition changes to determine the maximum fuel burn-up, it is necessary to solve the criticality problem many times, so that application of the above-mentioned methods is laborious and time consuming, even for large-capacity computers. Since fuel elements of different nuclear compositions are uniformly arranged over the whole reactor lattice, it is possible to define the material buckling for a uniformly mixed lattice and to reduce a criticality problem to a problem of finding the roots of the two-group critical equation.

With two types of fuel element, the validity of the definition of the material buckling for the mixed reactor core was experimentally investigated on the Yugoslav heavy-water zero-power reactor RB [23]. Some of the results are presented in Fig. 13. The validity of the adopted simple definition of the material buckling for mixed reactor core, as linearly dependent on the configuration factor, was confirmed.

3.2. Criticality

To be able to establish the best national fuel management policy, special efforts were devoted to the development of fast and reasonably accurate methods for overall reactor core calculation. In addition to the already mentioned burn-up program TER, a criticality program, REDIR, was developed [24]. Galanin's method of effective boundary conditions was used with a two-group calculated effective reflector thickness. The program calculates radial and axial macroparameters and fuel-element burn-up. To calculate U-Pu lattices, suitable averaging procedures have been included. The validity of the assumptions involved in the program was checked by the Feinberg-Galanin heterogeneous method applied in the criticality program HETERO [25], as well as by the experiments described in Section 3.1.

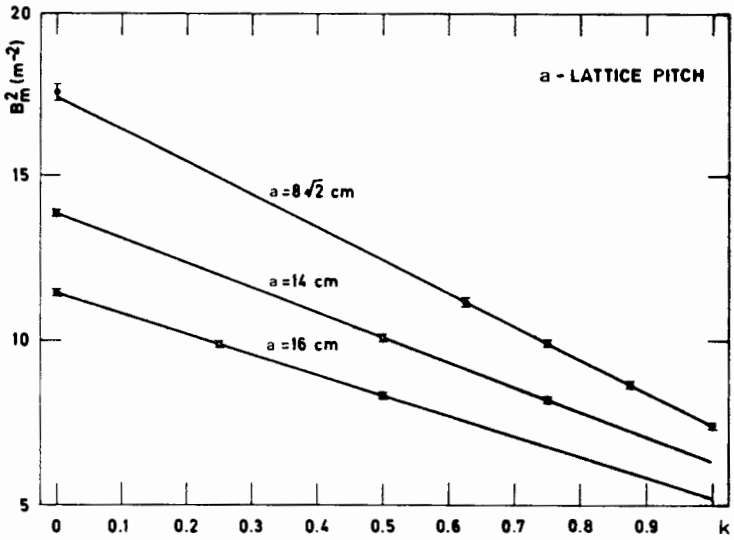

FIG. 13. Material buckling for reactor lattices composed of two types of uniformly arranged fuel elements as function of fraction factor k.

The criticality programs developed were used to study the different refuelling schemes with or without plutonium recycling. Some results of these investigations, including the recommendations for countries with rather small nuclear power programs, are presented in Refs [26] and [27].

3.3. Optimization of core configuration

In designing nuclear reactors and in operating them, a series of problems of variational nature is encountered. The extreme of a physical magnitude is always sought, e.g. minimum critical mass, minimum critical dimensions, maximum reactor power, maximum neutron flux, minimum poisoning after reactor shutdown, minimum time of reactor shutdown, and the like. The function that is varied depends on the nature of the problem investigated. This may be the space distribution of fuel or absorber concentration, the neutron flux, the reactor power, etc. For constructional, thermic, or other reasons, this function is always limited, making these kinds of variational problem non-classical, so that Pontryagin's maximum principle or Bellman's dynamic programming have to be applied.

In the case of U-Pu lattices, the particularly relevant problems are based on the possibility of varying the space distribution of fuel concentration. The shortcoming of all existing solutions is that this distribution is assumed in advance. Here, a direct approach to this problem is defined: to determine the space distribution of fuel concentration in the reactor core directly from the extreme of a desired physical magnitude, satisfying the thermic restrictions.

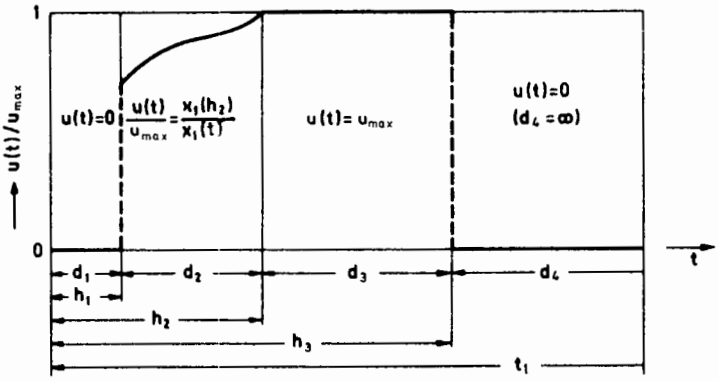

FIG. 14. Optimal core configuration for high-flux reactor.

The mathematical formulation of the above problem is given in Ref. [28] and some applications to the optimization of reactor core configuration are presented in Ref. [29]. For example, the optimal fuel configuration of the reactor core, obtained in the case of a high-flux reactor, consists of a reflector in its centre, a zone of constant (permissible) power density, a zone of constant (maximum) fuel concentration, and a peripheral reflector of infinite thickness (Fig. 14).

The choice of an adequate model of the process in the optimization procedure developed plays an important role. The model presented here is based on the two-group diffusion theory of neutrons, and is given for arbitrary core geometry. However, the same optimization procedure is also applicable with the multi-group diffusion model.

In a similar way, with a corresponding model of the process, numerous problems in reactor kinetics and the control of U-Pu cores can be solved.

REFERENCES

[1] FREDIN, B., BOŠEVSKI, T., MATAUŠEK, M., Discrete representation of thermal neutron spectra in U-Pu lattices, Nucl. Sci. Engng 36 (1969) 315.

[2] BOŠEVSKI, T., FREDIN, B., A Modified Gaussian Integration Method for Thermal Reaction Rate Calculation in U- and Pu-Isotopes, Rep. AE-RFR-556 (1966).

[3] FREDIN, B., A Method of Thermal Spectrum Calculation in Energy Points, Rep. AE-RFR-566 (1966).

[4] MATAUŠEK, M., FREDIN, B., A Method of Thermal Spectrum Calculation in Energy Points Applied to a Two Region Cell and Compared with Standard Multigroup Procedure, Rep. AE-RFR-628 (1967).

[5] MATAUŠEK, M., FREDIN, B., Scattering Matrix and Fictive Neutron Source for Thermal Neutron Spectrum Calculation by the Point Method, Rep. AE-RFR-638 (1967).

[6] MATAUŠEK, M., A Method to solve multigroup P_3 equations in cylindrical geometry, Nukleonik 12 (1968) 45.

[7] MATAUŠEK, M., MULTI – A Multigroup or Multipoint P_3 Program for Calculating Thermal Neutron Spectra in a Reactor Cell, Rep. IBK - 654 (1968).

[8] MATAUŠEK, M., Calculation of Multipoint Data for Thermal Neutron Spectra Determination, Program CAMP, Rep. IBK - 714 (1968).

[9] BOŠEVSKI, T., POP-JORDANOV, J., Collision Probabilities for Concentric Cylindrical Zones, Rep. IBK - 598 (1967).

[10] BOŠEVSKI, T., An improved collision probability method for thermal neutron flux calculation in a cylindrical reactor cell, Nucl. Sci. Engng 42 (1970) 23.
[11] BOŠEVSKI, T., "A generalization of the improved collision probability method to the multigroup neutron flux calculation in cylindrical reactor cell", to be published.
[12] BOŠEVSKI, T., SIMOVIĆ, R., An Improved Collision Probability Method for Determination of the Neutron Flux Space Distribution in a Cell with the Cluster Type Fuel Element, Rep. IBK - 970 (1970) (in Serbocroatian).
[13] POP-JORDANOV, J., 3rd UN Int. Conf. peaceful Uses atom. Energy (Proc. Conf. Geneva, 1964) 2, UN, New York (1965) 126.
[14] POP-JORDANOV, J., Neutron thermalization in reactor cell, Kernenergie 9 (1969) 288.
[15] POP-JORDANOV, J., Termalizaciya neitronov v geterogennom reaktore (Neutron Thermalization in the Heterogeneous Reactor), ITEF 133, Moscow (1963).
[16] POP-JORDANOV, J., BOŠEVSKI, T. et al., Methods and Programs for Physical Calculation of Heavy Water Power Reactors, TER 1, Rep. IBK - 241 (1965); TER 2, Rep. IBK - 504 (1966); TER 3, Rep. IBK - 590 (1967) (in Serbocroatian).
[17] AMOUYAL, A., BENOIST, P., HOROWITZ, J., Nouvelle méthode de détermination du facteur d'utilisation thermique d'une cellule, J. nucl. Energ. 6 (1957) 79.
[18] STAMM'LER, R., TAKAČ, S., WEISS, Z., Neutron Thermalization in Reactor Lattice Cells: An NPY Project Report (Technical Reports Series No. 68) IAEA, Vienna (1966).
[19] MATAUŠEK, M., "Slowing down of resonance neutrons in a reactor lattice cell", Proc. XVth ETAN Conf., Split (1971) (in Serbocroatian).
[20] STEFANOVIĆ, D., An exact solution of the neutron slowing down equation, Nucl. Sci. Engng 41 (1970) 393.
[21] STEFANOVIĆ, D., "An analytical solution of the energy and space distribution of neutron flux in a reactor cell", Proc. XVth ETAN Conf., Split (1971) (in Serbocroatian).
[22] MATAUŠEK, M., "Multigroup calculation of the space energy fast neutron flux distribution in a reactor lattice cell", Proc. XIVth ETAN Conf., Sarajevo (1970) (in Serbocroatian).
[23] BOŠEVSKI, T., REMŠAK, S., Material Buckling Measurements for Mixed Reactor Lattices, Rep. IBK - 657 (1968).
[24] BOŠEVSKI, T., STRUGAR, P., POP-JORDANOV, J., "Continual exchange of fuel in a reactor with a flat central zone", Burn-Up Predictions in Thermal Reactors (Proc. Panel Vienna, 1967), IAEA, Vienna (1968) 191.
[25] JOVANOVIĆ, S., RAIŠIĆ, N., Program HETERO, Rep. IBK - 490 (1966).
[26] POP-JORDANOV, J. et al., "Compensation of low burn-up limits of metallic uranium in heavy water reactors by adjusting the refuelling scheme", Heavy Water Power Reactors (Proc. Panel Vienna, 1967), IAEA, Vienna (1968) 697.
[27] STRUGAR, P. et al., "Steam superheating in BHWRs using plutonium enrichment", Heavy Water Power Reactors (Proc. Panel Vienna, 1967), IAEA, Vienna (1968) 299.
[28] STRUGAR, P., Maximum neutron flux in thermal research reactors, J. Optimization Theory and Applications 5 (1970) 4.
[29] STRUGAR, P., On the optimisation of reactor core configuration, Deutsches Atomforum E.V., Reaktortagung, Berlin (1970).

ETUDE NEUTRONIQUE DU PLUTONIUM ET DE L'EVOLUTION DES COMBUSTIBLES DANS LES REACTEURS A NEUTRONS THERMIQUES *

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Abstract–Résumé–Аннотация–Resumen

NEUTRON STUDY ON PLUTONIUM AND ON FUEL BURN-UP IN THERMAL REACTORS.

Since 1958, the Commissariat à l'énergie atomique has carried out an extensive series of studies in order to obtain a better knowledge of the properties of plutonium-containing fuels, in particular those of fuels irradiated in thermal power reactors. These studies have included integral measurements in heavy-water and graphite-moderated lattices on uranium-plutonium alloy fuels (progressive replacement and oscillations), chemical and isotopic analyses, and integral measurements (oscillations) on samples of fuels irradiated in power reactors. Theoretical models have been developed in order to analyse the experiments as a whole and take proper account of spectrum effects, cross-section variations as a function of temperature (experiments up to 400°C), and changes in the isotopic composition of plutonium during burn-up. A consistent set of cross-sections has been defined. In comparison with the U.K.A.E.A. 1967 library values normalized to the last recommended values at 2200 m/sec (Westcott, 1970), plutonium-240 absorption and the ratio of η of plutonium-239 to η of uranium-235 have been decreased, and the resonance amplitude of the absorption and fission cross-sections of plutonium-239 have been modified. The main parameters of the changes in the fuel during irradiation, such as the resonance integral of uranium-238 and the capture cross-sections of plutonium isotopes, are adjusted on the basis of the results of irradiated fuel analysis. With a knowledge of the plutonium data, the total capture of fission products can be inferred from the oscillations of the irradiated fuels. These methods and results are applied to the study of fuel burn-up in French power reactors and to evaluation of the plutonium produced and its isotopic composition. A direct application can also be made in the case of plutonium recycle studies.

ETUDE NEUTRONIQUE DU PLUTONIUM ET DE L'EVOLUTION DES COMBUSTIBLES DANS LES REACTEURS A NEUTRONS THERMIQUES.

Depuis 1958, le Commissariat à l'énergie atomique a réalisé un ensemble important d'études visant à connaître les propriétés des combustibles contenant du plutonium et notamment celles des combustibles irradiés dans les réacteurs de puissance à neutrons thermiques. Ces études ont consisté d'une part en des mesures intégrales dans des réseaux modérés à l'eau lourde et au graphite sur des combustibles d'alliages uranium-plutonium (remplacement progressif et oscillations), d'autre part en des analyses chimiques et isotopiques et des mesures intégrales (oscillations) sur des échantillons de combustibles irradiés dans les réacteurs de puissance. Des modèles théoriques ont été élaborés pour interpréter l'ensemble de ces expériences, tenir compte correctement des effets de spectre, de la variation des sections efficaces effectives avec la température (expériences jusqu'à 400°C) et de l'évolution de la composition isotopique du plutonium au cours de l'évolution. Un ensemble cohérent de sections efficaces a été mis au point: par rapport aux valeurs de la bibliothèque UKAEA 1967 normalisées aux dernières valeurs recommandées à 2200 m/s (Westcott, 1970), on est amené à diminuer l'absorption du plutonium-240 et la valeur du rapport des facteurs η du plutonium-239 et de l'uranium-235, et à modifier l'amplitude des résonances des sections d'absorption et de fission du plutonium-239. Les analyses de combustibles irradiés permettent

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* Cette étude a été réalisée avec la participation de plusieurs équipes et de nombreuses personnes, en particulier MM. M. Livolant et A. Hoffmann pour les méthodes de calcul, M.Y. Bouedo pour l'interprétation des expériences, M.R. Bir et ses collaborateurs pour les analyses de combustibles, et les équipes des réacteurs AQUILON, MINERVE et CESAR.

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l'ajustement des principaux paramètres de l'évolution du combustible, intégrale de résonance de l'uranium-238 et sections de capture des isotopes du plutonium. Les oscillations de combustibles irradiés donnent, à partir des connaissances relatives au plutonium, la capture totale des produits de fission. Ces méthodes et ces résultats sont appliqués à l'étude de l'évolution du combustible des réacteurs de puissance français et à l'évaluation du plutonium produit et de sa composition isotopique; cet ensemble trouve également une application directe dans les études du recyclage du plutonium.

НЕЙТРОННОЕ ИССЛЕДОВАНИЕ ПЛУТОНИЯ И ВЫГОРАНИЯ ТОПЛИВА В РЕАКТОРАХ НА ТЕПЛОВЫХ НЕЙТРОНАХ.

С 1958 года Комиссариат по атомной энергии провел крупные серии исследований для лучшего познания свойств топлива, содержащего плутоний и, в частности, свойства топлива, облученного в тепловых энергетических реакторах. Эти исследования проводились по интегральным измерениям в тяжеловодных графитовых решетках на урано-плутониевых сплавах с помощью методов регрессивного замещения и осцилляции, и по химическим и изотопным анализам, а также интегральным измерениям (осцилляции) на образцах облученного в энергетических реакторах топлива. Были разработаны теоретические модели для анализа серии экспериментов, правильного учета эффектов спектра, вариации эффективных сечений как функции температуры (эксперименты до 400°C) и изменения в изотопном составе плутония во время выгорания. Определен совместимый ряд сечений. По сравнению со значениями 1967 года библиотеки Управления по атомной энергии Соединенного Королевства, приведенными к последним рекомендованным значениям 2200 м/сек (Westcott. 1970), поглощение ^{240}Pu и отношение η ^{239}Pu к η ^{235}U были уменьшены, изменяются также амплитуда резонанса поглощения ^{239}Pu и сечения деления. Основные параметры изменения в топливе во время облучения, такие, как резонансный интеграл ^{238}U и сечения захвата изотопов плутония регулируются по результатам анализов облученного топлива. Осцилляции облученного топлива дают общее поглощение продуктов деления на основе знания значений плутония. Полученные таким образом методы и результаты применяются к исследованию выгорания топлива во французских энергетических реакторах и к оценке производства плутония и его изотопного состава. Они также могут непосредственно применяться в исследованиях повторного плутониевого цикла.

ESTUDIO NEUTRONICO DEL PLUTONIO Y DE LA EVOLUCION DEL COMBUSTIBLE EN LOS REACTORES DE NEUTRONES TERMICOS.

Desde 1958, la Comisaría de Energía Atómica de Francia ha realizado una serie importante de estudios destinados a conocer las propiedades de los combustibles que contienen plutonio y principalmente las de los combustibles irradiados en los reactores de potencia de neutrones térmicos. Estos estudios han consistido, por una parte, en medidas integrales sobre retículos de combustibles de aleaciones uranio-plutonio moderados por agua pesada y por grafito (sustitución progresiva y oscilaciones), y, por otra parte, en análisis químicos e isotópicos y en medidas integrales (oscilaciones) sobre probetas de combustibles irradiados, en los reactores de potencia. Se han elaborado modelos teóricos para interpretar el conjunto de estos experimentos, teniendo en cuenta correctamente los efectos de espectro, la variación de las secciones eficaces efectivas con la temperatura (experimentos hasta 400°C) y la evolución de la composición isotópica del plutonio durante la irradiación. Se ha puesto a punto una serie coherente de secciones eficaces; con relación a los valores de la biblioteca de la UKAEA 1967 normalizados a los últimos valores recomendados para 2200 m/s (Westcott 1970), se ha disminuido la absorción del plutonio y del uranio 235, y se ha modificado la amplitud de las resonancias de las secciones de absorción y de fisión del plutonio 239. Los análisis de combustibles irradiados permiten el ajuste de los principales parámetros de la evolución del combustible, integral de resonancia del uranio 238 y secciones de captura de los isótopos del plutonio. Las oscilaciones del combustible irradiado dan, a partir de los conocimientos relativos al plutonio, la captura total de los productos de fisión. Estos métodos y resultados se aplican al estudio de la evolución del combustible de los reactores de potencia franceses y a la evaluación del plutonio producido y de su composición isotópica; también encuentran una aplicación directa en los estudios del reciclado del plutonio.

INTRODUCTION

Un programme important a été réalisé au Commissariat à l'énergie atomique, depuis une dizaine d'années, sur l'étude des propriétés neutroniques du plutonium et l'évolution du combustible dans les réacteurs à neutrons thermiques à uranium naturel. Ces travaux ont été réalisés

en partie dans le cadre de deux contrats EURATOM sur le « Recyclage du plutonium » [1, 2] et en coopération avec Electricité de France.

Le programme expérimental a porté à la fois sur des alliages uranium-plutonium et sur des combustibles irradiés dans des réacteurs de puissance [3]. Dans le premier cas, on a fait des expériences critiques et des mesures par oscillations; on a fait varier largement les spectres en agissant, soit sur le rapport de modération (ainsi que sur la nature du modérateur: graphite ou eau lourde), soit sur la température. Dans le second cas, on a fait des analyses chimiques et isotopiques des teneurs en noyaux lourds à divers stades d'évolution, ainsi que des mesures de réactivité par oscillation.

Ce programme a comporté également des études théoriques, qui ont porté principalement sur la mise au point de méthodes de calcul précises des spectres de neutrons dans le domaine de la thermalisation. On a pu rendre ainsi toutes les mesures cohérentes et remonter aux données nucléaires de base. Dans ce qui suit, avant de décrire les programmes expériementaux et leurs résultats, on donne quelques indications, d'une part sur les études théoriques, d'autre part sur les méthodes d'analyse chimique et isotopique dont la précision est essentielle.

1. METHODES DE CALCUL [4]

Au début des années 60, les études théoriques ont porté sur l'élaboration de méthodes de calcul des spectres qui soient à la fois précises et rapides; elles ont abouti notamment à la mise au point d'un modèle synthétique de l'opérateur de thermalisation (modèle de Cadilhac, ou modèle « secondaire » [5]).

Dans la période récente, l'accent a été mis sur l'application des théories précédemment élaborées; on notera trois directions principales:
- constitution d'une bibliothèque de sections efficaces et surtout de sections de transfert en énergie des modérateurs,
- développement de méthodes de calcul de cellules et réalisation des codes correspondants,
- traitement des produits de fission.

1.1. Bibliothèques

Les données physiques nécessaires au calcul et à l'utilisation des spectres thermiques sont les sections efficaces, dans le domaine thermique, des éléments entrant dans la composition des combustibles et les sections de transfert en énergie et de diffusion des principaux modérateurs.

De plus, pour l'utilisation du modèle de Cadilhac, il est nécessaire de déterminer les fonctions G et H caractéristiques de ce modèle et, pour une prise en compte correcte des fuites macroscopiques, le coefficient de diffusion en fonction de l'énergie.

La plus grande partie des sections efficaces d'absorption et de fission utilisées provient de la bibliothèque UKAEA. Les autres sections

ont été extraites de la bibliothèque ENDF/B. En particulier, pour les isotopes les plus utilisés, les références sont les suivantes:

Bibliothèque UAKEA :	^{235}U	^{239}Pu	^{240}Pu	^{241}Pu	^{135}Xe
DFN	30	329	201	40	4
Bibliothèque ENDF/B:	^{149}Sm	^{241}Am	^{243}Am	^{237}Np	^{242}Pu
MAT	1027	1056	1057	1048	1055

On a établi un formalisme qui permet d'effectuer le calcul numérique des sections de transfert à partir d'une donnée physique fondamentale: le spectre des fréquences propres de vibration du noyau, compte tenu de toutes les liaisons auxquelles il est soumis. Les modèles physiques utilisés pour l'obtention des spectres de fréquences pour les différents modérateurs sont indiqués en [4].

L'ajustement des fonctions G et H est réalisé sur les flux: ces fonctions sont données en [4] pour les différents modérateurs. Le coefficient de diffusion est calculé à partir du spectre de fréquences du modérateur.

1.2. Méthodes de calcul de cellule

Pour les calculs de cellule, on a utilisé, soit le code THERMOS, basé sur la résolution directe des équations de transport-thermalisation exactes, soit des codes fondés sur l'emploi simultané d'un traitement simplifié de l'espace (méthode des zones-source) et du modèle de Cadilhac. Un certain nombre d'améliorations ont été apportées au code THERMOS initial: augmentation des possibilités du programme (50 points d'espace, 60 groupes d'énergie, 7 milieux), calcul plus correct des probabilités de collision, calcul de la source de ralentissement en utilisant les sections de transfert exactes des isotopes, calcul de sections multigroupes paraboliques, procédé d'itération plus sûr et plus rapide.

Les codes dits « secondaires » associent au modèle de thermalisation un traitement de l'espace simplifié, en exploitant les caractères spécifiques des problèmes traités: pour une cellule usuelle à eau lourde ou au graphite, le procédé de calcul spatial est une extension de la méthode ABH; pour les cellules modérées à l'eau ordinaire, on calcule les probabilités de première collision dans une géométrie cylindrisée, ou dans une géométrie réelle sans découpage du modérateur. Le code DEP 035 [6] est le plus couramment utilisé, en particulier pour l'interprétation des expériences sur les combustibles au plutonium.

Le principal avantage des codes secondaires est, pour des cellules de grandes dimensions réclamant de nombreux points d'espace, la rapidité de calcul (facteur 10 à 100 avec THERMOS). Actuellement, la comparaison des différentes méthodes de calcul montre que l'incertitude liée aux données nucléaires fondamentales est nettement supérieure à l'incertitude liée aux différences entre méthodes de calcul.

1.3. Traitement des produits de fission et calculs d'évolution

Les codes d'évolution [6] sont, pour le calcul des spectres, des codes secondaires. Ils traitent 32 produits de fission, la capture totale des produits de fission étant complétée au moyen d'un pseudo-produit. On a également mis au point une méthode plus élaborée, qui permet de calculer l'évolution d'un nombre quelconque de produits de fission (actuellement 188). Les sections efficaces effectives d'absorption sont calculées dans le formalisme de Westcott, en tenant compte des effets d'ombre et d'auto-protection des résonances. Les données nucléaires sont issues des ouvrages de Walker [7] et d'England [8].

2. ANALYSE DES ALLIAGES URANIUM-PLUTONIUM ET DES COMBUSTIBLES IRRADIES: METHODES ET RESULTATS [2]

La plus grande difficulté rencontrée dans ce type d'analyse est d'obtenir des solutions à doser représentatives des échantillons à étudier. Le choix des prélèvements sur les barreaux U-Pu ou sur les éléments combustibles irradiés, puis leur mise en solution, constituent des opérations très délicates, à cause des risques de pollutions aux différents stades des manipulations et de l'hétérogénéité des teneurs en plutonium. Les analyses physico-chimiques étant destructives, il est indispensable de s'assurer, par des méthodes physiques, que le prélèvement analysé est bien représentatif de la moyenne du barreau à étudier ou de mesurer sa différence avec une précision suffisante.

Pour les combustibles irradiés, l'examen de la répartition des taux de fission est effectué par spectrométrie γ.

Pour les alliages U-Pu, les comparaisons sont effectuées, pour les prélèvements, par spectrométrie α et, pour les barreaux, par comptage des neutrons émis par la fission spontanée du ^{240}Pu.

Les analyses sont toutes effectuées par spectrométrie de masse et elles concernent essentiellement la mesure de la teneur isotopique du plutonium et la détermination du rapport Pu/U par la méthode de la double dilution isotopique [1].

Pour les combustibles irradiés, on mesure en plus l'appauvrissement en ^{235}U et la teneur en ^{236}U, à l'aide d'un spectrometre de masse à UF_6, dont l'avantage réside essentiellement dans l'excellente précision que l'on peut atteindre, en particulier pour les faibles irradiations. Le principe de l'appareil est de déterminer directement la différence des teneurs entre deux échantillons voisins, l'un étant le combustible irradié appauvri en ^{235}U, l'autre le combustible initial ou un étalon d'enrichissement voisin de celui à mesurer. On exprime la différence, par rapport à leurs teneurs d'origine, des teneurs N_5 en ^{235}U et N_8 en ^{238}U par l'expression:

$$1 - \alpha = 1 - \frac{N_5/N_8}{(N_5/N_8)_0}$$

et la précision relative atteinte sur le terme $(1 - \alpha)$ est de 0,7%.

Les analyses isotopiques du plutonium sont effectuées avec un spectromètre de masse équipé d'une source à ionisation de surface, dont le signal de sortie est, après digitalisation, transféré sur bande perforée, puis traité sur ordinateur.

Le traitement des données, très élaboré, permet d'atteindre les précisions suivantes (N_i et N_j étant les concentrations respectives des deux isotopes du plutonium):

Pour $R = N_i / N_j > 10\%$	$\Delta R = \pm 0,1\%$
Pour $1\% < N_i / N_j < 10\%$	$\Delta R = \pm 1\% \times R$
Pour $N_i / N_j < 1\%$	$\Delta R = \pm 0,01\%$

La détermination de la teneur en plutonium, c'est-à-dire du rapport $^{239}Pu/^{238}U$, est effectuée par la technique de la dilution isotopique avec traceur double. En mélangeant ce traceur, constitué de ^{233}U et de ^{242}Pu, avec la solution à analyser, la dilution isotopique a lieu simultanément pour l'uranium et le plutonium. Les compositions isotopiques de ces deux éléments sont modifiées par le mélange, mais de telle façon que la relation existant entre le rapport des masses des éléments dans l'échantillon et la composition isotopique de ces éléments dans le mélange sera indépendante des masses mélangées, ce qui permet d'éviter la mesure précise des quantités de traceur et d'échantillon que l'on utilise. Toutefois, afin d'opérer dans les meilleures conditions, on optimise le mélange solution-traceur pour que le rapport des pics à mesurer soit le plus voisin possible de l'unité.

Une étude approfondie, portant sur plus de 200 déterminations, montre que la précision relative sur le rapport U/Pu est très voisine de 1%, qu'elle est remarquablement indépendante du temps et de la teneur en en plutonium, tout au moins dans la gamme couverte par cette étude.

3. ETUDE EXPERIMENTALE DE COMBUSTIBLES URANIUM-PLUTONIUM

On a fabriqué des combustibles d'alliages métalliques d'uranium et de plutonium, dont les compositions couvrent globalement les variations en cours d'évolution des teneurs en noyaux lourds des combustibles des réacteurs de puissance à uranium naturel ou faiblement enrichi. Les expériences effectuées constituent un ensemble cohérent de mesures portant sur un type de combustible, dans deux modérateurs (eau lourde et graphite), à plusieurs pas de réseau et plusieurs températures. Deux méthodes expérimentales ont été utilisées: le remplacement progressif et les oscillations.

Les expériences d'oscillation à différentes températures, qui portent sur un grand nombre de combustibles de différentes teneurs, permettent d'obtenir un jeu cohérent de sections efficaces du plutonium et de l'uranium-235. Ce jeu de sections « évalué » est appliqué ensuite aux autres types d'expériences.

TABLEAU I. ECHANTILLONS D'OSCILLATION

$\frac{^{235}U}{U}$ (‰)	$\frac{Pu}{U}$ (‰) / $\frac{^{240}Pu}{Pu}$ (‰)	0,15	0,20	0,25	0,30	0,35
0,45	3			×	×	×
	8	×	×		×	×
	20			×	×	
	35					
0,22	3			×		
	20			×		
0,35	35			×	×	×

Nota. Les croix correspondent aux échantillons. Les échantillons à 20‰ et 35‰ de ^{240}Pu contiennent respectivement 2‰ et 6‰ de ^{241}Pu. Les précisions sur les teneurs sont données au paragraphe 2 (la précision sur la teneur en bore des échantillons d'étalonnage est de ±1‰).

3.1. Expériences d'oscillations dans CESAR en température

La méthode d'oscillation [2] consiste à remplacer dans un réacteur expérimental critique l'élément combustible standard, placé au centre du réacteur (ou élément de référence), par l'échantillon U-Pu à mesurer; ce remplacement est effectué en imprimant un mouvement périodique à l'ensemble du canal de combustible ainsi constitué. On mesure deux quantités: le signal « Global », image de la réactivité de l'échantillon, et le signal « Local », image de la variation de la densité neutronique au voisinage de l'échantillon.

L'interprétation de ces expériences est faite par la méthode de l'échantillon équivalent [9]. En utilisant le double étalonnage du réacteur par des échantillons d'uranium enrichi ou appauvri et d'uranium naturel boré, on obtient de façon indépendante les taux de réaction d'absorption et de production du plutonium dans chaque échantillon, donc les sections effectives $\hat{\Sigma}_{aP}$ et $\nu\hat{\Sigma}_{fP}$, en fonction de celles de l'uranium-235.

Les expériences effectuées dans CESAR portent sur 14 échantillons (9 à 400 °C). Les échantillons sont des cylindres pleins de 29,2 mm de diamètre et 292,5 mm de longueur. Les teneurs en uranium-235 et en plutonium sont données au tableau I.

La zone centrale de CESAR est constituée par un réseau de cellules hexagonales (pas 225,16 mm, canal 70 mm); l'ensemble du réacteur est chauffé par circulation de CO_2; les expériences ont été effectuées à 20°C, 200°C, 400°C (précision sur les températures: ± 1°C). La comparaison entre calcul et expérience porte sur les quantités $\hat{\Sigma}_{aP}$ et $\nu\hat{\Sigma}_{fP}$. Les calculs des spectres et des sections effectives ont été effectués par les codes DEP 035 et THERMOS. Les données nucléaires

TABLEAU II. SECTIONS EFFICACES DE REFERENCE A 2200 m/s

	^{235}U	^{239}Pu	^{241}Pu	^{240}Pu	^{242}Pu	^{241}Am
ν	2,4229	2,3799	2,934			
σ_f (b)	580,2	741,3	1007,3			
σ_a (b)	678,5	1012,9	1375,5	297	30	587

fondamentales sont celles du paragraphe 1 en ce qui concerne les courbes des sections efficaces, normalisées aux valeurs à 2200 m/s qui figurent au tableau II.

Pour les noyaux fissiles, ce sont les dernières valeurs publiées par l'AIEA [10].

Les expériences en température offrent l'intérêt de pouvoir séparer les effets dus à la forme des sections de ce qui est dû à la valeur à 2200 m/s considérée comme une valeur de normalisation: dans le cas des isotopes du plutonium, cela revient à comparer l'importance de la première résonance à celle de la partie en 1/v.

Résultat: jeu « évalué » de données nucléaires de base

Ces mesures permettent ainsi d'obtenir un jeu évalué cohérent de sections efficaces caractérisé par rapport aux données initiales par les modifications suivantes:

a) Augmentation de 7% ± 2% et 5% ± 2% des hauteurs des pics des résonances d'absorption et de fission de ^{239}Pu.

b) Sections à 2200 m/s:

^{240}Pu: $\sigma^0_{a0} = 280 \pm 10$ b

^{241}Pu: $\sigma^0_{f1} = 1030 \pm 20$ b

^{239}Pu: $\sigma^0_{f9} = 1015 \pm 5$ b

^{239}Pu et ^{235}U: $\sigma^0_{f9} = 741,6$ b et $\sigma^0_{f5} = 584 \pm 3$ b (version 1)

ou $\sigma^0_{f9} = 737$ b ± 4 b et $\sigma^0_{f5} = 580,2$ b (version 2)

Dans les deux cas $\eta^0_9/\eta^0_5 = 1,009 \pm 0,007$

Les deux versions du jeu évalué sont équivalentes pour l'interprétation de nos mesures puisque ce sont des mesures relatives où on compare le plutonium à l'uranium-235.

Le tableau III illustre la comparaison entre le jeu de référence et le jeu évalué proposé.

3.2. Expériences de remplacement progressif [11]

Les éléments combustibles d'un réseau de référence sont remplacé de façon progressive par les éléments U-Pu à étudier. A chaque étape de cette substitution, on mesure la variation de réactivité de la pile,

ou de sa taille critique, et on en déduit, par extrapolation, le laplacien matière du réseau substitué comparé à celui du réseau de référence. Cette méthode n'utilise qu'une fraction du volume critique; cependant, dans la mesure où on cherche à s'affranchir des effets d'interaction entre zones, elle nécessite une quantité importante de combustible (de l'ordre de 1 tonne). On s'est donc limité à l'étude de trois jeux de teneurs différentes:

Jeu 1: uranium naturel + 0,05% de plutonium à 6% de ^{240}Pu.

Jeu 2: uranium appauvri à 0,22% de ^{235}U + 0,30% de plutonium à 8% de ^{240}Pu.

Jeu 3: uranium naturel + 0,05% de plutonium à 25% de ^{240}Pu.

On a étalonné les variations de réactivité par des jeux d'uranium enrichi et appauvri qui ont été mesurés de la même façon.

Les éléments combustibles ont la même géométrie que ceux des expériences d'oscillations. Ces expériences ont été réalisées dans CESAR (modérateur graphite, mesures de 20°C à 450°C) et dans AQUILON (modérateur eau lourde, mesures à différents pas). L'interprétation des résultats se traduit par la comparaison entre calcul et expérience des valeurs des laplaciens, ou du η du combustible, lorsqu'on utilise le jeu de données nucléaires de référence ou le jeu évalué proposé au paragraphe précédent.

3.2.1. Expériences dans CESAR

Les mesures ont été effectuées à 20, 100, 200, 300, 400 et 450°C. Les caractéristiques du réseau de mesure sont celles décrites pour les expériences d'oscillations. Les écarts entre calcul et expérience sur les laplaciens sont traduits en écarts sur le rapport de η (η') du combustible substitué U-Pu au η du combustible de référence uranium naturel. Le tableau IV donne les valeurs de cet écart selon le jeu de données nucléaires que l'on utilise.

Discussion des résultats

– Les modifications proposées des données fondamentales apportent peu d'amélioration pour les jeux 1 et 3. Ces combustibles contiennent d'ailleurs très peu de plutonium et les écarts obtenus sont en général à l'intérieur des marges d'incertitude liées à l'expérience, auxquelles il faut ajouter celles sur les teneurs en plutonium: on ne peut en tirer que peu d'enseignements.

– Par contre, les résultats du jeu 2 sont très sensibles à ces modifications (ceci est lié à la forte teneur en plutonium de ce combustible). Les variations constatées sont significatives: l'écart entre calcul et expérience est très nettement diminué lorsqu'on utilise le jeu évalué. Notons d'ailleurs que l'on doit ajouter à la marge d'incertitude expérimentale une marge égale due à l'incertitude sur les teneurs en plutonium.

En conclusion, les expériences de remplacement progressif dans CESAR, en température, confirment les modifications proposées des données nucléaires fondamentales.

TABLEAU III. ECARTS RELATIFS ENTRE JEU DE REFERENCE ET JEU EVALUE
(C-E)/E en 10^{-2} pour $\hat{\Sigma}_{aP}$ et $\nu\hat{\Sigma}_{fP}$
(C: valeur calculée; E: valeur mesurée; JR: jeu de référence; JE: jeu évalué)

Echantillons [a]	$\hat{\Sigma}_{aP}$						$\nu\hat{\Sigma}_{fP}$					
	20°C		200°C		400°C		20°C		200°C		400°C	
	JR	JE	JR	JE	JR	JE	JR	JE	JR	JE	JR	JE
45-15-8	-1,4	0,1	-1,4	0,8	-2,3	0,3	0,0	0,4	-0,1	0,8	-1,3	0,1
45-20-8	-1,5	0,0	-2,5	-0,3	-2,7	-0,2	-0,1	0,2	-0,9	0,0	-1,5	-0,1
45-25-3	-1,6	0,1	-3,2	-0,8	b	b	-0,2	0,2	-1,4	-0,4	b	b
45-25-20	-1,2	-0,1	-3,1	-0,3	b	b	-0,7	-0,3	-1,1	-0,1	b	b
22-25-3	-1,8	-0,1	-2,6	-0,2	-3,0	-0,2	-0,3	0,0	-1,0	0,0	-1,7	0,2
22-30-3	-1,4	0,3	-2,6	-0,1	-3,2	-0,4	-0,1	0,2	-1,0	0,0	-1,6	-0,1
22-35-3	-1,6	0,0	-2,3	0,1	-3,3	-0,6	-0,3	0,1	-0,8	0,2	-1,5	-0,1
22-30-8	-2,2	-0,7	-2,3	-0,1	-3,7	-1,2	-0,8	-0,4	-1,0	0,0	-2,0	-0,6
22-35-8	-0,9	0,6	-2,9	-0,7	-3,3	-0,8	0,3	0,7	-1,1	-0,1	-1,5	0,0
22-25-20	-0,9	0,2	-1,0	0,7	-1,1	0,9	-0,6	-0,1	-0,6	0,4	-0,7	0,7
22-30-20	-0,9	0,1	-1,3	0,4	-1,6	0,5	-0,8	-0,2	-0,8	0,1	-0,9	0,5
35-25-35	-0,6	0,2	-1,3	0,0	b	b	-0,4	0,1	-0,8	0,2	b	b
35-30-35	-1,1	-0,4	-2,4	-1,1	b	b	-1,0	-0,5	-2,0	-1,0	b	b
35-35-35	-0,5	0,3	-2,3	-1,1	b	b	-0,4	0,2	-1,9	-0,9	b	b
Moyenne [c]		0,0		-0,2		-0,2		0,1		0		0

[a] La nomenclature des échantillons correspond au tableau des teneurs: le premier nombre indique l'appauvrissement en 10^{-2}, le second la teneur en plutonium en 10^{-2}, le troisième la teneur isotopique en ^{240}Pu.

[b] Echantillons non mesurés à 400°C.

[c] La valeur moyenne n'a de sens que pour le jeu évalué correspondant aux sections ajustées.

TABLEAU IV. ECARTS ENTRE η'/η CALCULE ET EXPERIMENTAL $(\eta'/\eta)_{calc} - (\eta'/\eta)_{exp}$ $(\times 10^{-5})$.

Combustible U-Pu	Température (°C)	Jeu de référence (pcm)	Jeu évalué (pcm)
Jeu 1	20	- 56	- 78
	100	- 71	- 91
	200	+ 29	- 3
	300	- 92	- 112
	400	a	a
	450	- 53	- 70
Jeu 2	20	+ 330	+ 63
	100	+ 416	+ 155
	200	+ 490	+ 250
	300	+ 365	+ 110
	400	+ 206	- 8
	450	+ 127	- 121
Jeu 3	20	- 1	- 6
	100	+ 21	+ 52
	200	- 12	+ 3
	300	+ 36	+ 71
	400	a	a
	450	+ 40	+ 72

[a] Mesures non effectuées.

Nota. La marge d'incertitude expérimentale à associer à ces résultats est de $\pm 100 \times 10^{-5}$.

3.2.2. Expériences dans AQUILON [2]

Des expériences de remplacement progressif ont été faites dans l'installation critique à eau lourde AQUILON II sur des réseaux géométriquement identiques mais comportant des différences de composition des combustibles: le réseau de référence est en uranium naturel, les réseaux substitués en uranium enrichi ou appauvri (0,69%, 0,83%, 0,86% de ^{235}U) ou en alliage uranium-plutonium (jeux 1, 2 et 3). On a utilisé cinq pas du réseau (12 à 21 cm) pour mettre en œuvre une assez large variation de spectre.

Les différences de laplaciens entre les réseaux testés et les réseaux de référence sont déduites de la variation de la hauteur critique de la pile au cours du remplacement progressif. On a utilisé pour l'interprétation une méthode hétérogène qui permet de mieux prendre en compte les effets géométriques durant la substitution progressive et surtout les différences de composition isotopique entre les éléments testés.

Les fuites ayant été étalonnées au cours des expériences portant sur des combustibles à teneur variable en ^{235}U, on peut convertir les écarts de laplaciens entre les réseaux au plutonium et les réseaux à l'uranium naturel en écarts de réactivité qui portent principalement sur le terme η. On peut donc comparer ces valeurs expérimentales de η'/η (η': réseau au plutonium; η: réseau à l'uranium naturel) aux valeurs calculées a priori. Les valeurs « expérimentales » dépendent légèrement du calcul

TABLEAU V. ECARTS ENTRE $[(\eta'/\eta) - 1]$ calculé et expérimental

$[(\eta'/\eta)-1]_{exp} - [(\eta'/\eta) - 1]_{calc}$

Jeu combustible	Pas	Ecarts avec le jeu de référence JR (pcm)	Ecarts avec le jeu évalué JE (pcm)	Ecarts avec le jeu évalué modifié JE' (pcm)
1 P	12	69	112	
	13	69	121	
	17	- 43	- 4	
	19	1	29	
	21	- 33	4	
2 P	12	- 273	- 5	- 60
	13	- 324	- 50	- 86
	17	- 396	- 122	- 109
	19	- 444	- 166	- 141
	21	- 506	- 230	- 194
3 P	12	57	40	- 24
	13	55	57	5
	17	- 63	- 62	- 81
	19	- 62	- 56	- 66
	21	- 44	- 35	- 36

Nota. Pour ces comparaisons l'erreur sur la teneur en plutonium entraîne une incertitude de ± 30 pcm pour les jeux 1 et 3 et de ± 90 pcm pour le jeu 2. L'erreur neutronique varie de 35 à 70 pcm du pas 12 au pas 21 pour les jeux 1 et 3, et de 45 à 90 pcm pour le jeu 2.

(variations de f, L^2, L_s^2 principalement). Dans le tableau V on fait ces comparaisons en utilisant deux jeux de sections efficaces: le jeu de référence (JR) et le jeu évalué proposé (JE).

Discussion des résultats

– Les deux premières conclusions des expériences dans CESAR se retrouvent ici:
- pour les combustibles des jeux 1 et 3, les deux jeux de sections efficaces donnent des résultats compris dans les marges d'erreur;
- par contre, pour le jeu 2, le jeu évalué donne des résultats plus proche de l'expérience que le jeu de référence.

En effet, les écarts moyens (en pcm) $[(\eta'/\eta - 1)_{exp} - (\eta'/\eta - 1)_{calc}]$ sont les suivants:

	Jeu 1	Jeu 2	Jeu 3
JR	13	- 390	- 11
JE	52	- 115	- 11

– De plus, l'échelonnement des écarts (expérience-calcul) est significatif pour les trois jeux: on surestime l'efficacité du plutonium aux grands pas. Cet effet, particulièrement net pour le jeu 2, peut être expliqué en modifiant la forme de la section efficace du ^{240}Pu: une diminution de 10% de la capture effective de cet isotope dans le domaine de la résonance à 1 eV réduit la variation de l'écart du pas 12

au pas 21, pour le jeu 2 comme pour le jeu 3. Ceci est illustré dans le tableau V où la colonne JE' indique que l'on a utilisé le jeu évalué JE avec cette dernière modification relative à l'absorption du plutonium-240.

4. EXPERIENCES D'OSCILLATIONS DE COMBUSTIBLES IRRADIES: DETERMINATION DE LA CAPTURE TOTALE DES PRODUITS DE FISSION

Les expériences d'oscillations de combustibles irradiés sont tout à fait analogues à celles déjà décrites pour les combustibles U-Pu. Après une période de mise au point sur les réacteurs MINERVE et MARIUS, un programme important de mesures a été réalisé dans MARIUS en 1968 sur des combustibles en provenance des réacteurs G3 et CHINON 2. Ces expériences ont été effectuées avec mesure du signal Global seul. La méthode de l'échantillon équivalent et le double étalonnage par des échantillons enrichis ou appauvris en ^{235}U, ou borés, permettent d'aboutir à une relation entre l'absorption et la production de neutrons du combustible. Les teneurs en noyaux lourds des combustibles oscillés sont connues par les analyses.

La production de neutrons est calculée à partir de ces teneurs et du jeu de sections évalué à partir des expériences U-Pu, en tenant compte de la répartition radiale des noyaux lourds dans le combustible. Le résultat de l'expérience est alors l'absorption thermique totale de l'échantillon, et plus précisément l'absorption des noyaux lourds formés en cours d'évolution et des produits de fission. C'est donc sur le terme $(\hat{\Sigma}_{aP} + \hat{\Sigma}_{aPF})$ que porte la comparaison entre calcul et expérience. Le calcul permettant de déterminer le poids de l'absorption des produits de fission $[\hat{\Sigma}_{aPF}/(\hat{\Sigma}_{aP} + \hat{\Sigma}_{aPF})]$, on parvient ainsi à une estimation de $\hat{\Sigma}_{aPF}$ que l'on compare à la valeur calculée par le code d'évolution COREGRAF 2 [6] dans lequel le paramètre qui fixe l'irradiation est l'appauvrissement en ^{235}U mesuré par analyse:

$$\alpha = \frac{N_5/N_8}{(N_5/N_8)_0}$$

Les combustibles mesurés sont:

a) 9 barreaux d'uranium naturel de 31 mm de diamètre irradiés dans G3. Les taux d'irradiation varient de 1100 à 3500 MWj/t ($1 - \alpha$ varie de 0,16 à 0,40).

b) 33 tubes d'uranium naturel (de 18 mm de diamètre intérieur et 40 mm de diamètre extérieur) allié à du molybdène (teneur 1,1%) irradiés dans CHINON 2. Les taux d'irradiation varient de 500 à 2000 MWj/t ($1 - \alpha$ varie de 0,05 à 0,26).

Les résultats de cette étude sont illustrés par les figures 1 à 4.

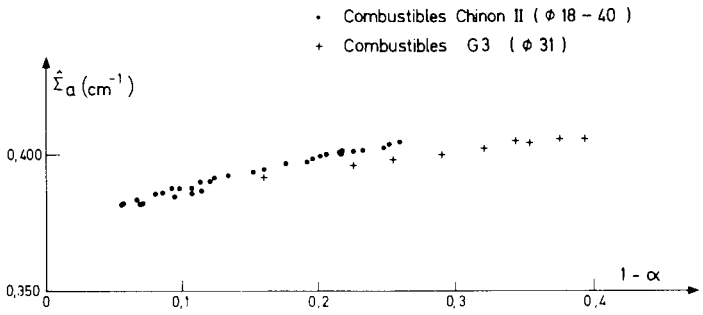

FIG. 1. $\hat{\Sigma}_a$ en fonction de l'appauvrissement en ^{235}U.

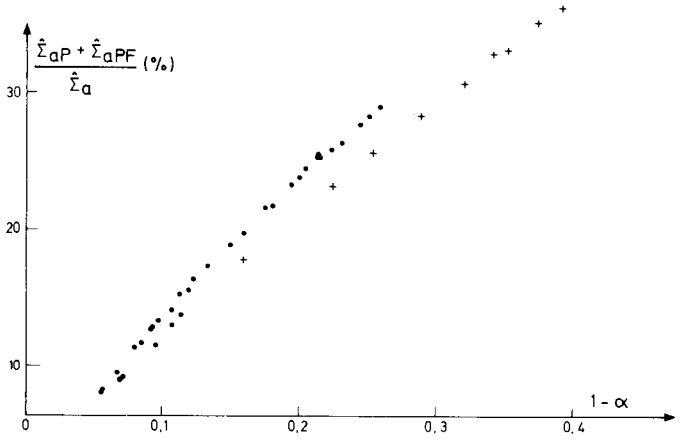

FIG. 2. $(\hat{\Sigma}_{aP} + \hat{\Sigma}_{aFF})/\hat{\Sigma}_a$ en fonction de l'appauvrissement en ^{235}U.

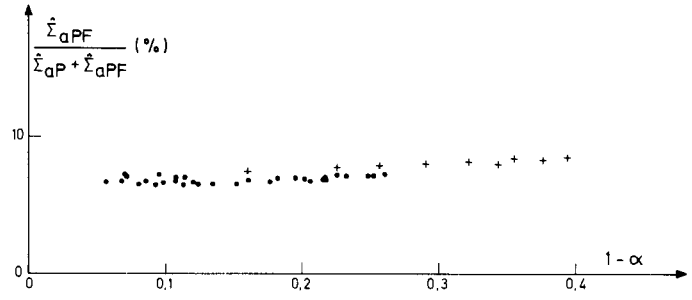

FIG. 3. $\hat{\Sigma}_{aPF}/(\hat{\Sigma}_{aP} + \hat{\Sigma}_{aPF})$ en fonction de l'appauvrissement en ^{235}U.

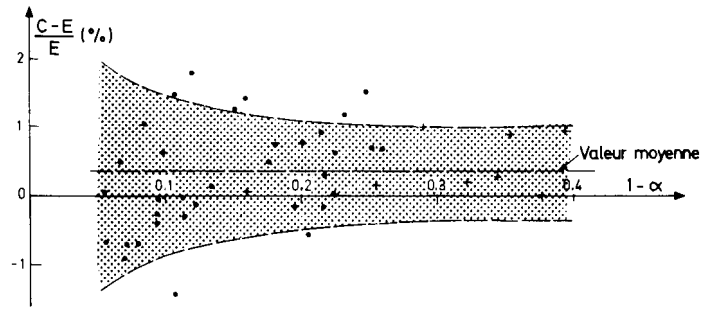

FIG.4. Ecart entre calcul et expérience sur $(\hat{\Sigma}_{aP} + \hat{\Sigma}_{aPF})/\hat{\Sigma}_a$ en fonction de l'appauvrissement en ^{235}U (C = valeur calculée, E = valeur mesurée).

En moyenne le calcul surestime la quantité $\hat{\Sigma}_{aP} + \hat{\Sigma}_{aPF}$ d'environ 0,35%. La marge d'incertitude sur cet écart varie entre ± 1,6% pour les faibles irradiation et ± 0,8% pour les plus fortes, définissant ainsi une zone d'incertitude qui contient la presque totalité des points. Exprimés en termes de capture totale des produits de fission, ces résultats montrent que le calcul surestime $\hat{\Sigma}_{aPF}$ d'environ 4,5% avec une marge d'incertitude allant de ± 21% aux plus faibles irradiations à ± 10% aux plus fortes.

On peut conclure de cette étude que les méthodes de calcul actuelles prédisent donc correctement les taux de réaction et la réactivité des combustibles irradiés à condition de diminuer globalement la capture totale des produits de fission d'environ 5%.

5. AJUSTEMENT DES CODES D'EVOLUTION A PARTIR DES ANALYSES CHIMIQUES ET ISOTOPIQUES DES COMBUSTIBLES IRRADIES [2]

La détermination des teneurs en noyaux lourds de combustibles irradiés dans des réacteurs de puissance permet de tester la validité des codes de calcul de l'évolution et d'ajuster éventuellement les principaux paramètres responsables de la formation et de la disparition des noyaux lourds. Les conditions d'irradiation étant connues avec une certaine incertitude liée aux variations de fonctionnement du réacteur, il est nécessaire d'analyser un grand nombre de combustibles pour obtenir des renseignements précis. On distinguera dans cette étude les résultats de l'ajustement, qui dépendent étroitement de la méthode de calcul que l'on utilise, et les méthodes mises en œuvre qui sont tout à fait générales: nous avons déjà parlé des analyses, nous exposerons ici les méthodes d'interprétation, puis les résultats obtenus.

5.1. Interprétation des analyses

On ajuste les paramètres suivants:

Intégrale de résonance de l'uranium-238: I_0

Sections à 2200 m/s: capture de ^{239}Pu (σ^0_{c9}), de ^{241}Pu (σ^0_{c1}), de ^{235}U (σ^0_{c5}), absorption de ^{240}Pu (σ^0_{a0}).

Dans une première méthode, on détermine pour chaque échantillon analysé les variations Δ qu'il faut appliquer à ces paramètres pour obtenir l'accord entre calcul et expérience sur les teneurs en noyaux lourds (^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{236}U). On trace ensuite les histogrammes relatifs aux Δ, ce qui fournit une valeur moyenne et un écart type qui permettent d'effectuer l'ajustement.

Dans une deuxième méthode, on traite par moindres carrés l'ensemble des résultats ponctuels relatifs à chaque analyse. On cherche les valeurs Δ des variations des paramètres qui décrivent l'ensemble des écarts sur les teneurs d'une façon optimale (détermination par la méthode du maximum de vraisemblance). Ces deux méthodes aboutissent à des résultats très voisins.

5.2. Echantillons analysés

Les 348 échantillons analysés sont prélevés sur des combustibles d'uranium naturel irradiés dans les réacteurs de puissance de la filière à uranium naturel-graphite-gaz. On trouvera aux tableaux VI et VII la répartition de ces échantillons et leurs teneurs en noyaux lourds aux irradiations maximales.

TABLEAU VI. ECHANTILLONS ANALYSES

Réacteur	Type de combustible.	Nombre d'échantillons analysés	Irradiation (MWj/t)
G2, G3	Barreau (Ø 28 mm)	48	600 à 4900
G3	Barreau (Ø 31 mm)	121	200 à 6500
CHINON 1	Tube (Ø 35-14 mm)	55	200 à 1400
CHINON 2	Tube (Ø 40-18 mm)	124	500 à 4500

TABLEAU VII. TENEURS EN NOYAUX LOURDS

Type de combustible	$N_5/N_8 \times 10^2$	$N_6/N_8 \times 10^2$	$N_9/N_8 \times 10^2$	$N_0/N_9 \times 10^2$	$N_1/N_9 \times 10^2$	$N_2/N_9 \times 10^2$
G2, G3, Ø 28 mm	0,37	0,06	0,19	29	5,5	0,9
G3, Ø 31 mm	0,30	0,07	0,22	40	8,0	1,9
CHINON 1	0,58	0	0,10	9,6	1,1	0
CHINON 2	0,41	0,05	0,21	26,4	5,9	0,9

TABLEAU VIII. VALEURS A 2200 m/s DES SECTIONS EFFICACES AJUSTEES (en barns)

σ^0_{C9}	σ^0_{C9}	σ^0_{a0}	σ^0_{C1}	σ^0_{C5}
Référence	273,9	297,3	382,3	100,6
$\Delta\sigma^0/\sigma^0 \times 10^{+2}$	- 5,0	-10,4	-12,3	- 2,0
Ecart type $\times 10^{+2}$	5,7	4,4	4,4	2,8
Ajustées	260,2	266,4	335,3	98,6

5.3. Ajustement du code d'évolution COREGRAF 2

COREGRAF 2 [6] est le code de calcul de réseau et d'évolution utilisé pour ce type de réacteurs; ses principales caractéristiques sont:

- calcul du spectre dans le domaine thermique par un code secondaire (voir paragraphe 2);
- calcul de l'intégrale de résonance à partir de l'expression classique $I_0 = A + B\sqrt{S/M}$, où A et B sont des paramètres ajustés sur des réseaux à uranium naturel;
- calcul d'évolution suivant la méthode exposée en 1.3 (32 produits de fission plus un pseudo-produit);
- sections efficaces: bibliothèque indiquée en 1, valeurs à 2200 m/s résultant de différents ajustements ou évaluations.

COREGRAF 2 permet des calculs très rapides mais il ne prend pas en compte les effets de la répartition des noyaux lourds et des produits de fission à l'intérieur du combustible. Ces effets peuvent être importants sur les sections effectives et on voit bien que la notion d'ajustement est ici étroitement liée au code utilisé et à un problème précis: la formation des noyaux lourds. La comparaison entre calcul et expérience sur les teneurs se fait en prenant la teneur en ^{235}U comme paramètre d'évolution.

Les résultats de l'ajustement sont les suivants:

a) Intégrale de résonance. La valeur de référence I_0 est calculée avec les paramètres A = 0,609, B = 28,87.
L'ajustement donne: $\Delta I_0/I_0 = -10,4 \times 10^{-2}$ (écart-type: $5,3 \times 10^{-2}$).

b) Sections efficaces. Elles figurent au tableau VIII.

Ces résultats sont très spécifiques au code d'évolution utilisé et ne peuvent être comparés directement aux résultats de caractère plus fondamental présentés aux paragraphes précédents.

CONCLUSION

On a cherché à faire de ce programme un ensemble logique, complet et structuré. En effet, dans une entreprise complexe comme celle-là, chaque ensemble de mesures ne peut éclairer qu'un aspect des problèmes,

et il faut donc les aborder de plusieurs côtés à la fois. D'autre part, si on veut pouvoir remonter aux données fondamentales qui sont nécessaires pour prédire correctement l'évolution des propriétés des combustibles, il faut faire des expériences « propres », dans des conditions aussi reproductibles que possible, et concentrer chaque fois l'effort sur les paramètres les plus sensibles. Enfin, un tel programme n'a de sens que si on développe simultanément les outils nécessaires à son interprétation.

Nous possédons désormais un ensemble de méthodes et de bibliothèques qui assurent un calcul correct, d'une part de la formation des noyaux lourds au cours de l'évolution du combustible, d'autre part des taux de réaction des combustibles contenant du plutonium et des combustibles irradiés. Il faut ajouter que, si le premier intérêt de ces travaux réside dans les résultats très complets qu'ils ont permis d'obtenir, ils ont été également l'occasion de la mise au point de nouvelles méthodes (méthodes de calcul, techniques expérimentales, interprétation rigoureuse des résultats obtenus) qui peuvent être appliquées à des problèmes différents et marquent un progrès important de la physique des réacteurs.

REFERENCES

[1] EURATOM, Etude neutronique du plutonium dans les réacteurs à neutrons thermiques, Rapport final, Contrat CEA-Euratom 037-60-12 RDF « Recyclage du plutonium », Rapport Euratom EUR-2853 f (1966).

[2] EURATOM, Etude neutronique du plutonium dans les réacteurs à neutrons thermiques, Rapport final, Contrat CEA-Euratom 002-64-9 TRUF «Recyclage du plutonium », Rapport Euratom EUR-4477 f (1970).

[3] TRETIAKOFF, O., « Physique des combustibles irradiés », Conf. int. util. énergie atom. fins pacif. (Actes Conf. Genève, 1964) 3, ONU, New York (1964) 357.

[4] LIVOLANT, M., Thermalisation ces neutrons, Bibliothèque de données, Méthodes et codes de calcul des cellules, Note CEA-N-1243 (1970).

[5] CADILHAC, M., SOULE, J.L., TRETIAKOFF, O., « Thermalisation et spectres de neutrons », Conf. int. util. énergie atom. fins pacif. (Actes Conf. Genève, 1964) 2, ONU, New York (1964) 153.

[6] COGNE, F., HOFFMANN, A., REUSS, P., COREGRAF 2: Code de réseaux et d'évolution des réseaux à graphite, Note CEA-N-1344 (1970).

[7] WALKER, W.H., Yields and Effective Cross-Sections of Fission Products and Pseudo-Fission Products, AECL-1054 (1960).

[8] ENGLAND, T.R., Time Dependent Fission Product Thermal and Resonance Absorption Cross-Sections, WAPD-TM-333 (1965).

[9] LAPONCHE, B., Méthode de l'échantillon équivalent pour interpréter les expériences d'oscillations d'échantillons fissiles dans une pile à neutrons thermiques, Rapport CEA-R-4152 (Thèse de Doctorat, Orsay, 1970).

[10] HANNA, G.C., et al., Revision of values for the 2200 m/s neutron constants for four fissile nuclides, Rev. Energie atom. (AIEA) 7 4 (1969) 3-93.

[11] NAUDET, R., « Interprétation d'expériences critiques portant sur des combustibles uranium-plutonium », Fuel Burn-Up Prediction in Thermal Reactors (C.r. Groupe d'étude Vienne, 1967), AIEA, Vienne (1968) 109.

STUDIES ON PLUTONIUM UTILIZATION IN POWER REACTORS AND SYSTEMS ANALYSIS OF LONG-TERM FUEL CYCLES

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Abstract–Résumé–Аннотация–Resumen

STUDIES ON PLUTONIUM UTILIZATION IN POWER REACTORS AND SYSTEMS ANALYSIS OF LONG-TERM FUEL CYCLES.

Studies on plutonium utilization in power reactors and systems analysis of long-term fuel cycles are reviewed. The evaluation of nuclear data and their conversion to group constants were processed on the basis of ENDF-A and -B data. Design assessments of plutonium recycling in LWRs were studied on the basis of direct substitution both in full and partial cores. A set of critical experiments on a PuO_2 - UO_2 fuelled core was also carried out to verify calculation methods. For HWR design, a computer code system, ATRASS, was developed, and the accuracy of the system is now being checked against experimental data. Through the assessment of work with the HWR, it was found that the plutonium self-sustaining cycle has the merit of reducing positive void reactivity, and may attain about 15 000 MWd/t fuel burn-up with natural uranium feeding. With the HTGR, the fuel cycle costs based on either low enriched uranium or on thorium mixed with highly enriched uranium become comparable when the price of natural uranium and the cost of separative work increase. When the LWR and the FBR are installed, about 6×10^5 metric tons of natural uranium will be consumed before 2030, when the nuclear power generating system will become self-supporting. To reduce the demand for natural uranium and decrease the fuel burn-up in the LWRs after the introduction of FBRs, improving the characteristics of the FBRs and introducing HWRs constitute an effective method. Time delay in the re-use of fissile material has a very large effect on natural uranium demand, so that it is very important to avoid it in the expanding power generating program. The role of Pu-recycled thermal reactors is discussed together with a strategic evaluation of the ATRs and FBRs for reduction of the uranium demand. The introduction of recycled Pu thermal reactors has the effect of reducing Pu pile-up; however, these reactors are responsible for a higher consumption of natural uranium. The effects of introducing thorium loaded HWRs (or HTGRs) and MSBRs into the LWR-FBR system are also presented.

UTILISATION DU PLUTONIUM DANS LES REACTEURS DE PUISSANCE ET ANALYSE DU CYCLE DU COMBUSTIBLE A LONG TERME: ETUDE D'ENSEMBLE.

Le mémoire passe en revue les études sur l'utilisation du plutonium dans les réacteurs de puissance et l'analyse du cycle du combustible à long terme. L'évaluation des constantes nucléaires et leur transformation en constantes de groupe ont été faites en fonction des données ENDF-A et -B. Les études sur le recyclage du plutonium dans les réacteurs à eau légère ont été faites en postulant un remplacement direct dans des cœurs tant complets que partiels. Une série d'expériences critiques sur un cœur chargé de PuO_2-UO_2 ont été faites pour vérifier les méthodes de calcul. Pour le calcul des réacteurs à eau lourde, un code d'ordinateur, ATRASS, a été élaboré; sa comparaison avec les données expérimentales en vue d'en vérifier la précision est actuellement en cours. L'analyse des expériences faites avec les réacteurs à eau lourde montre que le cycle auto-entretenu du plutonium a l'avantage de réduire la réactivité due aux vides et qu'un taux d'irradiation du combustible d'environ 15 000 MWj/t est possible avec une alimentation à l'uranium naturel. En ce qui concerne les réacteurs à haute température refroidis par un gaz, les coûts du cycle du combustible basés sur l'uranium légèrement enrichi ou sur le thorium mélangé avec de l'uranium fortement enrichi deviennent comparables lorsque le prix de l'uranium naturel et le coût du travail de séparation augmentent. Avec les

réacteurs à eau légère et les surgénérateurs rapides, environ 6×10^5 tonnes d'uranium naturel seront consommées avant que le système de production d'énergie nucléaire devienne autonome, vers l'an 2030. Pour réduire la consommation d'uranium naturel et abaisser le taux de combustion dans les réacteurs à eau légère après la mise en service des réacteurs surgénérateurs rapides, il serait utile d'améliorer les caractéristiques des surgénérateurs rapides et de multiplier le nombre des réacteurs à eau lourde. Les besoins en uranium naturel dépendent fortement des délais de recyclage des matières fissiles. Aussi importe-t-il d'éviter ces délais dans le programme élargi de production d'énergie. Le rôle des réacteurs thermiques avec recyclage de plutonium, ainsi que l'évaluation stratégique des réacteurs thermiques avancés et des surgénérateurs rapides sont examinés au point de vue de la réduction des besoins d'uranium naturel. L'adoption des réacteurs thermiques à recyclage de plutonium permet de réduire les stocks de cet élément; il s'ensuit cependant un accroissement de la consommation d'uranium naturel. On examine enfin les incidences de l'introduction de réacteurs à eau lourde (ou de réacteurs à haute température refroidis par un gaz) alimentés au thorium et des réacteurs surgénérateurs rapides à sels fondus dans le système «réacteurs à eau légère – réacteurs surgénérateurs rapides».

ИССЛЕДОВАНИЯ ПО ПРИМЕНЕНИЮ ПЛУТОНИЯ В ЭНЕРГЕТИЧЕСКИХ РЕАКТОРАХ И АНАЛИЗ СИСТЕМ С ДЛИТЕЛЬНЫМИ ТОПЛИВНЫМИ ЦИКЛАМИ.

В докладе сделан обзор исследований по применению плутония в энергетических реакторах и анализу систем с длительными топливными циклами. Оценка ядерных данных и их превращение в групповые константы были проведены на основе данных ENDF-A и B. Предварительное исследование методов переработки плутония для реакторов с обычной водой было изучено на основе прямой замены всего топлива или его части в активной зоне. Проведена также серия критических экспериментов с активной зоной на основе окисного топлива PuO_2-UO_2 для проверки расчетных методов. Для тяжеловодных реакторов была разработана специальная вычислительная программа ATRASS; в настоящее время проверяется точность этой программы при сравнении расчетных данных с экспериментальными. Благодаря оценке работы с тяжеловодными реакторами было обнаружено, что самоподдерживающийся плутониевый цикл способствует уменьшению положительного пустотного коэффициента реактивности и может обеспечить глубину выгорания примерно 15000 МВт·сутки/т с топливом из природного урана. Что касается высокотемпературных реакторов с газовым теплоносителем, то стоимости топливного цикла для топлива из слабообогащенного урана и для смеси из тория с высокообогащенным ураном становятся сравнимыми, если цены на природный уран и стоимость разделения изотопов будут повышаться. Для реакторов, охлаждаемых обычной водой, и быстрых реакторов-размножителей до 2030 года потребуется около $6 \cdot 10^5$ т природного урана, это будет продолжаться до тех пор, пока атомная энергетика не станет сама себя снабжать топливом. Для уменьшения потребления природного урана и снижения выгорания топлива в реакторах на обычной воде, до ввода в строй быстрых реакторов-размножителей, наиболее эффективным методом является усовершенствование параметров быстрых реакторов-размножителей и разработка тяжеловодных реакторов. Время, необходимое для повторного использования делящегося материала, оказывает весьма существенное влияние на потребности в природном уране, поэтому очень важно уменьшать это время для расширения программы производства электроэнергии. Обсуждается роль реакторов на тепловых нейтронах с топливом на основе переработанного плутония, а также стратегическое значение реакторов типа ATR и быстрых реакторов-размножителей для уменьшения потребностей в уране. Создание реакторов на тепловых нейтронах с топливом на основе переработанного плутония имеет значение для уменьшения накопления плутония, однако это приведет к еще большему потреблению природного урана. Обсуждаются также проблемы, связанные с разработкой тяжеловодных реакторов (или высокотемпературных реакторов с газовым теплоносителем) и реакторов типа MSBR с ториевым топливом.

ESTUDIOS SOBRE LA UTILIZACION DEL PLUTONIO EN REACTORES DE POTENCIA Y ANALISIS DEL CICLO DE COMBUSTIBLE A LARGO PLAZO.

Se revisan los estudios sobre la utilización del plutonio en reactores de potencia y los análisis del ciclo de combustible a largo plazo. Se han realizado evaluaciones de datos nucleares y sus conversiones a constantes de grupo, tomando como base los datos de ENDF – A y B. Se han estudiado estimaciones en proyectos del reciclado del plutonio en reactores de agua ligera, basándose en sustituciones directas tanto en núcleos completos como parciales. También se ha realizado una serie de experiencias críticas en un núcleo con combustible de PuO_2-UO_2 para comprobar los métodos de cálculo. Se ha desarrollado un sistema de programas de cálculo ATRASS para el diseño de reactores de agua pesada cuya precisión se está comprobando actualmente con datos experimentales. En los cálculos hechos en los reactores de agua pesada se ha comprobado que el ciclo autosostenido de plutonio tiene la ventaja de reducir el coeficiente de reactividad positiva de vacío y que es de

esperar un quemado del orden de 15 000 MWd/t con combustible de uranio natural. Por lo que se refiere a los reactores de gas a alta temperatura, los costos del ciclo del combustible, basados en uranio de bajo enriquecimiento o en torio mezclado con uranio de alto enriquecimiento, resultan comparables cuando suba el precio del uranio natural y el costo de los trabajos de separación. Si se instalan reactores de agua ligera y reactores reproductores rápidos, se consumirán cerca de 6×10^5 toneladas de uranio natural antes del año 2030, fecha en la que el sistema de generación de potencia nuclear llegará a ser automantenido. Para disminuir la demanda de uranio natural y reducir el grado de quemado del combustible en los reactores de agua ligera después de la introducción de los rápidos, constituye un método eficaz perfeccionar las características de los reactores reproductores rápidos e implantar los reactores de agua pesada. El retraso en la reutilización del material fisionable influye mucho sobre la demanda de uranio natural, de tal modo que su supresión se hace muy importante en el programa de producción de energía. Se discute la influencia de los reactores térmicos con reciclado de plutonio, junto con la evaluación estratégica de los reactores térmicos avanzados y rápidos, sobre la reducción de la demanda de uranio. La introducción de reactores térmicos con reciclado del plutonio provoca una reducción de la acumulación de plutonio, pero, al mismo tiempo, un consumo mayor de uranio natural. También se presenta el efecto de la introducción de reactores de agua pesada cargados con torio (o de gas a alta temperatura) y de reactores reproductores de sal fundida en el sistema de reactores de agua ligera y reproductores rápidos.

1. INTRODUCTION

In its latest estimation, the Japan Atomic Industrial Forum, Inc., forecasts that there will be a nuclear power generating capacity of 27-30 GW(e) in 1980 and of 200-240 GW(e) in 2000. These capacities account for about 16% and 50% of the total electric generating capacity in the respective years. In attaining such a capacity, the procurement of nuclear fuel and its effective utilization will become a serious problem. In fact, extensive research and development work on plutonium utilization is now proceeding in Japan, in relation to the development of both FBRs and ATRs and also for the purpose of establishing a plutonium recycling technique in LWRs. Furthermore, the construction of a fuel reprocessing plant is under way in the Tokai area.

This paper deals first with the studies on the core performances of power reactors and then with strategic systems analysis of long-term fuel cycles carried out at the Japan Atomic Energy Research Institute (JAERI). For the former item, the research activities on power reactor assessments, such as fuel management and fuel cycle analysis, calculation methods and computer codes, nuclear data and critical experiments, etc., are briefly reviewed. For the latter item, the systems analysis of the long-term fuel cycle, based on the latest estimation of nuclear power generating capacity, is explained with the intention of presenting the importance of strategic reactor installation through the demand for natural uranium and separative work.

2. STUDIES ON CORE PERFORMANCES

2.1. Light-water reactors

Since light-water reactors are now in commercial operation, optimal fuel management is very important. To obtain detailed information on this subject, far-reaching improvements of nuclear data and reactor calculation methods have been effected in Japan.

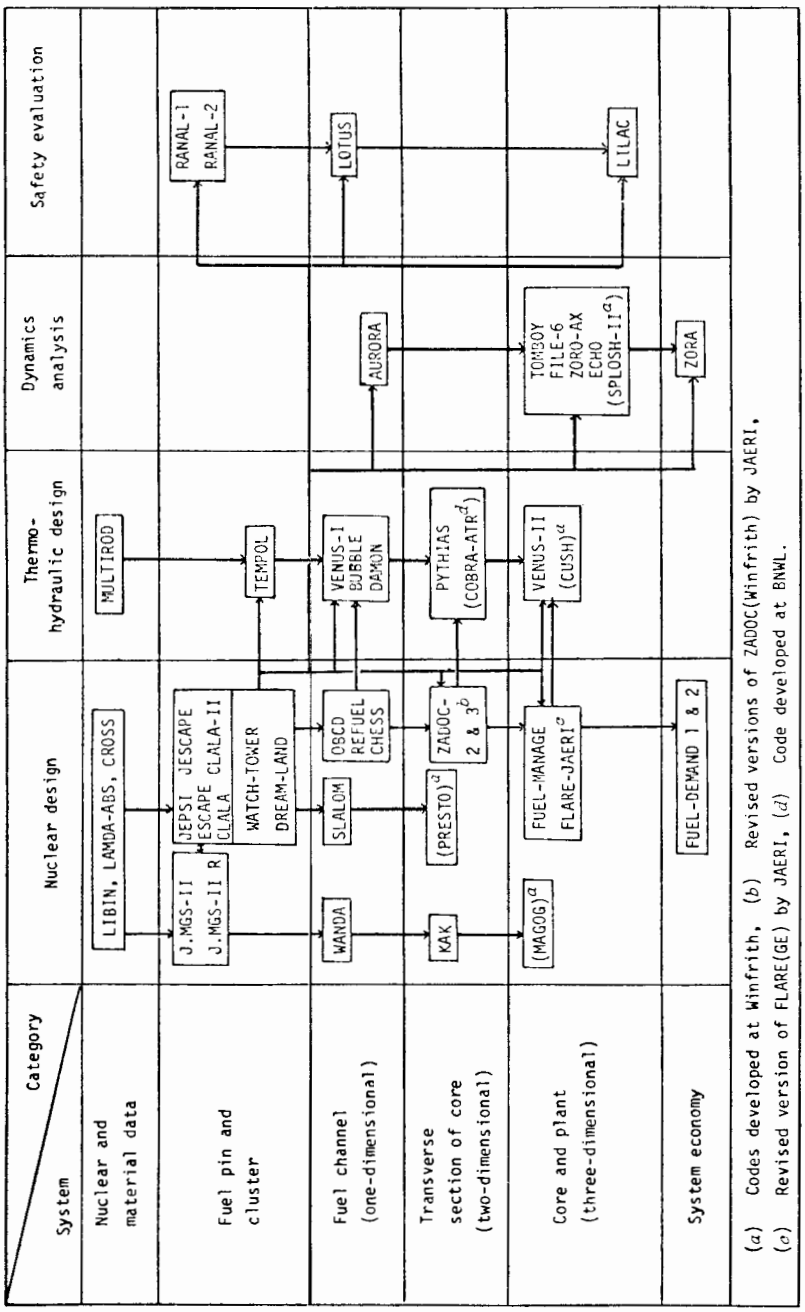

FIG. 1. Constitution of ATRASS code system for HWRs.

Recent progress includes the following developments, among others: (1) For the evaluation of nuclear data and their conversion to group constants, both the MUFT and GAM types of multi-group constants were processed on the basis of ENDF-A and B data, and then checked against critical experiments [1, 2]. (2) To make more precise calculations of neutron transport, several computer codes on first flight collision probabilities were developed, and their applications to various lattice systems, including cluster and tube-in-shell, are now conventionally in use [3, 4]. (3) Design assessments of plutonium recycling in LWRs have been studied on the basis of direct substitution both in full and partial cores [5-7]. One of the recent studies was the survey calculation for PuO_2-UO_2 cores in BWRs, which was performed to determine the average Pu enrichment necessary to achieve the same fuel burn-up as UO_2 cores by changing the rod diameter, the moderator to fuel volume ratio and the Pu isotopic concentrations. Pu distributions in an assembly were also analysed in minimizing the local power peaking factor and a Pu loading scheme was determined. Effects of Pu recycle on kinetic characteristics were assessed through the reactivity coefficient and the control-rod worth [6]. Another study was the survey of local power variation with burn-up. The amount of Pu loaded is assumed to be a quarter of the total fuel. Two categories of loading were considered: namely (a) one PuO_2-UO_2 assembly with three UO_2 assemblies, and (b) one mixed assembly equivalent to (a) in terms of Pu to U atomic number ratio. Two-dimensional burn-up calculation for a set of four assemblies in a scattered pattern confirmed that loading category (b) is preferable [7]. (4) Performance tests of Pu-bearing assemblies in JPDR (BWR) and Saxton (PWR) in the United States of America are planned. Further, a set of critical experiments on two regional water lattices with a central PuO_2-UO_2 region and a surrounding UO_2 region has been conducted. Two types of PuO_2-UO_2 fuel rod, one for BWRs and the other for PWRs, were employed. In these experiments, k_{eff}, power and flux distributions and β/l were measured parametrically with changes in volume ratio and poison concentration. The results were analysed to verify the present state of the art of PuO_2-UO_2 lattice calculation [8].

2.2. Heavy-water reactors

Research and development work on HWRs in Japan was initiated in 1963. A 165-MW(e) prototype of a boiling-light-water-cooled reactor is now under construction. From the beginning of the project, assessment work on core performances and the fuel cycle has been performed in JAERI. For this purpose, a versatile code system, ATRASS, was developed [9]. The outline of the system is presented in Fig. 1. The computer codes in the system are grouped into four categories, i.e. nuclear, thermo-hydraulic, dynamics, and safety. Of these, WATCH-TOWER, OBCD, REFUEL, CHESS, ZADOC-2 and -3 and FLARE-JAERI are frequently used for core burn-up analyses. The accuracies of the code system are being checked against the experimental data from several critical facilities [10].

The following results were obtained with this assessment work: (1) The use of 1.2-1.5% enriched uranium is more profitable than the use of natural uranium from the point of view of safety and economy. (2) With the plutonium self-sustaining cycle based on natural uranium feeding, a burn-up of about 15 000 MWd/t may be obtainable in a HWR of the BLW type with a capacity

TABLE I. FUEL CYCLE COST BASIS AND HTGR CORE PERFORMANCE

(A) Cost basis

Nat. U	10$/lb	U-233/U-235 parity	14/12	Interest rate	8%
Enrichment	30$/kgSWU	Thorium	17$/kg	Fuel loss	1.0 1.5%[a]
Pu value	10$/g-f	Load factor	80%	Lead time max.	3 yr[b]

(B) Core performance

Items	LEU cycle				HEU[c] - Th cycle						
					Cycle No.			1	2	3	4
Av. power density (kW/l)	6	6	6	6	6	6	8	8	8	8	8
Av. specific power (kW/kgHM)	78	78	62	62	47	31	62	42	42	42	42
Fuel enrich. (wt %) or Th/U	8	10	8	10	10	10	10	10	10	10	10
C. P. F. packing fraction (vol. %)	20	20	25	25	30	40	30	40	40	40	40
Nc/Nu or Nc/N_{Th}	360	360	288	288	315	238	315	238	238	238	238
Nc/Nu-235	4500	3600	3600	2880	3390	2560	3390	2560	5530	6960	8200
Fuel burn-up (MWd/kgHM)	94.6	127.5	71.1	98.7	117.9	124.1	110.5	116.4	87.4	69.9	58.6
U-235 in depleted U (g/kgHM)	7.4	5.5	20.0	18.0	10	20	20	4.5	5.0	6.7	7.5
Fissile Pu or U-233 (g/kgHM)	10.0	8.0	16.2	18.5	10.7	10.7	11.9	12.2	18.5	19.0	19.1

a 1% (UF_6-UO_2), 1% (fabrication), 1.5% (reprocessing).

b 1 yr (nat. U ore), 1 yr (conversion and enrichment), 1 yr (fabrication), 1 yr (reprocessing).

c 93% enriched U.

of 500-1000 MW(e). This cycle has the further advantage of possessing a large negative void coefficient, and it may be switched over to the plutonium-producing cycle without any difficulty [11, 12]. (3) A thorium-loaded HWR with heavy-water coolant may be operated as a near breeder. If the reactor is enriched with ^{235}U it will be an excellent ^{233}U producer, which facilitates the introduction of the thermal breeding cycle [13].

The relationship between the refuelling scheme and the burn-up performance has been studied in detail with the following results: (1) In-out and out-in refuellings have little merit in HWRs because they induce large power peaking in the core central region and at the core periphery, which is enhanced when the burn-up of the discharged fuel increases [14]. (2) In the axially refuelled HWR of the BLW type with a large positive void coefficient, power trapping may occur in the core central region when the coolant flows in the same direction as the fuels move. (3) In a radially scattered refuelled core, the stretch β of fuel burn-up is well represented by the relation $\beta = 2Cn/(n+1)$, where n is the number of the refuelling batches of the core and C is a form factor, depending both on fuel loading modes and on core size, and, for the 165-MW(e) prototype core, C is found to be 0.9-0.95 [14]. (4) In the continuously scattered refuelled HWRs, poor ordering of fuel loading deteriorates power distribution greatly, so that the optimization of the ordering becomes very important [15].

2.3. High-temperature reactors

With respect to high-temperature gas-cooled reactors, JAERI had the experience of promoting the Semi-Homogeneous Reactor development project 14 years ago [16]. In recent years, this type of reactor became attractive again in Japan because of the interest in direct utilization of

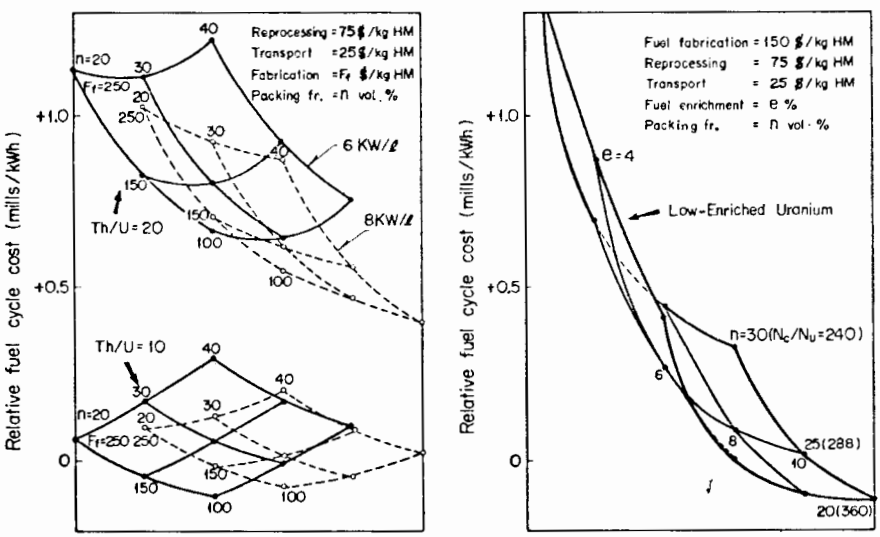

FIG. 2. Relative fuel costs for a HTGR with low-enriched uranium or highly enriched uranium-thorium loading.

nuclear energy for heat consuming industries, such as iron manufacturing. In this connection, JAERI completed the preliminary design study of an experimental multi-purpose high-temperature gas-cooled reactor early in 1971.

An evaluation of the fuel cycle was also made for several kinds of fuel use. A 2386-MW thermal power reactor was selected in the study and it employed prismatic fuel elements. The core parameters and the cost evaluation basis are given in Table I. The results of fuel cycle costing are presented in Fig. 2: (1) With a once-through uranium cycle, about 8 to 10% of the fuel enrichment is necessary to make the cycle economical, and the carbon to uranium atomic number ratio may have a relatively high value, such as 280 to 360. These values correspond to 25 to 20 vol.% of the coated particle fuel packing fraction, respectively. (2) With a once-through thorium cycle, a ratio of 10 of thorium to uranium atoms is preferable. We assume a relatively high cost for natural uranium and for fuel enrichment in the survey, so that this cycle has almost the same fuel cycle cost as a low enriched uranium cycle. (3) By recycling ^{233}U, the fuel cycle cost may be further decreased. One example, in which complete recycling of ^{233}U is employed, shows that its fuel cycle cost is about 0.2 mills/kWh less than that of a once-through cycle.

2.4. Fast-breeder reactors

JAERI has carried out two kinds of design study on 1000-MW(e) Na-cooled, oxide-fuelled, fast-breeder reactors. The first design, made in 1966, was aimed at obtaining a high breeding ratio and has two zoned cores with different plutonium enrichments [17]. A burn-up analysis of this reactor was done by several methods, among which an improved perturbation calculation was found to be most convenient [18]. Three- or four-batch scattered refuelling is assumed for core fuels. Blanket management was also studied in several patterns. A typical cycle gives a doubling time of 6.8 years at an 80% load factor.

The second design, made in 1967, placed emphasis on safety and has an annular core with an inner sodium region [19]. Because of increased neutron leakage, the doubling time becomes 12.7 years, which is rather conservative compared with current foreign designs. Burn-up calculations were done with 25-group-1-dimensional or 6-group-2-dimensional code, and JAERI recently developed their own cross-section set JAERI-FAST [20], which is being checked through critical experiments [21].

3. STRATEGIC SYSTEMS ANALYSIS OF THE LONG-TERM FUEL CYCLE

Studies on the long-term fuel cycle have been made by several authors [22-25]. The main items surveyed were the demand for natural uranium and separative work, system cost, etc. In this paper, a strategic systems analysis recently made by the authors is presented.

3.1. Nuclear power generating capacity

In Japan, the nuclear power generating capacities up to 1975 are already almost fixed. The main reactors are LWRs, but, after 1976, the reactor

TABLE II. ANTICIPATED NUCLEAR POWER GENERATING CAPACITY

Year	Nuclear power capacity (10^3 MW(e))	Average growth rate (%/yr)
1975	8.5	25.1
1980	27	17.3
1985	60	11.6
1990	104	8.5
1995	156	7.1
2000	220	5.5
2010	376	4.5
2020	584	3.5
2030	823	3.0
2040	1106	3.0
2050	1487	

TABLE III. CHARACTERISTICS OF BREEDER REACTORS

Items of performance	FBR	FBR(H)	MSBR
Specific power (kW/kg)	134	134	32
Fuel burn-up (MWd/kg)	93.0	66.3	-
Effective breeding ratio	1.20	1.27	1.06
Plant efficiency (%)	43.5	43.5	44.4
Fissile inventory (kg/MW(e))	2.17	1.90	1.40
Fissile yield (kg/MW(e)yr)	0.171	0.278	0.0458
Reprocess. time delay (yr)	1.0	1.0	0.0
References	19	17	27

types are not completely fixed so that it will be possible to introduce HWRs, Pu recycling LWRs, and, later on, perhaps from 1986, breeder reactors.

In the present study, the forecast of the total installed capacity of nuclear power by the year 2000 is adapted to accepted figures which are shown in Table II. After 2000, the growth rate of the nuclear capacity is assumed to be 5.5% per year for the first ten years and 4.5%, 3.5%, 3.0%, 3.0% for the following four decades until 2050. Throughout the entire period, reactor life and its load factor are taken, for the sake of simplicity, as 20 years and 80%, irrespective of reactor types.

TABLE IV. CHARACTERISTICS OF CONVERTER REACTORS

Items of performance	BWR	PWR	BWR (low burn-up)	PWR (low burn-up)	BWR (Pu)	PWR (Pu)	HWR (SEU)	HWR (Pu)	HWR (Th)	HTGR (LEU)	HTGR (Th)
Initial enr. (%)	2.19	2.37	1.45	1.85	1.558[a]	2.62[b]	1.20	1.20	1.90	6.0	4.76
Feed enr. (%)	2.56	3.30	1.85	2.06	1.558[a]	2.62[b]	1.20	0.588[b]	1.90	6.0	4.76
Pu discharge[e] (%)	0.554	0.647	0.439	0.517	0.948	1.08	0.481	0.588	1.17[c]	1.24	1.22[c]
Specific power (kW/kg)	22.0	34.8	22.0	34.8	20.0	34.8	15.6	15.6	17.0	62.3	37.5
Burn-up (MWd/kg)	27.5	33.0	14.0	16.0	23.5	30.0	19.7	15.4	22.0	45.9	30.3
Plant efficiency (%)	32.8	32.5	32.8	32.5	32.8	32.5	32.0	32.0	32.0	44.0	44.0
kg fuel/MW(e)	138.7	88.4	138.7	88.4	152.4	152.4	200.8	200.8	183.4	36.5	60.6
kg feed[d]/MW(e) yr	33.2	27.7	63.6	56.1	37.9	30.0	46.3	59.2	41.0	14.5	21.9
References	26	26	26	26	5	-	11	11	13	-	-

[a] Pu-enrichment with 0.89% E.U.

[b] Pu enrichment with natural uranium.

[c] U-233 discharge.

[d] At 80% load factor.

[e] Fissile Pu.

3.2. Reactor data

The reactor data used in the analysis are summarized in Tables III and IV. The characteristics of the LWRs are taken from the design data by the USAEC Reactor Development Technology Division [26]. Pu recycling LWRs are based on the data in Ref. [5] with some modifications. For the HWRs, two types are taken into consideration, one based on a Pu self-sustaining cycle [11] and the other on a 1.2% enriched uranium cycle [11]. JAERI's second design of a 1000-MW(e) FBR is taken as the reference FBR and, for sensitivity analyses, we introduced two kinds of modification: one in breeding gain and the other in specific power. For the purpose of comparison, JAERI's first design is occasionally used as a high gain breeder FBR(H). A further LWR with low fuel burn-up [26] was also introduced into the system. To investigate the effect of the thorium cycle, three types of thorium reactor are introduced as well. These are: a HWR with $^{235}UO_2$-ThO_2 fuel [13], HTGRs with 93% enriched UC_2+ThC_2 fuel designed by JAERI, and the molten salt breeder reactor designed by ORNL in 1969 [27], which has rather poor breeding gain compared with the earlier design [28] but the merit of continuous reprocessing is still retained.

3.3. Assumed reactor installation

For the uranium-plutonium cycle, the ratios of the installation capacities for thermal converters are given annually as inputs between 1970 and 1985. After 1986, the annual installed capacity of FBRs is determined by the amount of Pu available. Thermal converters are to be installed only when the available amount of Pu is not sufficient, and in this case the rest of the installed capacity is shared by thermal converters, whose ratios are given as inputs.

When the thorium cycle is introduced, MSBRs will be installed after 1996 or, occasionally, after 1986 for the purpose of comparison. Their share in the installed capacity will be determined, after that of the FBRs has first been fixed, by the amounts of ^{233}U available at that time. The above procedure for determining the shares of the different reactor types is called the material balance method.

Another way of determining the shares, the linear programming method, is used to optimize the cumulative demand for natural uranium and to minimize the system cost under the constraints imposed by excess plutonium kept at specified values and the introduction timing of breeder reactors. The computer code FUEL-DEMAND-II [29, 30] can be used in either method.

3.4. Results and discussion

3.4.1. LWR-FBR system

If only LWR and FBR reactors are introduced and if there is no improvement in their characteristics, the fuel cycle situation in Japan will proceed as in Fig. 3. It can be noticed that the supply of Pu by LWRs is insufficient for the complete introduction of FBRs between 1988 and 2020, and a total capacity of 232×10^3 MW(e) of LWRs will be installed before attainment

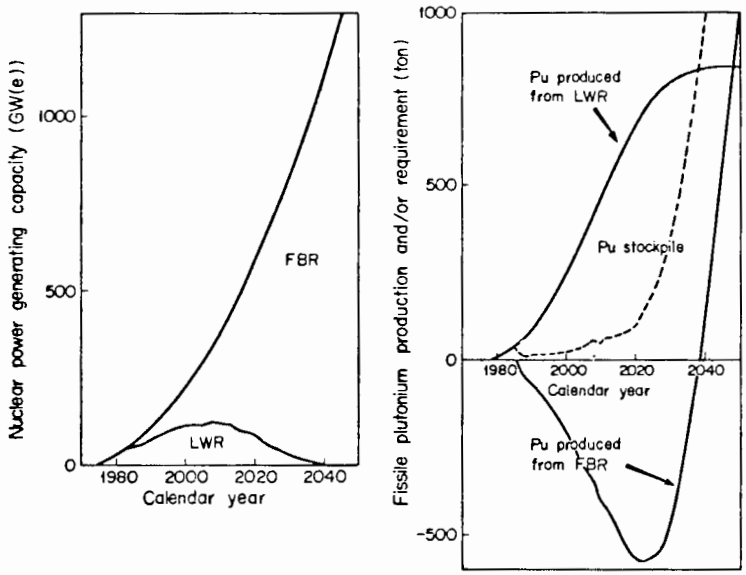

FIG. 3. Capacity and fissile material balance for a LWR-FBR system (case A).

of the self-supporting breeding cycle. As a result, the cumulative demand for natural uranium will be 632×10^3 metric tons of metal and for separative work 539×10^3 metric tons. The peak capacity of the LWRs will be 131×10^3 MW(e) in 2007 when the annual demand for separative work will exceed 16×10^3 metric tons/yr. In Fig. 3, there is a sharp contrast between the periods before and after 2020, when the FBRs will change from consumers of plutonium to suppliers.

3.4.2. Modifications to the LWR-FBR system

The demand for uranium and separative work in the above-mentioned reference case does not make for optimism. To relieve the situation within the framework of the LWR-FBR system, sensitivity analyses have been made.

One of these is to improve the effective breeding ratio[1], EBR, from 1.20 to 1.25. The cumulative demand for natural uranium will then decrease to 470×10^3 tonnes and for separative work to 399×10^3 tonnes. As opposed to this, if the EBR decreases to 1.15, the demand will increase to 902×10^3 tonnes and 769×10^3 tonnes, respectively, showing an approximately inverse proportionality between breeding gain and cumulative demand, and we will have a coefficient of about -40×10^3 t/0.01 of EBR. Another analysis is to improve the specific power. If the original design value of 59.8 kW(e)/kg fuel is changed by ±10%, the cumulative demand for natural uranium and for separative work will vary by about ±5% as shown in Table V.

[1] The effective breeding ratio is defined here as the annual discharge rate of fissile material divided by the annual charge rate.

If the improvements to the FBR characteristics do not proceed well, one easy way to improve the fuel economy is to reduce the fuel burn-up of the LWRs after the introduction of the FBRs. For this purpose, LWRs with a fuel burn-up reduced to about half of the original value will be introduced after 1986. The effect of such a modification is fairly large, as is shown in case G of Table V and in Fig. 4. The result is similar to that when a high gain FBR is introduced (case F of Table V).

Another modification to the LWR-FBR system is the introduction of LWRs (Pu), whose shares are determined by inputs or by the linear programming method. With the former, if LWRs (Pu) are introduced in the order LWR, LWR (Pu), and, lastly, FBR, the result is a reduction of the Pu pile-up between 1976 and 1985, but, in the long run, more uranium consumption. With the latter, one interesting result obtained is that the optimal introduction date of LWRs (Pu) to minimize the cumulative demand for natural uranium is after the introduction of the FBRs, and the cumulative demand for both natural uranium and separative work, and also for annual separative work, is somewhat reduced when compared with the reference LWR-FBR system.

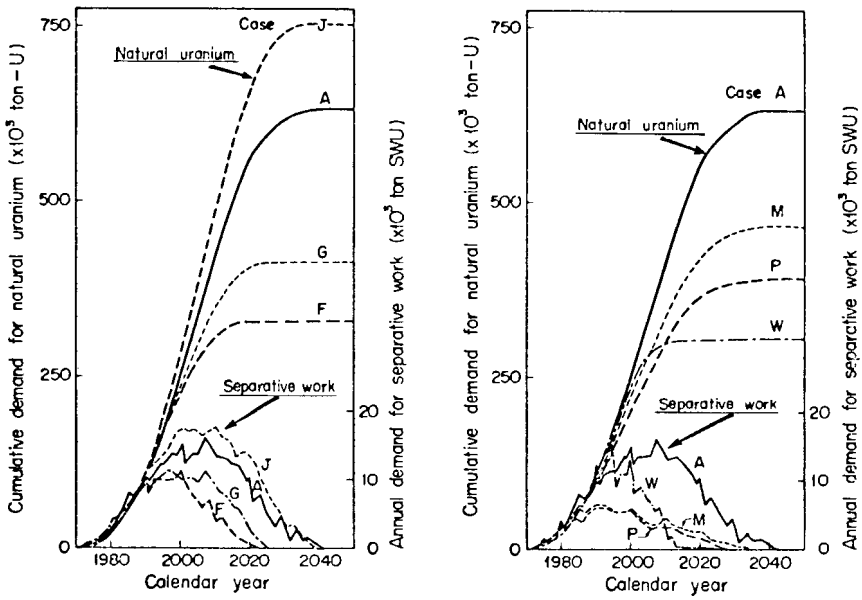

FIG. 4. Cumulative demand for natural uranium and annual separative work.

3.4.3. Effect of HWRs

In reducing the demand for separative work, the introduction of HWRs is quite effective. Cases L through R in Table V and Fig. 4 illustrate this. In case M, fifty-fifty installations of LWRs and HWRs (Pu) are postulated between 1978 and 1985, and after 1986 only HWRs (SEU) are supposed to

TABLE V. RESULTS OF LONG-TERM FUEL CYCLE ANALYSIS

LWR from 1970, LWR(Pu) from 1976 HWR from 1978, HTGR from 1981 FBR from 1986, MSBR from 1996		Peak capacity of converter		Cumulative nat. U	Separative work			Pu (or U-233) pile-up			
					Cum.	Annual peak		Fissile Pu (t)		U-233 (t)	
Case, combination of reactors	Date of installation	10^3 MW(e)	(yr)	10^3 t	10^3 t	10^3 t/yr	(yr)	1985	2000	1985	2000
A. LWR→FBR, LWR	1970 - 1986	131	(2007)	632	539	16.1	(2007)	31	24	-	-
B. LWR→FBR (EBR+0.05), LWR	1970 - 1986	110	(2007)	470	339	14.0	(2000)	31	27	-	-
C. LWR→FBR (EBR-0.05), LWR	1970 - 1986	154	(2020)	902	769	19.0	(2020)	31	22	-	-
D. LWR→FBR (s. p. +10%), LWR	1970 - 1986	115	(2007)	507	433	14.1	(2007)	31	24	-	-
E. LWR→FBR (s. p. -10%), LWR	1970 - 1986	147	(2007)	761	648	18.3	(2010)	31	25	-	-
F. LWR→FBR(H), LWR	1970 - 1986	89	(1998)	330	281	11.4	(2000)	31	30	-	-
G. LWR→FBR, LWR (low burn-up)	1970 - 1986	106	(1999)	414	359	11.8	(2000)	31	29	-	-
H. LWR→FBR(Nodelay), LWR	1970 - 1986	76	(1995)	288	246	9.4	(1995)	31	36	-	-
I. LWR→FBR (2-yr delay), LWR	1970 - 1986	200	(2018)	1158	988	24.3	(2017)	31	19	-	-
J. LWR→LWR, LWR(Pu)→FBR, LWR	1970-1976-1986	149	(2010)	756	640	17.8	(2010)	4	24	-	-
K[a] LWR, LWR(Pu), FBR	1970 - 1986	116	(2000)	610	446	13.0	(2010)	31	31	-	-
L. HWR(SEU)→FBR	1970 - 1986	94	(2007)	330	118	4.1	(2000)	48	28	-	-
M. LWR→LWR, HWR(Pu)→FBR, HWR(SEU)	1970-1978-1986	129	(2000)	468	206	6.0	(1992)	18	28	-	-
N. LWR→LWR, HWR(Pu)→FBR, LWR, HWR(SEU)	1970-1978-1986	135	(2000)	578	369	10.8	(2000)	18	26	-	-
O. LWR→LWR, HWR(Pu)→FBR, LWR, HWR(Pu)	1970-1978-1986	219	(2020)	901	534	14.3	(2010)	18	16	-	-
P. LWR→LWR, HWR(SEU)→FBR, HWR(SEU)	1970-1978-1986	104	(2007)	391	188	6.5	(1990)	36	28	-	-
Q. LWR→LWR, HWR(SEU)→FBR(H), HWR(SEU)	1970-1978-1986	79	(1995)	231	132	6.3	(1990)	36	34	-	-
R[a] LWR, LWR(Pu), HWR, HWR(Pu), FBR	1970 - 1986	122	(2000)	441	205	7.2	(1994)	2	0	-	-
S. HTGR→FBR, HTGR	1970 - 1986	124	(2007)	479	548	16.9	(2007)	39	25	-	-
T. HTGR[b]→FBR, HTGR[b]	1970 - 1986	80	(2007)	341	393	12.8	(2007)	57	30	-	-
U. HWR(Th)→MSBR	1970 - 1986	60	(1985)	158	205	14.2	(1985)	-	-	67	257
V. HTGR(Th)→MSBR	1970 - 1986	60	(1985)	294	388	5.3	(1985)	-	-	58	78
W[a] LWR, HWR(Th), FBR, MSBR	1970-1986-1996	67	(1990)	307	273	13.9	(1994)	0	7	0	13

[a] Linear programming to minimize cumulative demand for nat. U.

[b] Specific power and fuel burn-up are taken as 51.9 kW/kgHM and 31.9 MWd/kgHM, but enrichment retains 6%.

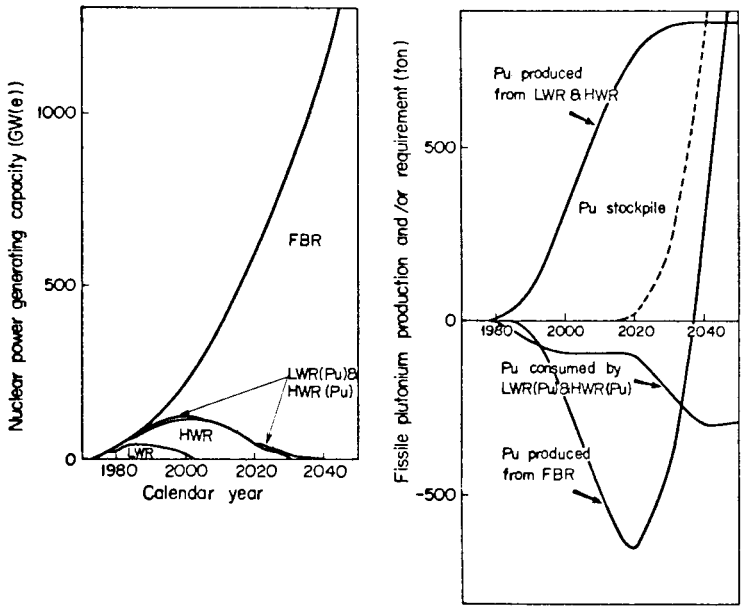

FIG. 5. Capacity and fissile material balance for LWRs, LWRs (Pu), HWRs, HWRs (Pu) and FBRs determined by the linear programming method (case R).

be built as converters. The cumulative demand for natural uranium is estimated as 486×10^3 tonnes, which is not less than in the case of the LWR-FBR (H) system, but the separative work amounts to only 206×10^3 tonnes (cumulative) and 6×10^3 tonnes/yr (annual peak in 1993).

The installation of Pu self-sustaining HWRs has the merit of reducing both Pu pile-up before 1986 and the annual peak for separative work, but such continuous installation after 1986 would result in a shortage of Pu for the installation of FBRs. In Table V, cases N and O are also shown, in which LWRs are continuously developed on a fifty-fifty installation basis with HWRs (SEU) even after 1986. These do not have any particular advantage compared with the LWR-FBR case, but show the importance of switching from LWRs to HWRs.

To optimize the installation of LWRs and HWRs, survey calculations with the linear programming method were made. One example is shown in Fig. 5, which is to minimize the cumulative demand for natural uranium under the constraint imposed by annual excess Pu pile-up, held within 1 tonne before 1986 and 500 tonnes afterwards. Six kinds of converter, BWR, PWR, BWR (Pu), PWR (Pu), HWR and HWR (Pu) can be installed at any time, but the FBR only after 1986. The cumulative demand for natural uranium and for separative work is 441×10^3 tonnes and 205×10^3 tonnes, respectively, and for annual separative work 7×10^3 tonnes/yr which is a remarkable reduction.

3.4.4. Introduction of the thorium cycle

A partial introduction of the thorium cycle into the U-Pu system is also effective for reducing uranium demand. Some results of this are given in Table V, including a result obtained with the linear programming method, which shows the complementary coexistence of Pu and Th cycles.

3.4.5. Importance of time delay

In the foregoing sections, the time delay for re-use of Pu discharged from the FBRs is fixed at one year for cooling, reprocessing and re-fabricating the fuel. But, for ^{233}U from the MSBRs, no time delay is assumed in virtue of continuous on-site reprocessing.

Since the time delay plays an important role in the expanding power program, sensitivity analyses were made to demonstrate this effect. The results are summarized in Table V, in which the time delay for FBRs is varied from zero to two years. The effect is drastic. If there is a two-year delay, forecasts for the long-term fuel economy become pessimistic. The cumulative demand for natural uranium exceeds 10^6 tonnes and will still be increasing in 2050. A similar situation exists with the MSBR system if there is a one-year delay. The merits of introducing the thorium cycle will be greatly reduced in this case.

4. CONCLUSION

From the surveys of work on power reactor assessment and of systems analysis of the long-term fuel cycle, the following may be concluded: (1) The more power reactors there are in commercial operation, the more important fuel management becomes, and, to assess this with high accuracy, the refinements of consistent sets of multi-group constants and of calculation methods, as well as experimental confirmations of these, are much more necessary. (2) From the studies on Pu loading schemes in boiling light-water reactors, one PuO_2-UO_2 mixed assembly is preferable to one PuO_2-UO_2 assembly with three UO_2 assemblies from the viewpoint of the power peaking factor. (3) Through the assessment studies on HWRs of the BLW type, the plutonium self-sustaining cycle was found to have the merit of reducing positive void reactivity, and it can attain about 15 000 MWd/t fuel burn-up with natural uranium feeding. This cycle may be switched over to the uranium cycle without any difficulty. (4) For HTGRs, the fuel costs, in a once-through cycle based either on low-enriched uranium or on thorium mixed with highly enriched uranium, become comparable to each other when the price of natural uranium and the cost of separative work increase. However, about 0.2 mills/kWh of cost saving may be attained by complete recycling of ^{233}U. (5) To reduce the demand for natural uranium, the characteristics of FBRs should be improved and HWRs introduced, in addition to reduction of the fuel burn-up of the LWRs after introduction of the FBRs. To reduce separative work, the introduction of HWRs is very effective. (6) Time delay in the re-use of fissile material has a very large effect on natural uranium demand, and it should thus be reduced to a minimum to ensure the success of an expanding power generating program. Where the thorium cycle is concerned, it can coexist naturally with the uranium-plutonium cycle. The above results underline the importance of system synthesis between converters and breeders.

ACKNOWLEDGEMENTS

The authors gratefully acknowledge the advice and encouragement received from Dr. R. Ueda and Dr. K. Nakamura and the helpful discussions with Dr. M. Nozawa.

REFERENCES

[1] KATURAGI, S., et al., JAERI Rep. JAERI-1176 (1969).
[2] NAITO, Y., TURUTA, H., SUGI, T., JAERI Rep. JAERI-M 2761 (1967).
[3] TSUCHIHASHI, K., JAERI Rep. JAERI-1196 (1971).
[4] TSUCHIHASHI, K., JAERI Rep. JAERI-M 3034 (1968).
[5] UEMATSU, K., YUMOTO, R., YASUKAWA, S., "Aspects of plutonium fuel design for a large boiling water reactor", Plutonium as a Reactor Fuel (Proc. Symp. Brussels, 1967), IAEA, Vienna (1967) 117.
[6] MATUOKA, K., private communication.
[7] MIZUTA, H., private communication.
[8] KOBAYASHI, I., YUMOTO, R., private communication, MATUURA, S., et al., JAERI Rep. JAERI-M 4259 (1970).
[9] YASUKAWA, S., et al., JAERI Rep. JAERI-M 3971 (1970).
[10] SHINDO, R., HIRANO, M., ARUGA, T., JAERI Rep. JAERI-M 3969, M 3973 (1970).
[11] SAWAI, S., YASUKAWA, S., SHINODA, W., "Design of Pu self-sustaining reactor with D_2O moderator and boiling H_2O coolant", Heavy-Water Power Reactors (Proc. Symp. Vienna, 1967), IAEA, Vienna (1968) 285.
[12] YASUKAWA, S., SHINDO, R., "Uses of plutonium fuel in pressure-tube-type, heavy-water-moderated thermal reactors", Plutonium as a Reactor Fuel (Proc. Conf. Brussels, 1967), IAEA, Vienna (1967) 561.
[13] FURUHASHI, A., SHIRAKAWA, T., TAKEDA, E., Bull. Tokyo Inst. Tech. No. 83 (1968) 23.
[14] YASUKAWA, S., JAERI Rep. JAERI-M 3724 (1969).
[15] MOTODA, H., Nucl. Sci. Engng 41 1 (1970) 1.
[16] UEDA, R., et al., Some Reports on Semi-Homogeneous Reactor Project, JAERI-Intern. Rep. (1962).
[17] NOZAWA, M., et al., JAERI Rep. JAERI-M 2244 (1966).
[18] YOSHIDA, H., HIRATA, M., Proc. BNES Fast Reactor Physics Conf. (1969) 257.
[19] NOZAWA, M., et al., JAERI Rep. JAERI-M 2654 (1967).
[20] KATURAGI, S., et al., JAERI Rep. JAERI-1195, 1199 (1970).
[21] KUROI, H., et al., Proc. BNES Fast Reactor Physics Conf. (1969) 82.
[22] NAGASHIMA, K., IZUMI, T., "Uranium requirements for the long-term nuclear power generating program in Japan", Economics of Nuclear Fuels (Proc. Symp. Gottwaldov, 1968), IAEA, Vienna (1968) 37.
[23] TAKEI, M., "Concepts of the econometric model and examination of its results", Economics of Nuclear Fuels (Proc. Symp. Gottwaldov, 1968), IAEA, Vienna (1968) 241.
[24] KIKUCHI, T., "Long-term nuclear fuel cycle and its economics relating to plutonium utilization", Nuclear Energy Costs and Economic Development (Proc. Symp. Istanbul, 1969), IAEA, Vienna (1970) 365.
[25] TAKEI, M., SUZUKI, S., WATANABE, H., "Evaluation of the growth of nuclear power in the future power system", Nuclear Energy Costs and Economic Development (Proc. Symp. Istanbul, 1969), IAEA, Vienna (1970) 501.
[26] Genshiryoku Shiryo No. 19, published by JAIF (1969).
[27] ROSENTHAL, M.W., et al., "Outlook for molten-salt power reactors", Nuclear Energy Costs and Economic Development (Proc. Symp. Istanbul, 1969), IAEA, Vienna (1970) 101.
[28] ROSENTHAL, M.W., et al., USAEC Rep. ORNL-4254 (1968).
[29] SHINOHARA, Y., YASUKAWA, S., SHIMAZAKI, J., J. nucl. Sci. Technol. 7 12 (1970) 15.
[30] YASUKAWA, S., FURUHASHI, A., JAERI Rep. JAERI-M 3946 (1970).

DISCUSSION ON AGENDA ITEM 2.7

Uranium-plutonium fuel cycle for thermal and fast reactors

DISCUSSION ON THE FOLLOWING GROUP OF PAPERS:

P/709	*USSR*	*Presented by V.V. Orlov*
P/185	*Italy*	*Presented by L. Sani*
P/496	*UK*	*Presented by J. Fell*

O. COMELLINI: In connection with the paper presented by Mr. Orlov (P/709) I should like to suggest that in view of the cost implications of having the plutonium immobilized in fast reactors, the doubling time is not a satisfactory criterion for assessing the economics of the various fast reactor types. I think that in trying to optimize the fast-reactor system one should take account of specific power as well as doubling time.

V.V. ORLOV: I agree entirely. The doubling time will become really important only later, when atomic power development depends solely on increasing the efficiency of the nuclear fuel. In considering fast reactors for the immediate future, we adopt criteria which take separate account of the excess production of plutonium and its specific loading in the fuel cycle, as can be seen from the written paper.

J. ARKUSZEWSKI: I gather from paper P/709 that the ROKBAR system includes various optimization possibilities pertaining to a given reactor performance. I should like to ask Mr. Orlov whether it also includes optimization of the cost of 1 kWh throughout the whole reactor lifetime?

V.V. ORLOV: With the ROKBAR optimization system it is possible to optimize fast reactors according to various criteria, including the cost of producing 1 kWh of electric energy or – when the costs vary with time – the overall cost (over the entire operating time of the reactor) per kW of installed capacity, referred to the 'present moment'.

J. BUSSAC: Several years ago it was realized that the value given to the ratio $\alpha = \sigma_c/\sigma_f$ was wrong, and the accepted value of α is now much higher. In view of this, does Mr. Orlov think it is really possible, with fast-neutron reactors (using sodium and oxide fuel), to obtain a plutonium doubling time of 11-12 years, as mentioned in the report, instead of 20-25 years?

V.V. ORLOV: In the data given in the paper account has been taken of the changes in uranium and plutonium cross-section resulting from measurements made in recent years (including changes in the value of α for ^{239}Pu). These changes have led to a slight deterioration in the characteristics of fast reactors, compared with the earlier published value. The 'exponential' doubling time of 11-12 years for a fast reactor with oxide fuel takes these changes into account. It should also be realized that the values indicated are based on a one-year fuel cycle and take account of the favourable influence of ^{240}Pu.

A. TEBOUL: I have a question on paper P/185, presented by Mr. Sani. By how much does the fabrication cost of a fuel element containing several per cent of plutonium exceed the cost of an element for a thermal reactor?

L. SANI: It depends on the price of the fissile plutonium employed. In our evaluations for a BWR we found that the increase in cost is about 40% for a fissile plutonium value of US $6.5/g and 60% for a plutonium value of $5/g. The correlation between the cost increase and the plutonium value is approximately linear. Of course, there are many other economic parameters that should be taken into account as well.

J. BUSSAC: I should like to ask Mr. Sani (P/185) what are the annual budgets for plutonium recycling in thermal neutron reactors at CNEN and ENEL.

L. SANI: Perhaps Mr. Schileo could answer for CNEN.

G. SCHILEO: The total cost borne by CNEN in the last six years for the development of plutonium-containing thermal fuel amounts to between US $8-9 million; this includes the cost of building the plutonium plant and acquiring completely original know-how regarding the design of nuclear fuel in general, and plutonium-containing fuel in particular (irradiation tests from capsule-scale to prototype full-scale; calculation codes for thermo-hydraulic, neutronic and mechanical design; out-of-pile tests, etc.).

L. SANI: Until 1970 ENEL had a research contract with EURATOM with a budget of about US $1.2 million. To this must be added the cost of the 16 plutonium prototype assemblies (about 3 tons of fuel), which was borne entirely by ENEL.

J. BUSSAC: Perhaps you could also say something about the penalty due to the higher percentage of ^{240}Pu?

L. SANI: We have studied the effect of different ^{240}Pu contents in detail because we have plutonium from different reactors. The results of these studies have shown no significant differences in the fissile plutonium value.

J. BUSSAC: Is it proposed to use the plutonium several times after irradiation in the reactor?

L. SANI: We have not thoroughly studied the second plutonium recycle because we think it is too early for plutonium utilization strategies in this field.

A. ARIEMMA: I should like to ask Mr. Fell a question about plutonium degradation during irradiation. Starting with fuel enriched by plutonium of Magnox origin and having a ^{240}Pu content of about 18%, one gets a plutonium content of about 30% after irradiation, which is similar to the case of plutonium of BWR origin. Does this mean that Magnox plutonium has a higher worth than BWR plutonium? Can the ^{240}Pu content be regarded as a 'fertile poison', which does not significantly affect reactivity lifetimes?

J. FELL: The results in Table II of paper P/496 are relevant to this question. The first of the four plutonium qualities for which data are given in this Table is essentially standard Magnox plutonium, and the third is very similar to reject plutonium from uranium-fuelled light-water reactors. The Table shows that Magnox plutonium has a higher worth per fissile atom for all the thermal reactors considered.

With reference to the role of ^{240}Pu as a fertile poison, I should mention that we have carefully studied this possibility, which would of course have attractive consequences. Our conclusion is that ^{240}Pu can operate in this way in the MK III GCR but not in light-water reactors. This point is also brought out in Table II, which shows a higher percentage of ^{240}Pu in the

reject plutonium for all the thermal reactors studied except the MK III GCR. We would of course be interested in studying any detailed calculations which lead to opposite conclusions.

DISCUSSION ON THE FOLLOWING GROUP OF PAPERS:

P/071	*USA*	*Presented by F.G. Dawson*
P/277	*Belgium*	*Presented by H. Bairiot*
P/346	*Austria*	*Presented by H. Gutmann*
P/351	*Yugoslavia*	
P/610	*France*	
P/822	*Japan*	

G. TESTA: My question is related to paper P/071, presented by Mr. Dawson. Figure 3 of the paper shows several gadolinium-uranium rods in the BWR fuel module. I should like to know whether the UO_2 mixed with Gd_2O_3 is enriched or natural UO_2. What criteria are applied in making the decision.

F.G. DAWSON: The UO_2 is natural uranium oxide. I am not aware of the criteria used by the General Electric Company in making the decision.

A. RADKOWSKI: I should like to ask Mr. Dawson (P/071) whether any studies have been made on the dependence of temperature coefficient and spatial stability on plutonium content?

F.G. DAWSON: Yes. Moderator temperature and void coefficients and fuel Doppler coefficients are substantially more negative for plutonium-fuelled systems than for uranium systems. The Doppler coefficient increases with ^{240}Pu content and could be 10-20% higher than for a uranium-fuelled reactor. The larger negative coefficients should improve reactor stability.

J. BUSSAC: I should like to make a few remarks regarding the paper presented by Mr. Dawson (P/071). The recycling of plutonium formed in uranium is a fundamental feature of the economics of water reactors and, by way of analogy, the utilization of plutonium that has already been recycled is an equally fundamental element of the economics of the nuclear power stations recycling plutonium. In this connection, two possible strategies can be considered: (1) the whole of the irradiated fuel and hence all the plutonium isotopes can be mixed for each cycle (this involves an increasing penalty) or, (2) the plutonium already irradiated can be separated and kept for fast reactors (an advantageous procedure for fast reactors because they then have a supply of cheaper fuel). This second possibility is being considered in France. Has either of these two strategies been studied in the United States of America? Do you know whether the presence of ^{241}Am constitutes a penalty in the fabrication of elements using plutonium that has been recycled several times?

F.G. DAWSON: Different compositions of plutonium would probably be mixed together for recycle if they were to be used in the same reload. Mixing, to obtain a single composition, would simplify accountability problems during fuel fabrication and handling as well as simplifying fuel sub-assembly design. These simplifications could very well offset the plutonium degradation penalty. Each case would have to be evaluated individually. The use of multiple-recycle plutonium for fast reactors is very likely for two reasons. First, its value in thermal reactors might be too low for economic use and, second, no storage would be required for fast reactors since three or four cycles represent a relatively long period of time and commercial fast reactors should be in operation when the plutonium is available after three or four cycles.

The build-up of ^{241}Am from the decay of ^{241}Pu represents a source of radiation which requires shielding. It increases with time after reprocessing of the irradiated fuel and with the ^{241}Pu content which in turn increases with repeated recycling of the plutonium. There is a fabrication penalty with this effect. Burnham and associates at Battelle (Pacific Northwest Laboratory) calculated a fabrication penalty of 1 to 2% for so-called 'high exposure' plutonium (about 20% ^{240}Pu), as compared with low exposure plutonium (6% ^{240}Pu). I believe the corresponding ^{241}Pu contents were of the order of 5 to 6% and less than 1%, respectively.

H.B. STEWART: Which characteristic of the plutonium fuel is most limiting, relative to the fraction of plutonium that can be used in the initial and equilibrium cores?

F.G. DAWSON: The high cross-sections of the fissile isotopes and their effects on power distributions and control worths.

AGENDA ITEM 2.8

Developments in the thorium fuel cycle

Progrès dans le cycle du combustible au thorium

Разработки в области ториевого топливного цикла

Progreso del ciclo del combustible de torio

Chairman

R. B. DUFFIELD, United States of America

Vice-Chairman

W. RUTKOWSKI, Poland

Scientific Secretaries

H. A. HUGHES, United Kingdom
K. N. KOSTADINOV, IAEA

LARGE-SCALE NUCLEAR ENERGY FROM THE THORIUM CYCLE

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Abstract–Résumé–Аннотация–Resumen

LARGE-SCALE NUCLEAR ENERGY FROM THE THORIUM CYCLE.

Nuclear reactors with capacities of several thousand megawatts offer great promise for water desalination, for agro-industrial complexes and for very large electrical power networks, but various delays have so far prevented the vision from becoming a practical reality. The attraction of large capacity is the prospect of very low fuelling, operating and maintenance costs – less than 1 m$/kWh (electric). Advances have been made since the 1964 Conference in recognizing the problems and identifying the means for their solution. An essential requirement is high availability; this demands simplicity of the plant and all auxiliaries, easy access to keep maintenance time short, and a long unattended life for all components. Energy from thorium is very promising, especially in conjunction with organic-cooled heavy-water-moderated reactors. A new design of thorium-oxide fuel promises ratings as high as 50 to 80 kW(th)/kg thorium, yielding 35 MWd/kg thorium over irradiations of two to four years with high neutron economy. Such a near-breeder fuel cycle requires a small ancillary supply of neutrons. Several cycles are discussed. We believe that a major advance in plant availability can be achieved by using organic-liquid coolant as discussed in another paper. Good neutron economy is preserved with cool heavy-water moderator. It is envisaged that large reactors with these features could be completed by about 1985. The capital cost would depend on the state of technology in the industry undertaking the work, but analysis of the component specifications indicates a substantially lower cost per kilowatt than for the CANDU reactors of today. When the initial provision of the reactor, fuel reprocessing plant and generating equipment is clearly separated from the subsequent low-cost operating and maintenance, such reactors are seen to be well suited to programs of capital aid.

LE CYCLE DU THORIUM PEUT DEVENIR UNE GRANDE SOURCE D'ENERGIE.

Les réacteurs nucléaires ayant une capacité de plusieurs milliers de mégawatts sont très prometteurs pour le dessalement de l'eau, pour les complexes agro-industriels et pour les grands réseaux électriques, mais divers retards ont empêché, jusqu'à présent, d'en tirer parti. L'intérêt des grandes capacités est qu'elles offrent la perspective de coûts très bas pour le combustible, l'exploitation et l'entretien – moins de 1 m$/kWh (électrique). Des progrès ont été effectués depuis la Conférence de 1964, en ce sens qu'on a délimité les problèmes et identifié les mesures à prendre pour les résoudre. Une exigence essentielle est un facteur élevé de disponibilité, ce qui requiert la simplicité de l'installation principale et de toutes les annexes, un accès facile pour réduire la durée des travaux d'entretien et une longue durée utile pour tous les composants. L'énergie tirée du thorium est très prometteuse, particulièrement pour les réacteurs ralentis à l'eau lourde et dont le caloporteur est un fluide organique. Un nouveau type de combustible à l'oxyde de thorium permet d'espérer de hauts rendements allant de 50 à 80 kW(th)/kg Th et donnant 35 MWj/kg Th, avec une bonne économie neutronique, pour les irradiations de deux à quatre ans. Ce cycle de combustible qui se rapproche de celui des surgénérateurs nécessite un petit approvisionnement auxiliaire de neutrons. Plusieurs cycles sont passés en revue par les auteurs. Il semble qu'un progrès considérable peut être réalisé pour ce qui est de la disponibilité en utilisant un caloporteur constitué par un fluide organique comme il en est question dans un autre mémoire. Une bonne économie neutronique est assurée avec de l'eau lourde froide comme modérateur. De grands réacteurs ayant ces caractéristiques pourraient être réalisés vers 1985. Les dépenses d'investissement dépendraient de l'état de la technologie, mais l'analyse des spécifications des composants donne un coût par kilowatt nettement plus bas que celui des réacteurs CANDU d'aujourd'hui. Lorsque les frais d'investissement initial pour le réacteur, l'usine de retraitement et la génératrice sont nettement séparés des frais (relativement bas) d'exploitation et d'entretien, on voit que ces réacteurs conviennent pour les programmes d'assistance financière.

ИСПОЛЬЗОВАНИЕ В БОЛЬШОМ МАСШТАБЕ ТОРИЕВОГО ЦИКЛА В ЯДЕРНОЙ ЭНЕРГЕТИКЕ.

Ядерные реакторы мощностью в несколько тысяч мегаватт имеют большие перспективы использования для опреснения соленых вод, для работы в агропромышленных комплексах и в исключительно крупных электроэнергетических системах, но различные обстоятельства до сих пор не дали возможности этой мечте превратиться в практическую реальность. Привлекательная сторона атомных электростанций с большой мощностью состоит в перспективе достижения очень низких затрат на топливо, эксплуатацию и ремонтно-техническое обслуживание – менее 0,001 долл/квт-ч (эл). Период, истекший со времени Конференции 1964 года, характеризуется некоторыми успехами в оценке проблем и в определении путей их решения. Важным требованием к атомным электростанциям является высокий коэффициент использования календарного времени; это требует простоты конструкции как реакторной установки, так и всех вспомогательных систем, легкого доступа к оборудованию, для того чтобы максимально сократить время, необходимое на проведение технического обслуживания, и увеличить срок работы всех узлов без какого-либо обслуживания. Использование тория для производства энергии имеет большие перспективы, особенно в тяжеловодных реакторах с органическим теплоносителем. Новая конструкция топлива из окиси тория обещает дать от 50 до 80 кВт(тепл) на 1 кг тория, обеспечивая глубину выгорания 35 МВт·сутки/кг тория в течение от двух до четырех лет облучения с высокой экономией нейтронов. Такой топливный цикл, близкий к циклу реактора-размножителя, требует небольшого вспомогательного источника нейтронов. Рассматриваются несколько вариантов циклов. По нашему мнению, большие достижения в повышении коэффициента использования календарного времени могут быть получены путем использования жидкого органического теплоносителя; эта проблема рассматривается в другом докладе. Хорошая экономия нейтронов сохраняется и при использовании тяжеловодного замедлителя, имеющего умеренную температуру. Предполагается, что разработки крупных реакторов с такими характеристиками могут быть закончены примерно к 1985 году. Размер капитальных затрат будет зависеть от уровня развития технологии в данной отрасли промышленности, выполняющей эти работы, но анализ спецификаций узлов указывает на значительно меньшие затраты, приходящиеся на 1 кВт, чем на современных реакторах типа CANDU. Если первоначальные затраты на поставку реактора, установки для переработки топлива и теплоэнергетического оборудования четко отделить от последующих низких затрат на эксплуатацию и ремонтно-техническое обслуживание, то можно увидеть, что такие реакторы хорошо подходят для программы оказания помощи в форме капитальных вложений.

UTILIZACION DEL CICLO DEL TORIO PARA OBTENER ENERGIA NUCLEAR EN GRAN ESCALA.

Parece posible construir reactores nucleares de varios miles de megavatios; estos reactores representan una gran promesa para la desalación del agua, para complejos agro-industriales y para redes muy grandes de energía eléctrica, pero varios retrasos han impedido por ahora que esta visión se convierta en una realidad práctica. Lo que más atrae de una gran instalación es la esperanza de obtener costes muy bajos, menores que una milésima de dólar por kilovatio-hora (eléctrico), sumando los costes del combustible, los de explotación y los de mantenimiento. Desde la conferencia de 1964 se ha progresado en la identificación de los problemas y de los medios de resoverlos. Se requiere esencialmente una gran disponibilidad, lo que exige simplicidad de la instalación y de todos los medios auxiliares, con acceso fácil para que el entretenimiento consuma poco tiempo, y que todos los componentes resistan una larga vida sin necesidad de atención. Promete mucho la energía obtenida del torio, especialmente utilizando reactores moderados por agua pesada y refrigerados por líquido orgánico. Un nuevo diseño de combustible de óxido de torio promete llegar a potencias específicas de hasta 50 a 80 kW(t)/kg de torio, con grados de quemado de 35 MWd/kg de torio e irradiaciones de 2 a 4 años con buena economía neutrónica. Este ciclo de combustible cuasi reproductor impone la servidumbre de une pequeña fuente de neutrones auxiliar. Se comentan distintos ciclos. Creemos que se puede conseguir una mejora importante en la disponibilidad de las instalaciones utilizando refrigerantes líquidos orgánicos, tal como se discute en otra memoria. Se logra una buena economía neutrónica empleando como moderador agua pesada fría. Se prevé que para 1985 más o menos, podrían haberse construido grandes reactores de estas características. El coste de instalación dependería del estado de la tecnología en la industria que llevara a cabo el trabajo, pero el análisis de las especificaciones de los componentes indican que el coste por kilovatio sería sensiblemente más bajo que el de los reactores CANDU de hoy día. Si se ponen aparte los gastos iniciales de la construcción del reactor, de la instalación de reelaboración del combustible y del equipo de producción de energía, y se considera que luego los gastos de explotación y mantenimiento son bajos, se ve que tales reactores se prestan bien para programas de ayuda financiera.

HIGH POWER HEAVY-WATER-MODERATED REACTORS

At the 1964 Geneva Conference, Canada discussed conceptual designs of prospective heavy-water-moderated power reactors with alternative heat transport fluids [1]. All have this fluid confined in channels flowing over multi-element fuel bundles. These channels lie within tubes that form a lattice in the cool heavy-water moderator.

Since the Conference, 500-MW(e) natural-uranium-fuelled reactors have been constructed [2] in a multiple-unit generating station, and the detailed design of 750-MW(e) units has advanced. These units all use heavy water for heat transport. A 250-MW(e) prototype reactor with boiling light water that needs no steam-raising heat exchanger is also operating. A further design, using an organic liquid (HB-40 a hydrogenated terphenyl) for heat transport at high temperature, is giving excellent service in a 40-MW(th) experimental reactor and in test loops in other experimental reactors.

These basic designs can be extended to even higher capacities very readily if the fuel-power density is increased [3]. To achieve this while preserving neutron economy, thorium fuel is particularly suitable. The larger capacity leads to a lower unit cost of power.

Such prospects were further discussed at the 1967 Symposium on Heavy Water Power Reactors (International Atomic Energy Agency) [4,7,11].

Operating experience has emphasized the economic value of continuous availability of all components [5,6]. To attain this, the total number of components and actions should be limited, in particular the total number of coolant channels and fuelling operations even when carried out at power. The combination of thorium fuel and organic liquid caloporteur [7,8] offers advantages in high channel power, few fuel changes and ease of maintenance such as the location and cure of leaks that develop in joints and valves.

Large-unit and large-station capacity offer lower prospective power costs. The chief advantage lies in the reduction of the capital-cost component, and there is also a prospective saving in the number of operators and the cost of chemical control. Fuelling costs may be reduced by the correct choice of fuel cycle, for an optimum combination of high burn-up, low fuel inventory value and low fuel fabrication and reprocessing costs. Overall neutron economy is an essential key as is high efficiency in thermal—electrical energy-conversion. In the prospect presented in 1964, the lowest unit-fuelling costs were associated with the organic liquid coolant and, although the relative parameters have changed as experience has been gained, this conclusion is unaltered. The characteristics that give the organic-cooled reactor its prospective advantages now are

- high surface-heat transfer-rate from fuel (up to 300 W/cm^2),
- high station efficiency from high coolant temperature (up to 40%), and
- ease of maintenance and access due to negligible radioactivity of coolant and self-disclosure of any leaks.

At the 1964 Conference there were discussions of the prospective use of low-cost large-scale nuclear energy for desalting water [9] and subsequently the scope of such prospects has been widened to cover nuclear-powered complexes for agro-industrial development [10]. Up to the present time, high capital costs have appeared as the main disadvantage of nuclear power. Nevertheless, for a long time it has been realized in principle that, when the capacity is high, availability and a low fuelling cost assume great importance. This applies particularly to the agro-industrial complex where applications can become much wider with lower operating costs.

There is an added reason for considering the capital and operating costs separately. The total investment in the complete industrial plant involved is much larger than that of the power reactor. It is indeed so large that the capital equipment will mostly be fabricated elsewhere and financed from other than the local source. Thus, the reactor becomes a candidate for capital aid, provided all operating is simple and of low enough cost to be provided locally.

THORIUM FUEL CYCLES

A peculiar property of thorium fuel was emphasized in a paper [11] to the 1967 Symposium mentioned above. Plain thorium oxide fuel might be fabricated for a total cost of about $30/kgTh, but after irradiation over two to four years yielding 35 MWd/kgTh (worth, for example, $84 at 0.1 m$/kWh thermal energy), the credit from the residual value of the fuel would be about $200/kgTh. The compensating disadvantage was the need to supply neutrons from some costly enriched-fissile material or from a neutron source of some other type. A subsequent report [12] embodied, as the "valubreeder" concept, the principle of using thorium fuel, for value and energy yield, and enriched uranium, as the lowest cost source of spare neutrons. It also discussed the relative prospective costs of several complete fuel cycles at equilibrium that ranged from 0.3 to 0.55 m$/kWh (1967 U.S. dollar values). Although this range covers a factor of nearly two, it is not to be concluded that the lowest cost would be optimum. Reference has been made to the importance of availability and, if electrical power is assigned a total value of even only 2 m$/kWh at 90% availability, then changing the availability to 80% could be represented as a penalty of 0.25 m$/kWh whereas the whole range of fuel costs mentioned was only 0.25 m$/kWh. Furthermore the higher the value set on power the higher the penalty for nonavailability. In some systems where power must be purchased from some alternative source when a reactor is not available, very much higher penalties can be assigned to nonavailability. In others where, for example, there is stored water capacity available the penalty may be less. If the absence of power leads to idle factories, penalties may be much higher than the cost of idling the power plant alone.

A number of thorium—uranium—plutonium fuel cycles have been discussed in earlier reports [13—16].

In this paper some further thorium fuel cycles are discussed in relation to one specific design of fuel channel for a large heavy-water-moderated reactor. The dimensions adopted for the reference fuel bundle and channel are given later in this paper. The channels are arranged in a square lattice and most calculations have been made for a lattice pitch of 280 mm, but some have been made for 360 mm. The channels are the largest in diameter so far studied extensively, and the neutron-flux depression in the fuel is even higher than in the steam-generating heavy-water (SGHW) reactor [17], the only operating power reactor with a comparable heavy-water lattice.

Because these large-diameter heavily fuelled channels are so desirable for large power reactors, it has seemed necessary, despite the complexity, to attempt a fresh analysis.

THORIUM FUEL DESIGN AND PERFORMANCE

Basis for Choice of Sintered ThO_2

In most nuclear-powered generating stations throughout the world the fuel is formed of sintered pellets of UO_2. The irradiation behaviour of UO_2 has been thoroughly investigated and reported at many conferences. Less work has been done on thorium fuels, but results that are available [18—22] show that sintered pellets of ThO_2 perform as well as, or better than UO_2 irradiated at the same specific power. For example, irradiations of ThO_2—1.3wt%U^eO_2 [1] fuelled assemblies, designed to produce high fast-neutron fluxes, are continuing in the NRU reactor at Chalk River Nuclear Laboratories (CRNL) [23]. The two types of assemblies being irradiated have the following characteristics and performance.

	Fast-Neutron Rods	Flux-Peaking Rods
Design	3 15-element bundles in one channel	1 19-element full-length assembly
Length (mm)	Each bundle, 500; total of 1500	2800
Element dimensions (mm)	15.2 outer diam. 0.38 wall thickness	12.5 outer diam. 0.38 wall thickness
No. irradiated	14	13
Max. linear heat output per element (W/cm)	900	380
Max. Burn-up (MWd/kg(Th+U))	25	50
Max. time in reactor (d)	800	1600

[1] U^e = U-235, and U^n = natural uranium.

The few defects that occurred are attributed to the unintentional inclusion of a hydrogenous contaminant inside the element, which resulted in the formation of radially oriented zirconium hydride platelets in the sheath. Stress on the low ductility Zircaloy resulted in very small defects.

In another assembly the fuel was ThO_2—1wt%U^nO_2. The power output from this assembly gradually increased as U-233 was formed, and the assembly now has a peak burn-up of over 20 MWd/kg(Th + U) at a linear power output of 300 W/cm.

In the ThO_2 assemblies examined, the fission-gas release and the thermal expansion were low. In another test [18] it was shown that the dimensional changes, the plasticity and the in-reactor resintering of ThO_2 and UO_2 were similar when compared at the same percentage of their absolute melting temperatures. With this knowledge, and with confirmation from the ThO_2 irradiations at CRNL and other laboratories, we feel confident that we can predict the irradiation behaviour of ThO_2 and can design fuel elements to operate at high power outputs to high burn-up.

Reference Fuel Bundle and Channel Design

The reference fuel bundle and channel have the following specifications:

Bundle diameter	129.9 mm
Bundle length	0.5 m
Number of elements	61
Diameter of elements	13.88 mm
Zirconium-alloy sheath thickness	0.38 mm
Channel inside diameter	133.36 mm
Channel wall thickness	3.18 mm
Channel to calandria radial gap	8.5 mm
Calandria-tube wall thickness	1.95 mm
Calandria-tube outer diameter	160.62 mm.

Fuel Element Design

The design (Fig. 1) of the individual fuel elements is discussed in detail in reference [19] and the bibliography therein. Four basic concepts are employed:

- thin sheathing, to reduce the neutron absorption, and keep the temperature of the interface with the fuel low;
- hollow pellets, to make sheath strain negligible;
- internal graphite discs, to provide better heat transfer from the centre of the fuel [24] and also to provide for absorption of released fission-gases; and
- thin graphite interlayer between the pellets and the sheath, to minimize local stresses and possible ratchetting.

Calculations well supported by experiment indicate that this element can operate at linear power outputs up to 1000 W/cm with a surface heat flux of 230 W/cm^2. There should be negligible strain of the sheath due to

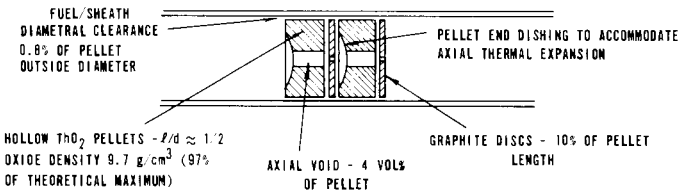

FIG. 1. Fuel pellet assembly in sheath.

thermal expansion of the fuel, or due to pressure from released fission-product gases up to a burn-up of at least 35 MWd/kgTh. The graphite discs limit the maximum temperature in the fuel so that

$$\int_{\text{Surface}}^{\text{Maximum}} \lambda d\theta \qquad \text{is only 45 W/cm.}$$

In a practical reactor design such fuel elements incorporated in the reference 61-element bundles can give a maximum of 3.5—4.5 MW thermal power per metre of in-core length. Assuming an overall average—maximum power ratio for the core of 0.65 and a channel thermal—electrical efficiency of 40%, a 1500-MW(e) reactor would need 290 channels 5 m long.

NEAR-BREEDER CHARACTERISTICS OF CANDU REACTORS

It was reported to the 1964 Geneva Conference [25] that the near-breeder characteristics of Canadian Deuterium Uranium (CANDU) reactors operating on a thorium fuel cycle, initiated and maintained by a supply of separated uranium-235, was such that low cost power could be supplied to meet all the world's needs for hundreds of centuries. The energy yield, mostly from fission of uranium-233, could be expressed as 60—100 thermal MWd per kilogram of natural uranium supplied [15]. Subsequently [12] the fuelling costs for such cycles were indicated as in the range 0.3—0.55 m$/kWh(e) in terms of 1967 U.S. dollar values including inventory charges at 7.2% per year and natural uranium costing $8/lb$U_3O_8$ or $21/kgU. Doubling that cost of natural uranium would increase the fuel supply cost by only 21/(24x60x0.375) $\simeq$ 0.04 m$/kWh(e).

All these earlier analyses were made for well-moderated reactors (with Westcott epithermal r = 0.07) [15], and although the general conclusions are valid for any heavy-water-moderated reactors of good neutron economy there are significant changes in the details of the fuel cycles when under-moderated lattices with heavily fuelled channels of large diameter are studied.

BASIS OF POWER AND BURN-UP EVALUATIONS

The debit and credit neutron-balance is the essential basis for the evaluation of different fuel cycles. The precise evaluation in a completely specified reactor involves a detailed evaluation of the neutron spectrum and

flux and the consequent reaction rates to a degree that strains to the limit the capacity even of large digital computers and the limits of knowledge of nuclear data. For this reason a number of different approximations are made, and, fortunately, the risk of significant error in fuel-cycle costs arising from the irradiation is not large, and indeed is very much smaller than the uncertainties of processing costs outside the reactor. It is important to understand this because the fuel-cycle costs predicted are as low as or even lower than the fuelling costs of natural-uranium CANDU reactors which are the lowest yet established in the world, being in the range 0.4—0.8 m$/kWh depending on the credit allowed for plutonium produced. Moreover the need for high availability leads to the selection of high average fuel burn-up between processing stages, which can be achieved with thorium fuels.

To approximate to the conditions in an undermoderated reactor with large fuel channels the LATREP code has been used. The lattice cell code LATREP [26] is a multi-group, collision probability, fuel burn-up code using 32 epithermal and one thermal neutron energy groups. The thermal group, up to 0.625 eV, uses a modified Westcott formalism. Cluster geometry effects are allowed for in the calculation of effective U-238 resonance integrals, in the self-shielding of the major Pu-240 resonance and in the thermal-flux calculation. Burn-up proceeds independently in each ring of fuel and the neutron spectrum is recalculated at fixed irradiation intervals. The output contains summary tables, which are useful for fuel-cycle calculations, of reaction rates as a function of irradiation.

Some of the differences from the previous BOUT code [27] are evident from Fig. 2. The range over which the LATREP code can be relied on is also restricted and this has to be kept in mind in the method of its use.

It was apparent from the cycles considered previously [15] that the highest energy yield from a given supply of uranium would involve reprocessing fuel after a relatively short irradiation such as 2.5 n/kb reached in 400 full-power days at 7×10^{13} n/cm^2.s. Operating at a lower neutron flux increases the yield further and extends the period but increases the inventory cost.

In order to keep fuel movement to a minimum while achieving a low fuelling cost, the principle is adopted of using two kinds of fuel bundles, which may be described as driver fuel and power fuel. At all times a reactivity balance, a limited power range and a materials balance must be satisfied. Examples of the compromise achieved by the use of separate driver and power fuel may be seen from a few cases in Table I.

For eventual U-233 recycle, the sequence number 4 (Table I) appears a good compromise achieving the high average burn-up of 33.3 MWd/kgHE [2] with the power fuel giving no more than 39.6 MWd/kg at an irradiation of 4 n/kb in 2.8 years[3]. The loss of U-233 is 0.093 g/MWd and the fuelling cost 0.52 m$/kWh(e) on the assumptions adopted. This choice is clearly weighted to availability and low fuelling cost rather than to the highest conservation of fissile material.

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[2] HE = heavy elements.

[3] See data in the notes to Table I.

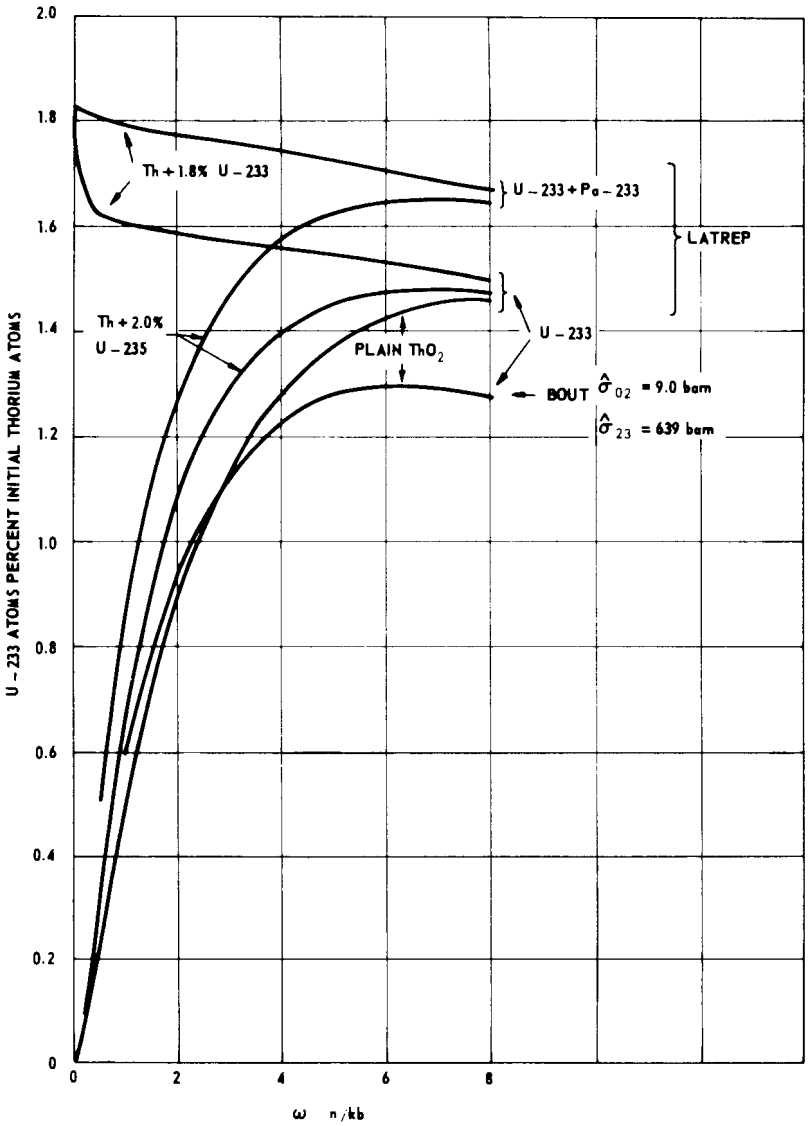

FIG. 2. Comparison of LATREP and BOUT calculations for ^{233}U and ^{233}Pa content at 5×10^{13} n/cm$^2 \cdot$ s.

TABLE I. CHARACTERISTICS OF SELECTED 'DRIVER' AND 'POWER' FUEL COMBINATIONS

Sequence No.	FUEL SPECIFICATIONS: Blended Oxide Fuel On-Power Fuelling 280-mm Square Lattice — Driver B: %	Driver B: Fiss.[a]	Driver B: ω_B n/kb	Power C: % U-233	Power C: ω_C n/kb	FACTORS AFFECTING OPERATIONAL "AVAILABILITY": Ratio in Reactor N_C/N_B	Feed Ratio $\bar{\eta} = 1.05 \left(\frac{\omega_C/\omega_B}{N_C/N_B}\right)$	Burn-up MWd/kgHE[a]: Driver B	Power C	Av.	FISSILE FUEL CONSERVATION: g Fissile Fuel lost per MWd: U-235	U-233	FUEL COST: Equilibrium Specific Inventory: Fiss. Fuel $/kW(e)	Fabn.[a] $/kW(e)	Unit Energy Cost (Fuel) m$/kWh(e)
1	2.0	U-233	1.75	1.8	3.0	1.38	1.24	19.31	29.79	23.98	–	0.060	14.54	2.97	0.57
2	2.0	U-233	1.75	1.8	4.0	0.78	2.94	19.31	39.57	24.46	–	0.067	14.57	2.93	0.58
3	2.3	U-233	1.75	1.8	4.0	1.88	1.21	21.29	39.56	29.54	–	0.084	14.83	2.89	0.54
4	2.3	U-233	2.5	1.8	4.0	1.01	1.58	29.34	39.56	33.30	–	0.093	15.21	2.86	0.52
5	2.3	U-233	3.0	1.8	3.0	0.98	1.02	34.57	29.79	32.21	–	0.088	15.34	2.87	0.52
6	2.3	U-233	3.0	1.8	4.0	0.55	2.42	34.57	39.56	36.03	–	0.101	15.56	2.82	0.51
7	2.3	U-233	3.0	1.8	6.0	0.33	6.06	34.57	59.11	38.05	–	0.112	15.72	2.79	0.52
8	1.5	U-233 + U-235	2.5	1.8	3.0	2.89	0.53	33.17	29.79	30.95	0.122	−0.009	15.36	2.82	0.54
9	1.5	U-233 + U-235	2.5	1.8	4.0	1.29	1.24	33.17	39.56	36.03	0.168	−0.026	15.70	2.72	0.54
10	1.5	U-233 + U-235	2.5	1.8	6.0	0.77	3.10	33.17	59.11	39.49	0.209	−0.040	15.99	2.62	0.55
11	1.5	U-233 + U-235	2.0	1.8	3.0	3.45	0.44	27.39	29.79	29.06	0.105	−0.004	15.03	2.86	0.55
12	1.5	U-233 + U-235	2.0	1.8	4.0	1.95	1.03	27.39	39.56	33.40	0.153	−0.022	15.28	2.76	0.54
13	1.5	U-233 + U-235	2.0	1.8	6.0	1.16	2.58	27.39	59.11	36.26	0.200	−0.039	15.51	2.65	0.56
14	3.0	U-235	1.5	1.8	3.0	1.28	1.56	19.63	29.79	23.60	0.464	−0.294	15.06	2.73	0.64
15	3.0	U-235	1.5	1.8	4.0	0.72	3.68	19.63	39.56	23.89	0.591	−0.385	15.22	2.63	0.67
16	3.0	U-235	1.5	1.8	6.0	0.43	9.24	19.63	59.11	23.49	0.690	−0.454	15.34	2.55	0.69
17	3.0	U-235	1.75	1.8	3.0	0.85	2.03	22.57	29.79	24.96	0.521	−0.333	15.30	2.68	0.64
18	3.0	U-235	1.75	1.8	4.0	0.48	4.78	22.57	39.56	25.51	0.630	−0.410	15.48	2.59	0.65
19	3.0	U-235	1.75	1.8	6.0	0.29	11.99	22.57	59.11	25.38	0.706	−0.463	15.59	2.53	0.67
20	2.3	U-233	3.0	1.6	3.0	0.45	2.22	34.57	27.98	35.52	–	0.092	15.41	2.85	0.53
21	2.3	U-233	3.0	1.6	4.0	0.36	3.74	34.57	37.66	35.22	–	0.100	15.48	2.81	0.52
22	2.3	U-233	3.0	1.6	6.0	0.26	7.61	34.57	57.25	37.20	–	0.109	15.56	2.78	0.52
	Breeder Fuel										Net Gain				
23	1.8	U-233	1.0	1.6	1.0	2.50	0.40	10.22	9.19	9.49	–	0.010	14.02	3.25	1.02

[a] Abbreviations: HE = heavy elements; Fiss. = fissile; Fabn. = fabrication.

Data and Assumptions Useful for Table I.

1. $\bar{\eta}$ may be considered as characteristic of the fuel or of the reactor, since the reactivity balance requires these to be equal [12,15].

2. ω is the irradiation measured in neutrons per kilobarn (Westcott flux x time x 10^{21} n/cm^2). Reaction rates were normalized in the LATREP program to a thermal neutron flux of 5 x 10^{13} n/cm^2.s. Reactor utilization is 90.5% so that the residence time of fuel = 0.70 ω yr.

3. The fuel fabrication cost assumed is uniformly $50/kgHE including the supply of thoria. The more highly enriched fuels, however, generate more power (and cost more); hence the specific inventory values are not constant. Average core power rating is about 43.5 kW(th)/kgHE.

4. N_B, N_C are the numbers of fuel bundles of B and C fuel in the reactor.

5. In estimating fuel conservation the convention introduced in reference [15] is maintained: 98% of Pa-233 is assumed to decay to U-233 before extraction. A recovery of 99% of the uranium so determined is assumed.

6. The value assigned to fissile materials is $11/gU-235 and $13/gU-233; all other costs are considered included in fabrication ($50/kgHE) and extraction ($20/kgHE). Inventory, credit and interest charges are as explained in the valubreeder study [12]. Charge on inventory is at 7.2% per year and interest rate on credit is 5.5% per year. A delay time of 0.75 yr between fuel discharge and processing is included.

7. The rate at which fuel has to be supplied may be found directly from the average hurn-up in MWd(th)/kgHE (i.e., U + Th). Relative rates of feed are given for equilibrium conditions. The net station efficiency for electrical output/fission power is 0.375. (Note: 0.375 x 24 = 9.0 MWh(e)/MWd(th).

It is of interest to note that by shortening the irradiations and using the lowest practicable fissile content, a fuel cycle that gives a small breeding gain is indicated (Table I, sequence number 23).

In the absence of a supply of uranium-233 it is practicable to start a reactor with 2% U-235 oxide blended with thoria and replenish with 3% U-235 as driver fuel. After reprocessing, the driver fuel can be 1.5% U-233 + 1.5% U-235 or a similar mixture that results from the first irradiation. In all the cases in the table the power fuel is 1.8% or 1.6% U-233. In the absence of pure U-233 these could be substituted by 2.0% U-235 or 1% U-233 + 1% U-235, which behave similarly.

A number of other combinations have been evaluated and will be presented in a more detailed discussion [28].

SEPARATE ENRICHED FUEL (SPIKE FUEL)

The basic principle for achieving fuel conservation is to remove fission products from continued irradiation. One method that has some practical advantages is to make the driver fuel without thorium. For example, one fuel that has operated satisfactorily consists of highly enriched UO_2 or PuO_2 particles coated with pyrolitic graphite and dispersed in a graphite matrix. These elements are clad in zirconium alloy and assembled in bundles of dimensions similar to the ThO_2 fuels. The advantages lie in the possibility of reducing the total cost of fabrication and processing. Estimates of these costs depend on the timing of the processes. Uranium extracted from thoria is best fabricated before the decay products of U-232 build up. Thorium would not be used for recycle for about 15 years to allow time for the decay of Th-228 (RdTh) and its more radioactive decay products. The use of separate enriched fuel makes the eventual elimination of U-236 relatively simple. A disadvantage of using separate enriched fuel is that the neutron wastage by absorption in the reactor components is increased.

OFF-POWER FUELLING

Another important variant of the fuel cycle arises if the reactor is refuelled when off-power. Thorium fuel has some advantage over uranium fuel in that the average rate of loss of reactivity with burn-up can be significantly lower. Some cycles have been evaluated that suggest, however, that refuelling of about one tenth of the reactor at intervals of about 60—80 days would be needed to limit the range of reactivity swing to an acceptable level. It is possible, however, to improve on this if self-shielding in highly enriched separate fuel bundles is exploited [13].

IMPROVEMENT OF NEUTRON ECONOMY IN THE REACTOR

In earlier studies [12,15,16] results were presented in terms of the neutron wastage in the reactor expressed by the approximate parameter $\bar{\eta}$ (the ratio of the total neutron absorption and leakage rate in the reactor to the absorption rate in the fuel isotopes and their reaction products).

For the fuel discussed above and some typical irradiations $\overline{\eta} \simeq 1.051$ to 1.055, neglecting neutron leakage which is small from a large reactor — 1—1.5%. After the fission products, the neutron wastage in the zirconium and organic liquid coolant is highest (Table II). It might be possible to effect some reduction of this wastage: if, e.g., the zirconium absorption could be halved $\overline{\eta} \simeq 1.04$. The effect of changes of $\overline{\eta}$ on the three important characteristics of fuel conservation, average burn-up (a factor affecting availability) and fuel cycle cost are illustrated in Fig. 3, which may be compared with corresponding figures in [12,15,16]. The numbers shown against the curves in Fig. 3 identify the cases by the sequence number in Table I; for convenience certain key characteristics are also indicated. For Fig. 3a each curve requires two cases one of which introduces U-235; the other must consume U-233. The results are calculated very simply from the fuel conservation data in Table I, assuming that 1 kg natural uranium supplies 5 g of separated U-235.

BREAK-EVEN VALUE OF URANIUM-233 AND THE VALUBREEDER

It will be seen from Table I or Fig. 3b that the cycles involving U-235 give a higher fuelling cost than those involving only U-233. The difference would be removed if U-233 were given a higher value relative to U-235. For $\overline{\eta} = 1.05$ for cases 3, 4, 6, 9, 12, 15 and 18 it is found that a value of U-233 of \$15.34/g instead of \$13/g relative to U-235 at \$11/g would bring all the fuelling costs close to 0.57 m\$/kWh.

It will be noted that with the exception of the breeder case the fuelling costs do not cover a very wide range. It follows that the fuels and irradiations may well be chosen on the basis of engineering and operating convenience.

No valubreeder cycle is included in Table I because it was not easy to evaluate by the LATREP code in its present form. The difficulty arises from the changing power output and neutron spectrum during the irradiation of initially plain thorium fuel. This does not mean that valubreeder cycles are inappropriate for the fuel, channel and lattice dimensions

TABLE II. INTEGRATED NEUTRON ABSORPTIONS IN REACTOR COMPONENTS(%)

Initial fuel U–235(%)	3.0	1.5	–	–	–
Initial fuel U–233(%)	–	1.5	2.3	1.8	1.8
Irradiation (n/kb)	3.0	3.0	3.0	3.0	4.0
Thorium	37.80	37.59	40.12	42.58	41.84
Uranium & protactinium	48.31	48.74	47.08	45.13	44.92
Fission products (including U–236)	9.16	9.06	8.01	7.23	8.20
Organic caloporteur	1.52	1.46	1.58	1.74	1.73
Oxygen, H_2O, D_2O, and C	1.10	1.08	1.10	1.12	1.12
Zirconium	2.11	2.07	2.12	2.21	2.21

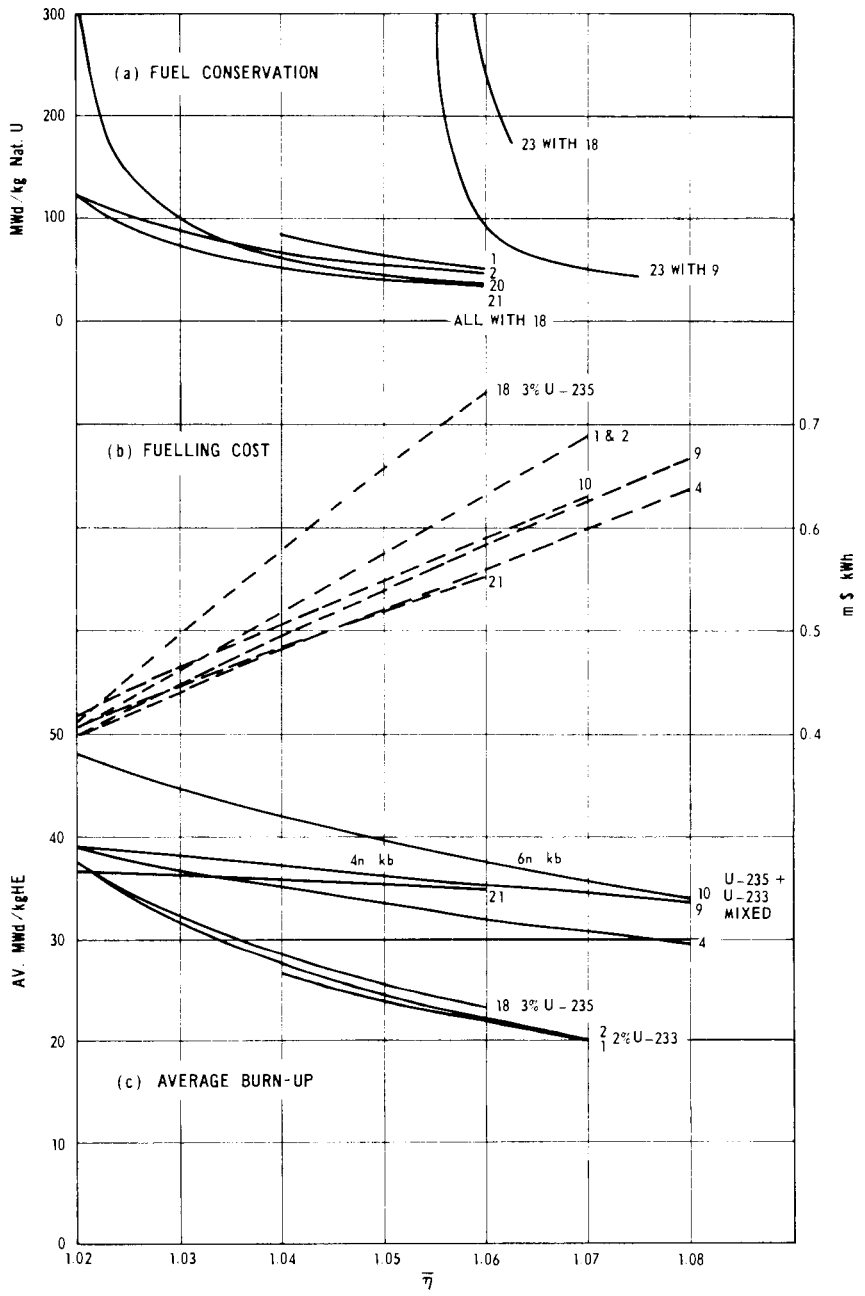

FIG. 3. Effect of neutron wastage on fuel conservation, fuelling cost and average burn-up for different fuel combinations. The numbers against each curve are the sequence numbers of the fuels in Table I.

specified. General considerations suggest that since the U-233 break-even value established above is so high, it would be more economical to operate some heavy-water CANDU reactors on the valubreeder cycle and supply U-233 to other CANDU reactors for operation on the cycles discussed above.

The lattice pitch of 280 mm is too small for satisfactory operation of the reactor on natural UO_2 fuel. A pitch of 360 mm would allow a burn-up of 6.5 MWd/kgU^n on continuous refuelling, but the optimum power rating would be lower so that it would be preferable to redesign the fuel for a smaller number of elements and reduce the organic coolant somewhat.

Optimizing the reactor for a valubreeder cycle would lead to dimensions intermediate between those most suitable for natural-uranium fuel and for those most suited for U-233 and thorium recycle.

CONCLUSION

The studies outlined in this paper indicate that a thorium-fuelled organic-cooled reactor with heavy-water moderator offers great potential for large-scale nuclear energy. Studies are continuing and will be reported in more detail in forthcoming publications now in preparation.

ACKNOWLEDGEMENTS

The authors wish to acknowledge the contributions made by J. Griffiths concerning LATREP and F.P. Blackstein in the many calculations involved in evaluating the results for presentation.

REFERENCES

[1] PON, G.A., et al., *Prospective D_2O-moderated power reactors,* Proc. 3d Int. Conf. Peaceful Uses Atomic Energy, 5, United Nations (1964) 333.

[2] WILSON, I.L., et al., *Studies of CANDU-type reactors in the 500 MW(e) range,* Proc. 3d Int. Conf. Peaceful Uses Atomic Energy, 5, United Nations (1964) 291.

[3] HART, R.G., et al., *The CANDU nuclear power system: competitive for the foreseeable future,* paper 151 at this Conference.

[4] MORISON, W.G., *Multi-unit aspects of the Pickering generating station,* Proc. IAEA Symp. Heavy-water Power Reactors, Vienna (1967) 267.

[5] LEWIS, W.B., FOSTER, J.S., *Canadian operating experience with heavy-water power reactors,* Proc. American Power Conference 32 (1970) 200; also in expanded form as Publication AECL-3569, Atomic Energy of Canada Limited (1964).

[6] BLACKSTEIN, F.P., Ed., *Proc. AECL Symposium on Reliability,* publication AECL-3670, Atomic Energy of Canada Limited (1970).

[7] MOORADIAN, A.J., et al., *Present status of Canadian organic-cooled reactor technology,* Proc. IAEA Symp. Heavy-water Power Reactors, Vienna (1967) 383.

[8] TEGART, D.R., *Operation of the WR-1 organic-cooled research reactor,* paper AED-Conf-69-357-031 presented at ANS Conf. Reactor Operating Experience, San Juan, Puerto Rico (1969); publication AECL-3523, Atomic Energy of Canada Limited (1970).

[9] RAMEY, J.T., et al., *Nuclear reactors applied to water desalting,* Proc. 3d Int. Conf. Peaceful Uses Atomic Energy, 6, United Nations (1964) 428.
[10] *Nuclear energy centers industrial and agro-industrial complexes,* USAEC Rep. ORNL-4290 (1968).
[11] LEWIS, W.B., *Outlook for heavy-water reactors,* Proc. IAEA Symp. Heavy-water Power Reactors, Vienna (1967) 545.
[12] LEWIS, W.B., *The super-converter or valubreeder; a near-breeder uranium-thorium nuclear fuel cycle,* publication AECL-3081, Atomic Energy of Canada Limited (1968).
[13] LEWIS, W.B., *High burn-up from fixed fuel,* publication AECL-531, Atomic Energy of Canada Limited (1957).
[14] LEWIS, W.B., *Fuelling high conversion ratio reactors,* publication AECL-1911, Atomic Energy of Canada Limited (1963).
[15] LEWIS, W.B., *How much of the rocks and oceans for power? Exploiting the uranium-thorium fission cycle,* publication AECL-1916, Atomic Energy of Canada Limited (1964).
[16] LEWIS, W.B., *Heavy water reactor review and prospect,* publication AECL-2274, Atomic Energy of Canada Limited (1965).
[17] MOORE, R.V., HOLMES, J.E.R., *The SGHWR system,* Proc. Int. Conf. Steam Generating and Other Heavy Water Reactors, London, British Nuclear Energy Society (1968) 3.
[18] BAIN, A.S., *Crack healing and void movement during irradiation of ThO_2 –2wt% UO_2,* publication AECL-3008, Atomic Energy of Canada Limited (1967).
[19] LEWIS, W.B., *Designing UO_2 and ThO_2 fuel for high power rating, long life and negligible sheath strain,* publication AECL-3737, Atomic Energy of Canada Limited (1971)(in press).
[20] OLSEN, A.R., et al., *Irradiation behaviour of thorium–uranium alloys and compounds,* Rep. IAEA Panel Utilization of Thorium in Power Reactors, Vienna (1965) 246; USAEC Rep. ORNL-TM-1142 (1965).
[21] KASTEN, P.R., *The role of thorium in power-reactor development,* Atom. Energy Rev. 8 3 (1970) 473.
[22] JEFFS, A.T., *Thermal conductivity of ThO_2–PuO_2 under irradiation,* publication AECL-3294, Atomic Energy of Canada Limited (1969).
[23] DURET, M.F., *Experience with thorium oxide fuel at Chalk River,* paper AED-Conf-68-083-009 presented at IAEA panel on the Utilization of Thorium in Power Reactors, Vienna (1968); unpublished report CRNL-141, Atomic Energy of Canada Limited (1968).
[24] MacDONALD, R.D., *Irradiation performance of Zircaloy sheathed fuel elements with metallic or ceramic discs between UO_2 pellets,* publication AECL-3606, Atomic Energy of Canada Limited (1970).
[25] LEWIS, W.B., CHURCH, T.G., *Electricity supply in Canada and the role of nuclear power,* Proc. 3d Int. Conf. Peaceful Uses Atomic Energy, 1, United Nations (1964) 53.
[26] PHILLIPS, G.J., GRIFFITHS, J., *LATREP users manual,* publication AECL-3857, Atomic Energy of Canada Limited (1970).
[27] HALSALL, M.J., *BOUT, a program for integrating the burnup equations for thorium and uranium fuels,* publication AECL-2693, Atomic Energy of Canada Limited (1967).
[28] LEWIS, W.B., *Thorium-fuelled CANDU reactors,* to be published as AECL-3905, Atomic Energy of Canada Limited.

CNEN WORK ON THE URANIUM-THORIUM CYCLE APPLIED TO WATER REACTORS

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Abstract–Résumé–Аннотация–Resumen

CNEN WORK ON THE URANIUM-THORIUM CYCLE APPLIED TO WATER REACTORS.

Over the past few years CNEN has been working on a program aimed at assessing the competitiveness of the thorium-^{233}U cycle in water reactors. The program carried out so far includes a preliminary economic evaluation for the optimization of a 500-MW(e) CIRENE Th plant (pressure tube, heavy-water moderated and two-phase light-water-cooled reactor) fuelled with either thorium-^{233}U or uranium-Pu. To confirm the satisfactory results obtained, further research was carried out. To check nuclear parameters of the uranium-thorium cycle, physics measurements were carried out in co-operation with AB Atomenergi and the OECD Halden Project (material buckling, axial flux distributions, spectral indexes, conversion ratios, etc.) on seven 2% enriched ^{235}U 7-pin cluster fuel assemblies, fabricated by extrusion at CNEN's Laboratory of Saluggia. In co-operation with Euratom, in the ECO reactor (Ispra), the following experiments were performed: substitution of 1, 5 and 9 assemblies, with different square lattices and coolants, for buckling measurements; activation measurements (spectral indexes, initial conversion ratios, flux distributions, etc.) with different lattices and coolants. All the results obtained are presented and discnssed in the paper. To determine (U-Th)O_2 fuel behaviour, seven fuel elements (those made at the Saluggia Laboratory) and five high burn-up and heat rating rigs were irradiated in the Halden reactor. The paper reports the results of the irradiation and of the post-irradiation examinations. CNEN's greatest effort in the field of uranium-thorium cycle applications is represented by the ITREC pilot plant. Its operation, described in a separate paper, enables the technical-economic data for a complete evaluation of the uranium-thorium cycle costs to be obtained.

ACTIVITES DU CNEN CONCERNANT LES APPLICATIONS DU CYCLE URANIUM-THORIUM DANS LES REACTEURS A EAU.

Le CNEN a entrepris il y a quelques années un programme à long terme visant à évaluer la compétitivité du cycle thorium/^{233}U dans les réacteurs à eau. Les activités ont jusqu'ici consisté tout d'abord à procéder à une évaluation préliminaire de rentabilité ayant pour but d'optimiser l'installation CIRENE-Th de 500 MW(e). Il s'agit d'un réacteur à tubes de force, ralenti à l'eau lourde et refroidi à l'eau légère en double phase. Le combustible est soit du thorium-^{233}U, soit de l'uranium-plutonium. Pour confirmer les résultats encourageants obtenus, on a procédé à d'autres recherches visant tout d'abord à vérifier, en coopération avec l'AB Atomenergi et le Projet Halden de l'OCDE, les paramètres nucléaires du cycle uranium-thorium (mesure du laplacien matière, de la distribution axiale du flux, des indices spectraux, des rapports de conversion, etc.), sur des assemblages combustibles constitués par des grappes de sept aiguilles d'uranium-235 enrichi à 2%, fabriquées par le procédé d'extrusion au Laboratoire du CNEN de Saluggia. En coopération avec EURATOM, on a procédé, dans le réacteur ECO d'Ispra, à l'expérience suivante: substitution de 1, 5 et 9 éléments, avec différents réseaux de forme carrée et différents caloporteurs, pour des mesures du laplacien; mesures par activation (indices spectraux, rapport initial de conversion, distribution du flux, etc.) avec différents réseaux et caloporteurs. Tous les résultats obtenus sont présentés dans le mémoire. Pour déterminer le comportement du combustible (U-Th)O_2, on a irradié ensuite dans le réacteur Halden sept éléments combustibles (fabriqués au Laboratoire de Saluggia), ainsi que des cellules à taux de combustion et à puissance thermique élevés. Le mémoire rend compte des résultats de cette irradiation et des examens effectués après l'irradiation. Le plus grand effort du CNEN dans le domaine du cycle uranium-thorium est représenté par la centrale pilote ITREC. Son exploitation, décrite dans un autre mémoire, a permis d'obtenir les données techniques et économiques pour une évaluation complète des coûts du cycle uranium-thorium.

РАБОТЫ НАЦИОНАЛЬНОГО КОМИТЕТА ПО ЯДЕРНОЙ ЭНЕРГИИ ИТАЛИИ В СВЯЗИ С ПРИМЕНЕНИЕМ УРАН-ТОРИЕВОГО ЦИКЛА В ВОДО-ВОДЯНЫХ РЕАКТОРАХ.

В течение последних нескольких лет Национальный комитет по ядерной энергии Италии разрабатывал программу, направленную на оценку преимуществ применения цикла то-

рий-уран-233 в водо-водяных реакторах. Работы, проведенные в этих целях, сводятся к следующему: предварительная экономическая оценка оптимизации установки CIRENE мощностью 500 МВт (эл) (канальный тяжеловодный реактор с двухфазным охлаждением обычной водой) с топливным циклом торий-уран-233 или уран-плутоний. Для подтверждения полученных обнадеживающих результатов проведены дополнительные исследовательские работы. Для проверки ядерных параметров уран-ториевого цикла в сотрудничестве с Объединением "АВ Атомэнерги" и с Организацией экономического сотрудничества и развития (Халленский проект) были проведены физические измерения материального параметра, осевого распределения потока, спектральных коэффициентов, коэффициента воспроизводства на семи топливных сборках, состоящих из семи топливных стержней каждая, с обогащением по урану-235 до 2%, изготовленных методом экструзии в Лаборатории Национального комитета по ядерной энергии в Салюдже. В сотрудничестве с Евратомом на реакторе ECO в Испре были проведены следующие эксперименты: измерение лапласиана при замене 1, 5 и 9 элементов топливной сборки с различным шагом решетки и различными теплоносителями; измерение активации (спектральные характеристики, начальный коэффициент воспроизводства, распределение потока и т.д.) с различными вариантами решетки и различными теплоносителями. Полученные результаты изложены в докладе с комментариями. Для определения поведения топлива $(U-Th)O_2$ в Халленском реакторе подвергали облучению семь тепловыделяющих элементов, изготовленных в лаборатории в Салюдже, и пять образцов с повышенной степенью выгорания и повышенной тепловой мощностью. Результаты облучения и пострадиационных исследований приведены в докладе. Результаты работы Национального комитета по ядерной энергии в области применения уран-ториевого цикла нашли отражение в создании опытной установки ITREC. Изучение работы этой установки, которая подробно описана в отдельном докладе, позволит получить технические и экономические данные, необходимые для полной оценки стоимости уран-ториевого цикла.

ACTIVIDADES DEL CNEN RELACIONADAS CON EL CICLO DEL URANIO-TORIO APLICADO A REACTORES CON AGUA.

El CNEN ha trabajado estos últimos años en un programa destinado a establecer si, en los reactores de agua, el ciclo del torio-^{233}U puede ser competitivo. Hasta el momento se lleva realizado lo que sigue: una evaluación económica que consiste en un cálculo preliminar para optimar una instalación CIRENE (reactor de tubos de presión, moderado por agua pesada y refrigerado por una mezcla bifásica de agua ligera) de 500 MW(e), bien sea quemando torio-^{233}U o uranio-plutonio. A fin de corroborar los satisfactorios resultados obtenidos se han realizado diversos experimentos. Para comprobar los parámetros nucleares del ciclo de uranio-torio, se han realizado los siguientes experimentos de física: En colaboración con la AB Atomenergi y la organización del Programa de Halden de la OCDE, medidas de laplacianas de material, distribuciones axiles del flujo, índices espectrales, razones de conversión, etc., hechas con siete conjuntos combustibles de haces de siete varillas de uranio enriquecido al 2%, fabricadas por extrusión en el Laboratorio de Saluggia (CNEN). En colaboración con la Euratom, utilizando el reactor ECO de Ispra, se han efectuado los siguientes experimentos: Medidas de laplacianas de material, por el método de sustitución, de 1, 5 y 9 conjuntos, con diferentes pasos de red cuadrada y con diversos refrigerantes; medidas de activación con diferentes redes y refrigerantes (índices espectrales, razones iniciales de conversión, distribuciones de flujo, etc.). Todos estos resultados se exponen y se comentan en la memoria. Para comprobar el comportamiento del $(U-Th)O_2$ como combustible, se irradiaron en el reactor de Halden los siete elementos fabricados en Saluggia y otros cinco conjuntos, de alto grado de quemado y fuerte desprendimiento de calor. La memoria da cuenta de los resultados de las irradiaciones y de los exámenes posteriores. El máximo exfuerzo del CNEN dedicado al ciclo del uranio-torio está representado por la planta-piloto ITREC. Su funcionamiento se describe en otra memoria y permite obtener los datos técnicos y económicos necesarios para evaluar por completo los costes del ciclo del uranio-torio.

1. INTRODUCTION

To utilize completely all the available energy resources, many countries have begun to exploit the uranium-thorium cycle in addition to the uranium-plutonium cycle.

The Italian program on the uranium-thorium fuel cycle began in 1960 with the design and construction of the ITREC pilot plant for remote reprocessing and refabrication of irradiated fuel elements for water reactors. The plant is located in the province of Matera in southern Italy.

After a general evaluation of several reactor systems that have thorium as the fertile material, which was carried out in the early sixties, in 1965 CNEN, in relation to the Italian program on Cirene-type reactors (pressure tube, heavy-water-moderated and two-phase light-water-cooled reactors), started a long-term program aimed at assessing the competitiveness of the thorium-uranium cycle in these reactors.

First of all, an economic optimization study, extended to all plant costs, was carried out to determine the minimum overall cost of energy produced by a 500-MW(e) Cirene-Th reactor, with an associated reprocessing plant satisfying the requirements of 10 000 MW(e) [1, 2]. The economic optimization was accomplished essentially by taking into account the most significant design parameters (i.e. specific fuel power, irradiation level, moderator-to-fuel ratio, number of rods per bundle, channel height, clearance between rods) and by utilizing a consistent set of economic and design data, with regard to the Italian situation, both for plant construction and for fuel cycle operation and nuclear material supply. The economic influence related to several uncertain design hypotheses was also evaluated.

TABLE I. ECONOMIC EVALUATION – CONCLUSIVE DATA

Fuel cycle	U-Th	Enriched U		Natural U
		Euratom method	Cise method	Cise method
Specific power (W/g)	40	20		20
Irradiation level (MWd/t)	25 000	15 000	15 070	5925
Radial form factor	0.80	0.85		0.85
Number of rods in the bundle	37	37		37
Linear conductivity integral (W/cm)	64.25	46		46
Clearance between the rods (cm)	0.3	0.3		0.1478
Pellet diameter (cm)	1.125	1.319		1.387
Clad rod diameter (cm)	1.267	1.485		1.473
Pressure tube i.d. (cm)	10.985	12.473		11.201
Lattice pitch (cm)	23.985	25.473		27.811
Moderator-to-fuel ratio	12.16	9.60		11.50
Core active height (cm)	270	480		350
Coolant max. specific flow ($g/cm^2.s$)	350	250		376
Mean steam quality	0.177	0.255		0.225
Make-up U specific requirements ($kg\ U_3O_8/MWd$)	0.064	0.159	0.131	0.177
Fuel cycle cost (total) (mills/kWh)	1.119	1.043	0.889	1.001
Heavy water cost (mills/kWh)	0.108	0.146	0.146	0.167
Capital cost (mills/kWh)	2.687	2.644	2.644	2.699
O. & M. cost (mills/kWh)	0.569	0.560	0.560	0.572
Total energy cost (mills/kWh)	4.483	4.393	4.239	4.439

A similar economic evaluation was also done later on for other interesting fuel cycles (natural uranium or slightly enriched uranium, once-through or with plutonium recycle), which could be utilized in a Cirene-type reactor [3]. It was performed for a 500-MW(e) plant by using, to the extent possible, a consistent set of economic data and a consistent method of analysis. The neutronic calculation method used for the uranium-thorium cycle is, in fact, rather dubious, while the method chosen for the enriched uranium once-through cycle (EURATOM method) gives more pessimistic results than that used for the natural uranium once-through cycle (CISE method).

From Table I, it can be concluded that the enriched uranium cycle is the cheapest one, even though the more pessimistic calculation method is used (with regard to the plutonium recycle cycles, their energy costs are slightly higher than those for the corresponding once-through cycles, although very favourable economic assumptions are made). Nevertheless, from the point of view of saving fuel reserves, the uranium-thorium cycle seems to be the most attractive (see Table I, the specific rate of fuel consumption per unit of energy produced). Therefore, to be able to start on a definite design of a Cirene-Th plant when fuel costs increase in the future, CNEN decided to undertake an experimental program aimed at verifying the most important uncertain design hypotheses and the promising results obtained in the uranium-thorium cycle economic evaluation. The experimental activities concerned mainly neutron physics, irradiation tests, and reprocessing and remote refabrication of thorium-uranium fuels.

2. NEUTRON PHYSICS EXPERIMENTAL PROGRAM

A complex neutron physics program is being carried out as part of CNEN's effort in the evaluation of the thorium-uranium cycle with special emphasis on heavy-water-moderated reactors. The main part of the program has already been completed and consisted of:

(1) buckling measurements, by the substitution method, and detailed parameter measurements on 7-rod bundle elements with 2% ^{235}U content;
(2) buckling measurements, by the substitution method, and detailed parameter measurements on 19-rod and 37-rod bundle elements with 2.4% ^{235}U content.

The results of the buckling measurements are reported in this paper.

2.1. Seven-rod bundle experiment

2.1.1. General

This experiment was performed in the R-0 reactor of Studsvik (Sweden) as a common effort by CNEN, AE of Sweden and the Halden Project [4].

Four different pitches were studied (11, 13, 15 and 17 cm in a triangular lattice) with D_2O and air coolants. Seven elements were available for the experiment.

2.1.2. Fuel element data

Fuel rod	Cladding (Zr-2)
Composition: 97.6 wt% ThO_2	i.d. : 12.76 mm
2.0 wt% $^{235}UO_2$	o.d.: 13.96 mm
0.4 wt% $^{238}UO_2$	Total length: 890.5 mm
Diameter: 12.59 mm	
Density: 9.57 g/cm^3	
Active length: 828.5 mm	

Assembly	Shroud tube (Zr-2)
Geometry: 7-rod bundle with 23-mm pitch. Two such bundles were assembled, one on top of the other in the shroud tube.	i.d. : 71 mm o.d. : 73 mm

The fuel of the reference lattice was 3.05 cm in diameter, aluminium canned, natural uranium metal.

2.1.3. Experimental technique

Because of the length of the test assemblies, the critical height was limited to 1.85 m. To keep the critical height of the reference lattice below this value, enriched uranium oxide bundles were placed in the peripheral part of the reactor. The buckling of the 11-cm (more precisely: $19/\sqrt{3}$ cm) reference lattice was obtained with a substitution experiment by adding reference rods to the 19-cm reference lattice. The substitution steps with ThO_2 fuel assemblies are shown in Fig.1.

Persson's method [5] was used for the analysis. Minor changes were introduced in the theory to accommodate eccentric regions and the triangular lattice pattern.

For the elements with air coolant, the $\delta D/D$ correction necessary for the interpretation of the measurements was experimentally determined in the 17-cm reference lattice. The $\delta D/D$ experiment was analysed according to the theory described in Ref. [6]. The results were used to normalize the calculated $\delta D/D$ variations for all the other lattice pitches.

2.1.4. Final results

In the interpretation of the experiment, two possible cell definitions were considered: the first, with triangular cells, is shown on the left-hand side of Fig.1; the second, with rhombic cells, is shown on the right-hand side of the same figure. Moreover, the fuel assemblies were intercompared in the 13-cm lattice and, since small but significant differences were found, correction for fuel element inhomogeneity was applied to the experimental values.

The final results are reported in Table II.

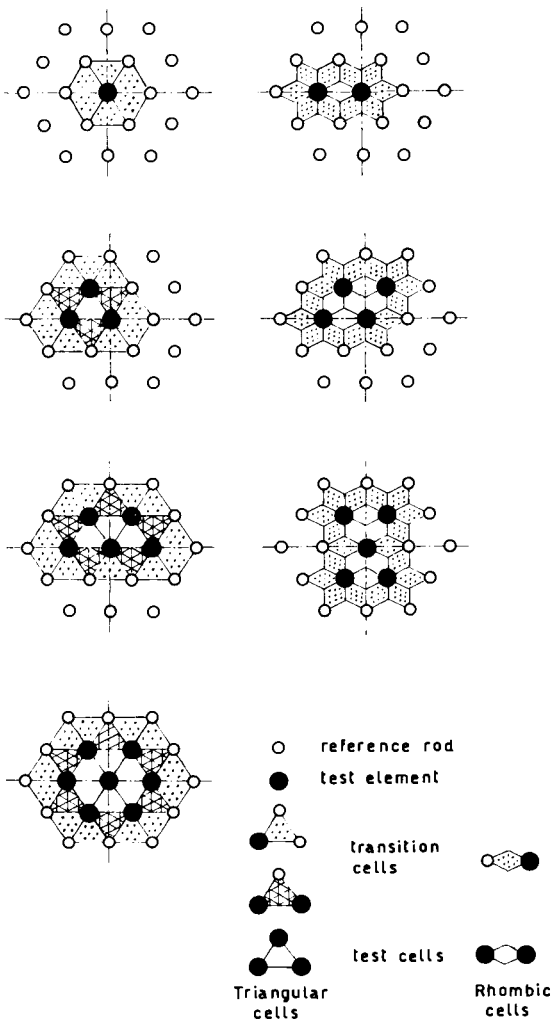

FIG. 1. ThO_2 substitution steps.

2.2. 19- and 37-rod experiments

2.2.1. General

The experiments were performed in the ECO reactor at Ispra in co-operation with CCR Ispra [7]. The total number of reference and fuel elements in the core was 89 for all substitutions. The measurements were performed for 18.8-, 22.3-, 25.5- and 28.05-cm-square lattice pitches and for H_2O, D_2O, air, 30% H_2O-70% D_2O, 50% H_2O-50% D_2O coolants.

TABLE II. 7-ROD BUNDLES – EXPERIMENTAL RESULTS
Temperature: 22°C; moderator composition: 99.645 wt% D_2O

Triangular lattice pitch (cm)	B^2_{ref} (m^{-2})	Type[a] of analysis	B^2_{test} Coolant: D_2O (m^{-2})	B^2_{test} Coolant: air (m^{-2})
11	6.76	Ia	8.36 ± 0.04	
		Ib	8.31 ± 0.04	3.22 ± 0.07
		IIa	8.31 ± 0.07	
		IIb	8.22 ± 0.07	3.07 ± 0.06
13	8.32	Ia	9.87 ± 0.06	
		Ib	9.78 ± 0.06	5.87 ± 0.07
		IIa	9.89 ± 0.08	
		IIb	9.76 ± 0.07	5.87 ± 0.11
15	8.23	Ia	9.83 ± 0.13	
		Ib	9.70 ± 0.08	6.80 ± 0.09
		IIa	9.91 ± 0.16	
		IIb	9.72 ± 0.11	6.79 ± 0.15
17	7.61	Ia	9.14 ± 0.09	
		Ib	9.03 ± 0.05	7.19 ± 0.03
		IIa	9.26 ± 0.14	
		IIb	9.09 ± 0.07	7.22 ± 0.06

[a] Ia: Rhombic transition cell, no fuel inhomogeneity correction.
Ib: Rhombic transition cell, fuel inhomogeneity correction applied.
IIa: Triangular transition cell, no fuel inhomogeneity correction.
IIb: Triangular transition cell, fuel inhomogeneity correction applied.

2.2.2. Fuel element data

Fuel rod (ThO_2-UO_2)

Composition: 2.67 wt% U/(U+Th)
2.40 wt% ^{235}U/(U+Th)
Diameter: 9.86 mm
Density: 9.35 g/cm^3
Active length: 1800, 2000, 2200 mm

Canning (Al 99.5)

i.d. : 10 mm
o.d.: 11.5 mm

Assembly

19 rods with 15.2-mm pitch
37 rods with 13.64-mm pitch

Shroud and calandria tubes (Al 99.5)

19 rods: 76.8/79.8 mm i.d./o.d.
81.8/84.8 mm i.d./o.d.
37 rods: 93/97 mm i.d./o.d.
103/106 mm i.d./o.d.

The normal ECO reference elements were utilized. Nine fuel elements were available for both 19-rod and 37-rod experiments.

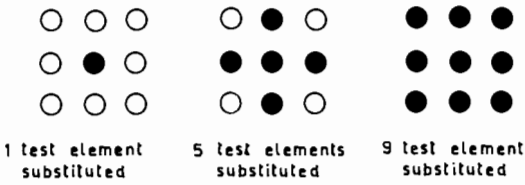

FIG. 2. Test element substitution steps.

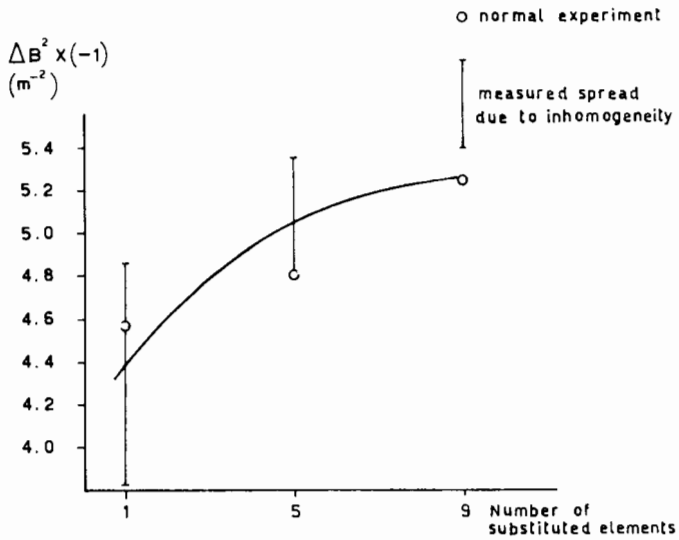

FIG. 3. Inhomogeneity effect in the lattice with 37 rods, 22.3 cm pitch, and D_2O coolant.

2.2.3. Experimental method

The test elements were added to the critical reference lattice stepwise, as shown in Fig. 2, and the critical height measured for one, five and nine substituted elements.

The experimental axial reflector savings of the reference lattices were added to the measured heights to obtain effective critical heights. Calculations were made to find the order of magnitude of the error introduced in the interpretation of the experiments by utilizing fixed reflector savings. This error is of the order of 0.05 m^{-2}.

The analysis was carried out by the direct method with homogenized reactor regions [8]. The nuclear parameters for the reference and test lattices were calculated with the cell code PINOCCHIO [9].

From the analysis, the ΔB_m^2 values of the test lattices were obtained with respect to the reference lattice. These values are given in Table III. By adding the B_m^2 values of the reference lattice obtained with the flux mapping technique, the final B_m^2 of the test lattice were determined and are given in Table IV.

The anomalous trend of the ΔB_m^2 as a function of substituted elements, which may be observed in Table III for the 37-rod experiment, is due to fuel element inhomogeneity. By interchanging in all possible ways the fuel

TABLE III. ΔB_m^2 [m^{-2}]

Coolant	Pitch (cm)	Substituted elements 1	5	9
19-rod elements	D_2O temperature: 23.20 - 24.32°C			
	D_2O purity: 99.725 - 99.963 wt%			
Air	18.8	-6.55	-4.98	-4.94
	22.3	-5.06	-4.07	-4.07
	25.5	-4.22	-3.76	-3.76
	28.05	-3.28	-3.33	-3.33
D_2O	18.8	-7.34	-5.86	-5.86
	22.3	-5.35	-4.55	-4.45
	25.5	-4.18	-3.83	-3.85
	28.05	-3.50	-3.27	-3.29
H_2O	18.8	-4.46	-3.54	-3.41
	22.3	-2.59	-1.63	-1.50
	25.5	-1.35	-0.71	-0.62
	28.05	-1.14	-0.34	-0.24
0.3 H_2O	18.8	-6.16	-5.35	-5.40
	22.3	-4.77	-3.81	-3.79
0.7 D_2O	25.5	-3.88	-2.99	-2.96
	28.05	-1.14	-2.46	-2.42
37-rod elements	D_2O temperature: 21.48 - 26.10°C			
	D_2O purity: 99.600 - 99.686 wt%			
Air	18.8	-3.93	-3.76	-4.06
	22.3	-4.92	-4.47	-4.8
	25.5	-4.92	-4.68	-5.01
	28.05	-4.52	-4.53	-4.81
D_2O	18.8	-4.45	-4.58	-5.07
	22.3	-4.57	-4.8	-5.25
	25.5	-4.81	-4.81	-5.17
	28.05	-4.68	-4.54	-4.81
H_2O	18.8	-5.97	-6.02	-6.62
	22.3	-4.5	-4.45	-4.89
	25.5	-3.41	-3.59	-3.97
	28.05	-2.42	-2.95	-3.32
0.3 H_2O	18.8	-4.96	-5.25	-5.8
	22.3	-4.82	-4.81	-5.25
0.7 D_2O	25.5	-4.31	-4.44	-4.81
	28.05	-3.60	-3.97	-4.31
0.5 H_2O	22.3	-5.00	-4.8	-5.23
0.5 D_2O	28.05	-3.37	-3.7	-4.04

TABLE IV. EXPERIMENTAL B_m^2 [m^{-2}] OBTAINED FROM THE 9-ELEMENT SUBSTITUTIONS

Coolant \ Pitch (cm)	18.8	22.3	25.5	28.05
19-rod elements				
Air	9.14	8.59	7.82	6.93
D_2O	10.07	8.97	7.90	6.88
H_2O	7.61	6.02	4.67	3.82
0.3 H_2O/0.7 D_2O	9.60	8.30	7.01	6.01
37-rod elements				
Air	8.27	9.32	9.06	8.41
D_2O	9.27	9.77	9.22	8.40
H_2O	10.83	9.41	8.02	6.91
0.3 H_2O/0.7 D_2O	10.00	9.77	8.86	7.90
0.5 H_2O/0.5 D_2O	-	9.75	-	7.63

elements for the 22.3-cm pitch D_2O coolant, the results shown in Fig.3 were obtained. The "normal experiment" refers to the loading sequence which was followed for all substitution experiments. While the inhomogeneity practically does not affect the ΔB_m^2 relative to the 9-element substitution, chemical analyses are being performed to determine more accurately the average ^{235}U content, which is known at present with a $\pm 1.5\%$ uncertainty.

3. IRRADIATION PROGRAM

A series of irradiation experiments on metal-clad fuel rods containing cold-pressed and sintered ThO_2-UO_2 pellets began in 1967, aimed at testing U-Th mixed oxide fuels for Cirene-type reactors.

All the tests were carried out in the OECD Halden reactor and concerned only single-rod performance, because no suitable facility exists in the reactor for testing full-scale Cirene fuel elements (37-rod bundles).

Two groups of experiments were planned. The first one consisted of the irradiation, in the central zone of the Halden reactor, of seven fuel elements, the same as those used for the physics experiments mentioned above, produced with the extrusion and sinterization method adopted in the ITREC plant. The main aims of this experiment were to test the in-pile behaviour of $(U\text{-}Th)O_2$ fuel made with the ITREC's fabrication process, despite the fact that the operating conditions of the fuel were lower than those of the reference 500-MW(e) Cirene-Th reactor, because of the relatively low neutron flux at Halden. The fuel elements, irradiated in the period 1967-1970 at a linear heat rating not higher than 300 W/cm reached an average burn-up of 6000 MWd/t (Th+U) and no rod failures were detected.

The remaining group of experiments had the main aim of obtaining information on the behaviour of $(U\text{-}Th)O_2$ fuel at high burn-up and heat

rating (IFA 125, 126 and 127) and of studying the influence of certain parameters such as pellet length, pellet fabrication method, etc. (IFA 175 and 176).

Table V lists the test section series, their main characteristics and objectives, and their present status.

The IFA 175 and 176 fuel rod diameter and the IFA 125, 126 and 127 ^{235}U enrichment values, larger and higher, respectively, than those determined in Ref. [2] as optimal, were utilized in reaching a significant value of $\int k d\theta$ even in the low neutron flux conditions of the Halden reactor.

Central thermocouples were placed in six rods; all but three, of the tantalum-sheathed type, behaved satisfactorily.

Temperature values up to 2400°K were measured and the related thermal conductivity data are in reasonable agreement with those reported in Ref. [10].

Fission gas retention was better than in UO_2; a release as small as 4% was measured in rods at 2500-2800°C peak central temperature. The post-irradiation gas pressure in the plenum of rods irradiated up to 2000 MWd/t (Th+U) was lower than the pre-irradiation pressure; the closed pores in the fuel are likely to be the reason for this.

The post-irradiation examinations showed that all the rods but two (IFA 176) were undamaged.

Since: (a) in the 25-mm-diameter rod samples, the transversal gamma scanning and the autoradiographies revealed remarkable dissymmetry in neutron flux and specific heat rating distribution, and (b) the moisture content measured in ThO_2 pellets was seven times higher than in similarly handled UO_2 pellets, the failures in the IFA 176 rods were attributed to the local stress concentration (a) in the Zircaloy-2 cladding material embrittled by the high fuel moisture content (b).

The metallographic and ceramographic analyses showed that the ThO_2-UO_2 behaviour was similar to that of uranium oxide fuel, with some peculiarities relative to the temperature at which the structural changes take place.

In the fuel, with operating central temperatures of:

up to 1400°C, as in IFA 175, non-significant structural changes are evident;

up to 1800°C, as in IFA 176, a large zone is evident with equiaxed grain growth, but no columnar grain growth (Fig.4(a));

up to 2300°C and with a burn-up of 2500 MWd/t (Th+U), as in IFA 125, there is no columnar grain growth in the circumferential inner zone, but only some along the cracks (Fig.4(b));

up to 2800°C and with a burn-up of about 15 000 MWd/t (Th+U), as in IFA 127, a large internal zone is evident showing columnar grain growth, but only one sample out of three shows an internal hole as small as 0.06 radius (Fig.4(c)).

4. REPROCESSING AND REMOTE REFABRICATION

Research aimed at developing a fuel fabrication procedure which could be adopted by the remotely operated ITREC plant began very early at CNEN's Fuel Fabrication Laboratory in Saluggia.

TABLE V. SUMMARY OF CNEN THORIUM FUEL CYCLE PROGRAM: IRRADIATION OF COLD PRESSED AND SINTERED PELLET ZIRCALOY-2 CLAD RODS

Test assembly No.	No. of rods	Fuel material	Fuel density (%) T.D.	Fuel rod dimensions				Irradiation period		Bundle average burn-up MWd/t(U+Th)	Status	Objective
				Length (mm)	o.d. (mm)	Wall (mm)	Diametral clearance (mm)	From	To			
IFA 125	6	ThO_2- 10 wt% UO_2[a]	94	770	14	0.6	0.18 - 0.30	Feb. 1968	Apr. 1968	2310[c]	Examined	Obtain high heat rating and examine the influence of diametral clearance on fuel central temperature and canning elongation.
IFA 126	3	ThO_2-10 wt% UO_2[a]	94	770	14	0.6	0.18 - 0.30	Feb. 1968	In progress	27400[d]	In pile	Study ThO_2-UO_2 at high heat rating and high burn-up.
IFA 127	3	ThO_2-10 wt% UO_2[a]	94	770	14	0.6	0.18 - 0.30	Feb. 1968	Oct. 1969	15600	Examined 2 rods	Same as for IFA 126; unloaded for instrumentation failure.
IFA 175	8	ThO_2 - 3.3 wt% UO_2[b]	90-95	300	25	1.0	0.16 - 0.24	Sep. 1968	Jan. 1969	1340[c]	Examined 3 rods	Study Cirene-thorium type material. Compare cold pressed and extruded fuel[e].
IFA 176	8	ThO_2- 3.3 wt% UO_2[b]	90-95	300	25	1.0	0.16 - 0.24	Feb. 1969	May 1969	1240[c]	Examined 4 rods	Same as IFA 175.

[a] Uranium enriched in ^{235}U to 93 wt%.
[b] Uranium enriched in ^{235}U to 90 wt%.
[c] Based on post-irradiation examinations (Nd-method) and on 200 MeV/fission.
[d] As up to January 1971, based on calorimetry.
[e] So far no successful production of 23-mm-diam. extruded fuel.

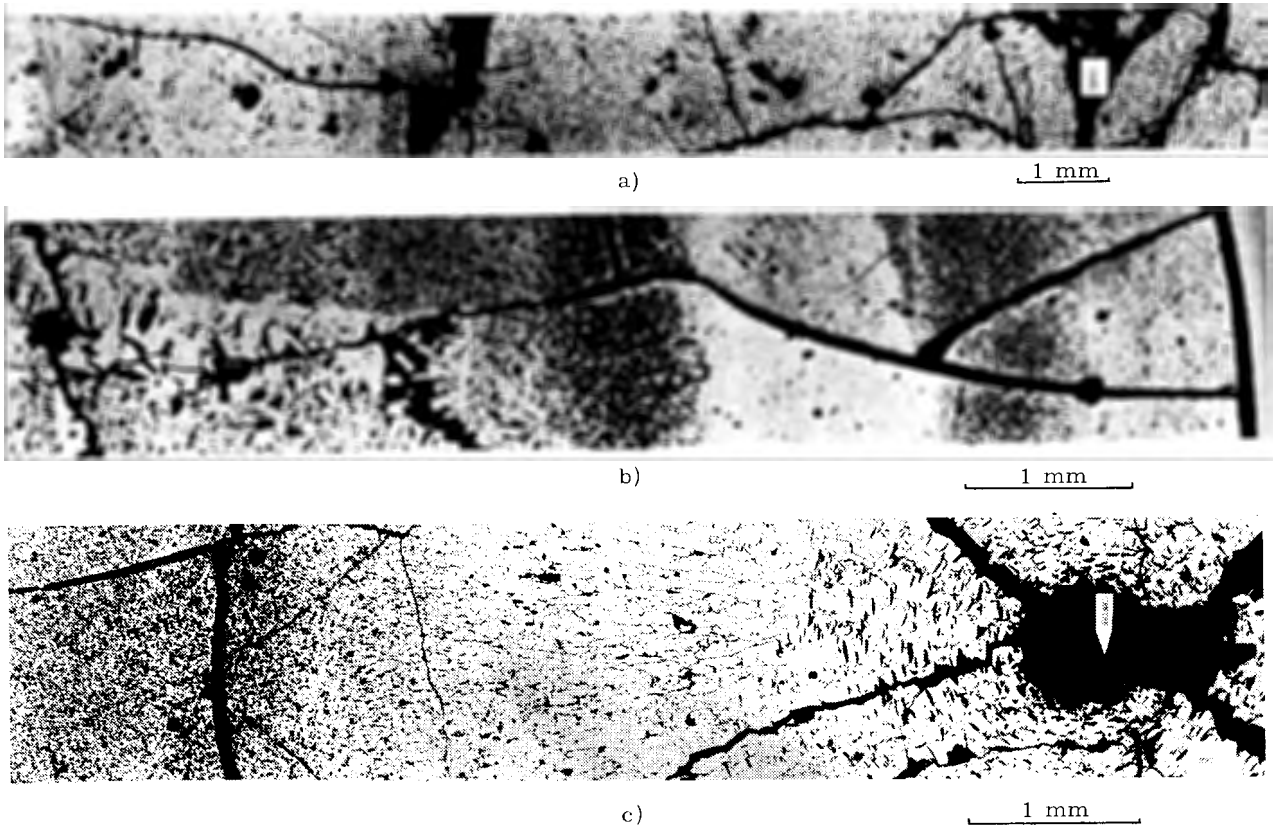

FIG. 4. Radial micrographs of three differently rated rods: (a) from IFA 176, estimated central temperature 1800°C; (b) from IFA 125, estimated central temperature 2300°C, burn-up 2500 MWd/t; (c) from IFA 127, estimated central temperature 2800°C, burn-up 15 000 MWd/t.

The steps of the experimental program were:

(a) chemical research for developing an acceptable preparation procedure for mixed thorium-uranium oxides with good and reproducible sintering behaviour;

(b) development in ceramics aimed at setting up the best operating conditions for obtaining high-density pellets;

(c) fuel test fabrication for irradiation experiments;

(d) pilot-scale fabrication of approximately one tonne of sintered pellets to verify the sinterability of the procedure developed.

The Th-U oxide preparation procedure chosen was characterized by co-precipitation at 60°C of a mixed Th-U-(IV) oxalate, followed by a calcining step at 900°C in air. Strict control of the operating conditions is very important to obtain powder with constant properties (such as specific surface, shape and size of the grain) [11].

Ceramic research was extensively performed, and two compacting processes in particular were studied: pressing and cold extrusion. Both these methods gave good results as far as pellet soundness, density and structure were concerned. The pressing method consists of a pre-compacting stage followed by pressing the granulated powder with binder and lubricant at 4 t/cm^2; sintering is then performed in hydrogen at 1750°C. Approximately 200 kg of pellets for the Halden irradiation tests were produced.

Extrusion methods allow the fabrication of pellets much longer (4 or 5 times) than those obtained by pressing, but the pellet soundness specification is more difficult to meet; only strict control of all the operations (paste preparation, pre-extrusion, extrusion) [12-15] can avoid this drawback.

The values reported above were confirmed by two pilot fabrication runs made with the pressing and extrusion processes (500 kg each).

The greatest effort in fuel reprocessing and remote refabrication made by CNEN is represented by the ITREC pilot plant, the detailed description of which is given by Orsenigo et al. elsewhere in these Proceedings. Its operation will enable us to obtain the technical-economic data for a complete evaluation of the uranium-thorium cycle.

REFERENCES

[1] CARDOSI, G., VENDITTI, P., Valutazione economica del ciclo uranio-torio in un reattore tipo Cirene, CNEN Rep. RT/FI(66)42 (1966).

[2] CARDOSI, G., MAZZANTI, A., VENDITTI, P., Progetto preliminare e valutazione economica di un reattore tipo Cirene alimentato ad ossido di torio, CNEN Rep. RT/FI(67)29 (1967).

[3] CARDOSI, G., MAZZANTI, A., VENDITTI, P., Analisi comparativa di vari cicli di combustibile in un reattore tipo Cirene, CNEN Rep. RT/FI(68)39 (1968).

[4] PERSSON, R., ANDERSSON, A.J.W., GRIFONI, S., SCHITTENHELM, C., SOKOLOWSKI, E.K., TOSI, V., Reactor Physics Measurements on $ThO_2/^{235}UO_2$ Fuel Assemblies in R-0, AE Rep. AE-FFR-113 (1967).

[5] PERSSON, R., Perturbation Method of Analysis Applied to Substitution Measurements of Buckling, AE Rep. AE-256 (1966).

[6] PERSSON, R., One Group Perturbation Theory Applied to Measurements with Void, AE Rep. AE-248 (1966).

[7] CCR Ispra Euratom - CNEN, to be published.
[8] CASINI, G., PAILLON, N., Buckling measurements with a reduced number of fuel elements, J. nucl. Energy 21 (1967) 7.
[9] AMYOT, L., CASINI, G., CUNIBERTI, R., DAOLIO, C., PINOCCHIO: a Computer Program for all Reactivity Calculation, Euratom Rep. EUR-4231 e (1969).
[10] SPRINGER, J.R., ELDRIDGE, E.A., GOODYEAR, M.U., WRIGHT, T.R., LAGEDROST, J.F., Fabrication, Characterization and Thermal Property Measurements of ThO_2-UO_2 Fuel Material, USAEC Rep., BMI-X-10210 (October 1967).
[11] COGLIATI, G., DE LEONE, R., FERRARI, S., GABAGLIO, M., LISCIA, A., "Preparation of ceramic-grade thorium-uranium oxide", New Nuclear Materials Including Non-Metallic Fuels (Proc. Conf. Prague, 1963) 1, IAEA, Vienna (1963) 37.
[12] LISCIA, A., "Problèmes sur le frittage de l'oxyde de thorium et des oxydes mixtes à faible teneur d'uranium", Attività tecnologiche italiane ed iugoslave, CNEN, Roma (1964).
[13] MILLER, R.S., LISCIA, A., "Extrusion fabrication of recycled thoria-urania in the PCUT Plant", Proc. 1st Int. Symp. on Thorium Fuel Cycle, USAEC Rep. TID-7650 (1962) 553.
[14] GABAGLIO, M., LISCIA, A., Fabbricazione di corpi ceramici in ThO_2-UO_2, Energia Nucleare (April 1966).
[15] LISCIA, A., ZANELLI, S., Sulla fabbricazione di pastiglie sinterizzate di ossidi misti di torio ed uranio a bassa concentrazione di uranio per elementi di combustibile, La Metallurgia Italiana 4 (1971).

THE KEMA SUSPENSION TEST REACTOR

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Abstract–Résumé–Аннотация–Resumen

THE KEMA SUSPENSION TEST REACTOR.

Elaborate studies of reactor strategies for the decades to come have shown the potential benefit of using homogeneous liquid fuel reactors as thermal breeders or near breeders. Their low fissile fuel inventory can be of the utmost importance for the economy of a rapidly expanding nuclear power system, even if fast breeder reactors are available at the same time. Apart from fuel salt solutions or molten salts, aqueous fuel suspensions based on the ^{233}U-Th cycle could be used in this respect. The possible uses and limitations of the aqueous suspensions have been studied extensively for this purpose. On the basis of the results of these studies, a 1-MW(th) facility has been constructed to explore the dynamic behaviour of a reactor system operating with different types of fuel suspension. The facility, which will begin operation shortly, is described together with its experimental possibilities and its program. Several specific features and technical and fundamental limitations of the application of different suspensions are discussed on the basis of existing knowledge and experience with non-critical loops.

LE REACTEUR D'ESSAI A SUSPENSION DE LA KEMA.

Des études approfondies de l'avenir des réacteurs pendant les décennies qui viennent ont montré les avantages éventuels des réacteurs à combustible liquide homogène utilisés comme surgénérateurs ou quasi-surgénérateurs à neutrons thermiques. Les réacteurs qui n'exigent qu'une petite quantité de combustible fissile peuvent être extrêmement importants pour la rentabilité d'un réseau nucléo-électrique en développement rapide, même si les réacteurs surgénérateurs à neutrons rapides existent en même temps. Mis à part le combustible sous forme de sels en solution ou de sels fondus, les suspensions de combustible dans l'eau fondées sur le cycle ^{233}U-Th peuvent être utilisées à cette fin. Les emplois possibles et les limites des suspensions aqueuses ont fait l'objet d'études approfondies. Sur la base des résultats obtenus, on a construit une installation de 1 MW(th) pour étudier le comportement dynamique d'un réacteur fonctionnant avec différents types de suspensions de combustible. Le mémoire décrit cette installation, dont l'exploitation commencera prochainement, ainsi que ses possibilités expérimentales et son programme. Les auteurs signalent plusieurs caractéristiques spécifiques ainsi que les limites techniques et théoriques de l'emploi de différentes suspensions sur la base des connaissances actuelles et de l'expérience acquise avec des boucles non critiques.

ОПЫТНЫЙ РЕАКТОР "КЕМА" С ТОПЛИВНОЙ СУСПЕНЗИЕЙ.

Тщательные исследования реакторной стратегии на предстоящие десятилетия показали потенциальные преимущества использования реакторов с жидким топливом в качестве размножителей или почти размножителей на тепловых нейтронах. Малая загрузка делящихся веществ для этих реакторов может иметь решающее значение с точки зрения экономически быстро расширяющейся ядерно-энергетической системы, даже если к этому времени войдут в строй реакторы-размножители на быстрых нейтронах. В таких реакторах могут быть использованы не только топливные растворы солей, но и водные топливные суспензии, основанные на топливном цикле уран-233 – торий. С этой точки зрения были тщательно изучены возможности и ограничения водных суспензий. На основе этих работ сконструирована опытная установка мощностью 1 МВт(тепл) для изучения динамики реакторной системы, работающей с различными видами топливных суспензий. Описана установка, которая должна войти в строй в ближайшем будущем, ее экспериментальные возможности и программа экспериментов. На основе современных знаний и опыта, полученного на некритических петлях, обсуждаются некоторые особые характеристики и фундаментальные ограничения применения различных суспензий.

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* Central laboratory of the Netherlands electricity supply undertakings.

EL REACTOR PARA PRUEBAS DE SUSPENSION DE KEMA.

El estudio detallado del desarrollo de los reactores en la próximas décadas muestra los beneficios potenciales del empleo de reactores de combustibles líquidos homogéneos como reactores reproductores térmicos o casi reproductores. La pequeña carga de combustibles fisionables que estos reactores precisan puede ser un factor de importancia capital para la economía de un sistema de energía nuclear en rápida expansión, aunque al mismo tiempo se utilicen reactores reproductores rápidos. Aparte de las soluciones salinas o de las sales fundidas, pueden emplearse con este objeto suspensiones acuosas de combustible basadas en el ciclo ^{233}U-Th. Se han estudiado detenidamente las posibilidades y las limitaciones de las suspensiones acuosas. Basándose en los resultados de estos estudios se ha construido una instalación de 1 MW(t) para investigar el comportamiento dinámico de un reactor que funcione con diferentes tipos de suspensiones de combustible. Se describe la instalación, que entrará en funcionamiento en breve, así como sus posibilidades experimentales y su programa. Las características más específicas y las limitaciones fundamentales y técnicas inherentes al empleo de diferentes suspensiones se estudian sobre la base de los conocimientos y experiencias actuales con circuitos no críticos.

1. INTRODUCTION

The general consensus of opinion is that, in the long run, the production of nuclear energy with present-day light-water or high-temperature gas-cooled reactors will be economically unattractive. It even appears to be impossible for the energy need in the coming decades to be fulfilled along these lines. This is due to the limited availability of reasonably cheap ^{235}U and the fact that only 1-2% of the uranium atoms in natural uranium can be fissioned in these reactors.

In breeder reactors, however, fertile uranium and thorium can, in addition, be converted into heat. The application of breeders therefore enables uranium and thorium reserves to be used which would be available abundantly at high prices. The assumption is that the rate of introduction of fast breeders is only limited by the Pu stock available at any given time from already existing reactors.

The large difference, by a factor of more than 100, of the relevant fission cross-sections in thermal and fast breeders results in a considerably lower fissile inventory per kW(e) for the thermal breeder. This inventory is diminished in fast breeders by a substantial increase of the rate of heat extraction. Nevertheless, the inventory of the molten-salt liquid fuel reactor remains smaller by a factor of 3 [1] and that of the aqueous suspension liquid fuel reactor by a factor of 4 (Table I).

The factor of 3 to 4 in the fuel inventory can be decisive for the manner of introducing breeder reactors. In a separate study [2], it is demonstrated that the available quantity of fissile material in the form of Pu produced in light-water reactors will seriously limit the introduction of fast breeders, whereas the same does not hold in the case of thermal liquid fuel reactors (Fig. 1). In the same publication, the fuel cycle costs for different reactor strategies are calculated, resulting in considerably lower costs per kWh for thermal breeders, or even near breeders, than for fast reactors (Fig. 2).

In these calculations, which were carried out for an estimation of Western European nuclear energy policy, it was assumed that in the years to come LWRs will be constructed and the Pu produced by them stocked as inventories for commercial breeder reactors. In this program, the fast breeder will use the ^{238}U-Pu cycle, and the thermal breeder will also be started with Pu, but thereafter operated on the Th-^{233}U cycle. Further-

TABLE I. CHARACTERISTICS OF THE ZERO ENERGY EXPERIMENT AND THE KEMA SUSPENSION TEST REACTOR IN COMPARISON WITH A SUSPENSION POWER REACTOR

	Zero energy experiment	KSTR	Suspension power reactor
Fuel	15% $^{233}UO_2$ - 85% ThO_2	25% $^{235}UO_2$ - 75% ThO_2	1.5% $^{233}UO_2$ - 98.5% ThO_2
Concentration	330 g/l	280 g/l	200 g/l
Moderator	H_2O	H_2O	D_2O
Reflector	BeO-graphite	BeO-graphite	Graphite
Thermal power	-	1 MW	788 MW [a]
Electrical	-	-	250 MW
Power density		50 kW/l	35 kW/l
Temperature	20 - 80°C	250°C	300°C
Conversion ratio			0.99
g ^{233}U/kW(e)	-	-	0.65
kg D_2O/kW(e)	-	-	0.2
Core volume	17 l	20 l	22 m^3
Outer volume	16 l	50 l	26.4 m^3

[a] 1000 MW(e) calculated for 4 units of 250 MW(e).

more, it is assumed that fast breeders can be introduced in the nineteen-eighties and thermal liquid fuel breeders will be available only in the nineteen-nineties.

The results of these calculations are summarized in Fig. 2. The bands represent fuel cycle costs for Western Europe in the years to come and for different reactor strategies. The lower limit of each band is based on a constant price of the uranium mined ($8-$10/lb). For the upper line, a rising uranium price has been supposed, based on the exhaustion of cheap uranium reserves. Both the estimates of the energy need and of the rising price are taken from recent literature [3].

Another important point is the ^{235}U production capacity required for the different strategies. This is smaller for LFRs introduced in 1990 than for FBRs introduced in 1980 (Fig. 3). However, a strategy in which FBRs are introduced first, followed by LFRs, might even be better in this respect.

From the results presented above, the potential of thermal breeders is evident for economic reasons. A more detailed study, to be published elsewhere, will show that an extra benefit could be derived from the simultaneous utilization of thermal and fast breeders.

Consequently, the development of thermal breeders is definitely more than an insurance for the serious material damage to essential constructional parts by fast neutrons currently encountered in fast breeders. It is our strong conviction that the development of thermal breeders must proceed far enough to enable a technical and economic comparison of both types.

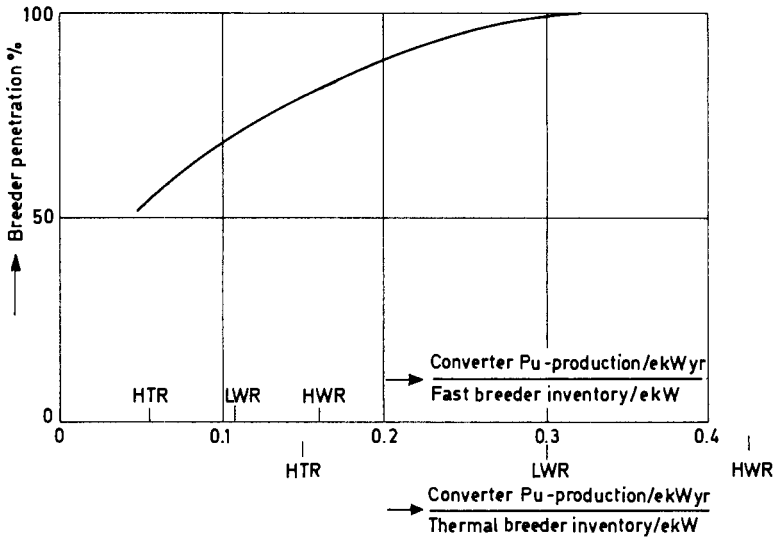

FIG. 1. Breeder (FB or LFR) penetration by the year 2010 in relation to the ratio of Pu production and breeder inventory for different converter reactors: High Temperature (HTR), Light Water (LWR) or Heavy Water (HWR) Reactor.

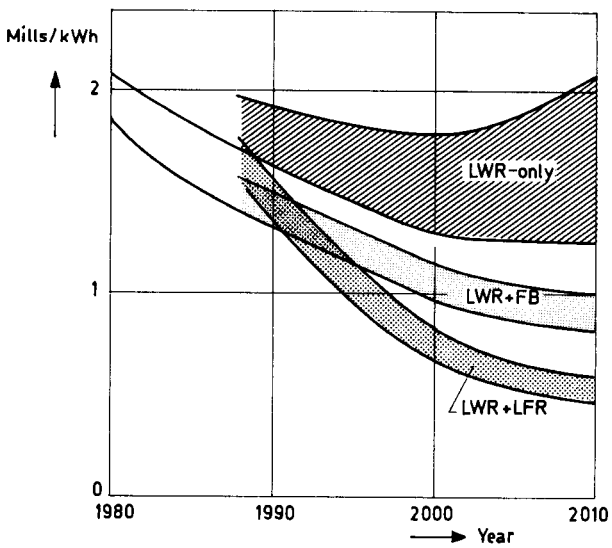

FIG. 2. Annual system fuel cycle costs for total nuclear demand, estimated for Western Europe: LWR = Light Water Reactor, FB = Fast Breeder Reactor, LFR = Liquid Fuel Reactor.

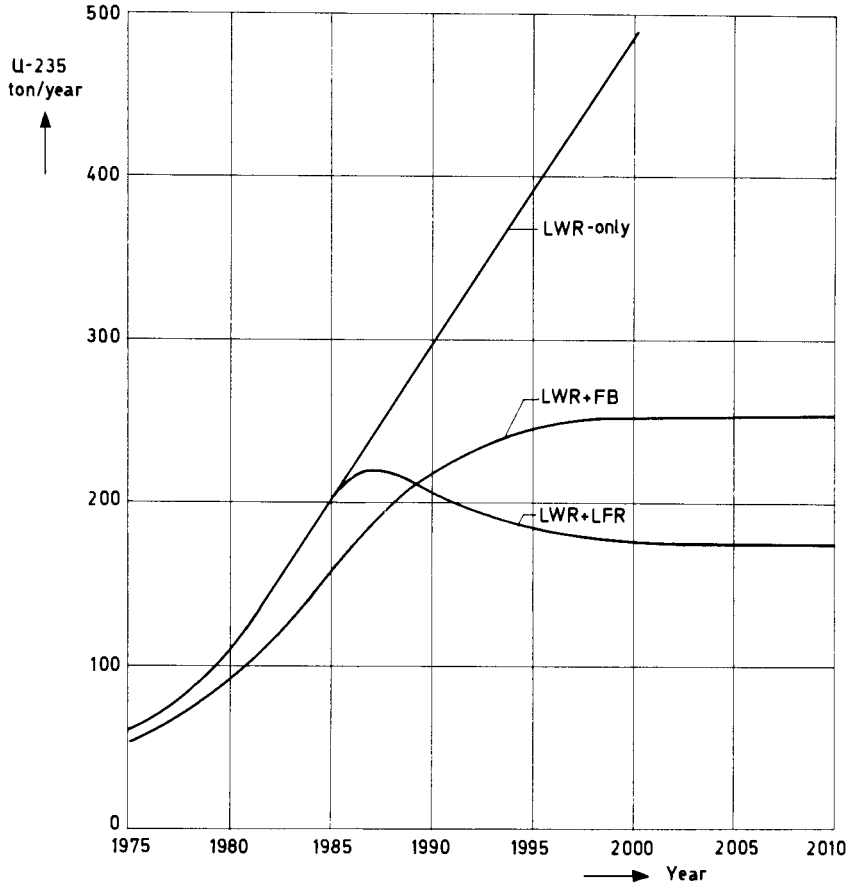

FIG. 3. Annual ^{235}U requirements for different reactor strategies.

Only two centres in the world – Oak Ridge National Laboratory (molten salt reactor) and the KEMA laboratories (aqueous suspension reactor) – are contributing, albeit on a relatively small scale, to this development. The effort spent in the development of Liquid Fuel Reactors (LFR) is therefore surprisingly small in view of their potential in a rapidly expanding system of nuclear power production. Their low fuel investment, high conversion ratio and low fuel refabrication costs seem decisive in this respect.

At the KEMA laboratories, development has reached a stage where an acceptable fuel seems to be available and all the components have been developed for a suspension test reactor. The construction of this reactor has been completed and the operational tests have started. In this paper, this test facility, its program and the development preceding its operation are described.

Certain activities in the project have already been mentioned at previous Geneva conferences [4, 5]. The results reported were obtained by a team of about 120 persons, who, unfortunately, cannot be mentioned

by name. This decisive point in the evaluation of the future of thermal breeders, however, could only be reached through their perseverance and skilled co-operation.

2. LIQUID FUEL REACTORS

The LFRs envisaged so far are characterized by the mobility of the fuel. This requires the development of certain specific components and procedures, not yet available in nuclear technology. A dependable evaluation of the different types of LFR therefore means a considerable amount of reseach and development before conclusions can be drawn. Three reactor projects concerning LFRs might be of importance in the evaluation:

(1) The HRT (Homogeneous Reactor Test) of ORNL. In this facility an aqueous uranium salt solution was used as a fuel. The experiment was stopped in 1962 and this type of fuel is no longer envisaged for an LFR.

(2) The MSRE (Molten Salt Reactor Experiment), which is also an ORNL project. A solution of fissile and fertile material in molten salt is used as the fuel medium The first stage of evaluation (the operation of a 5-MW(th) critical facility) was successfully completed at the end of 1969.

(3) The KSTR (the 1-MW(th) KEMA Suspension Test Reactor) of the KEMA laboratories. An aqueous suspension of (U, Th) oxide is used as a fuel. Research and development have been carried to the stage where the KSTR has been constructed as a critical facility which will be taken into operation shortly.

According to the view of the Netherlands electricity supply undertakings, the evaluation of the suspension reactor must be an integral part of the evaluation of LFRs in general; in particular, the suspension system should be compared with the molten salt concept. Each of the systems has its specific pro's and con's. It cannot be stated off-hand which presents the best perspective:

> Clear advantages of the molten salt reactor are its high temperature and low pressure, which give a low-pressure mechanical construction and high steam quality. The molten salt fuel is a homogeneous system, which simplifies inventory problems. On the other hand, the fuel salt is rather complex and expensive, specific material problems have been encountered and there may be complications with solidification of the salt.
> The suspension reactor has a lower fuel inventory (see Table I), cheaper fuel and a higher conversion ratio. The fuel system can segregate. This may be a nuisance in certain respects, but an advantage in others. For example, the fuel concentration can be adjusted by means of hydrocyclones. Finally, the fuel may attack the enclosing system mechanically by erosion.

An elaborate study is required of both systems before the advantages and disadvantages of each can be quantified to such an extent that a full technical and economical evaluation appears realistic.

3. EVALUATION OF THE SUSPENSION REACTOR

With LFRs, the most characteristic features of a suspension system are:

(1) the direct contact of a solid fuel particle with the transporting liquid;
(2) the homogeneous dispersion of the fuel particles cannot be preserved under all conditions.

Extensive research and development work has been performed with respect to the consequences of these characteristics. With respect to the direct contact between solid fuel and water, it has been demonstrated that the presence of water can increase the radiation damage to the fuel particles quite extensively.

The 5-μm fuel spheres of $(U, Th)O_2$ envisaged for the KSTR have the advantage that fission fragments escape from the fuel by recoil and are thereby accessible for continuous removal. They have the disadvantage that the fission fragments damage the fuel surface at the point of exit and at the same time create a track of very aggressive radiolytic species just outside the surface. It could be shown from capsule irradiations at ambient temperature that roughly 10^3 heavy metal atoms are removed from the fuel surface per damaged surface spot. Under the conditions of the reactor, the debris covers the fuel surface and may even grow together with it because of the enhanced rate of crystallization and crystal growth. Experiments have indicated:

(1) that, at ambient temperature, the fuel does not survive a burn-up of 200 MWd/t;
(2) that, at 250°C, the rate of damage has decreased by a factor of roughly 3,
(3) that hydrogen overpressure, radical scavengers, and a modified fuel composition do not provide a technical means of preventing the damage;
(4) that, at 250°C, an equilibrium fuel size of about 1 μm may be expected, which would be fully acceptable;
(5) that, in preliminary experiments, the rate of damage at 300°C is about 1% of that at ambient temperature (Fig. 4).

A stable fuel thus seems to be available for a power reactor. However, further studies of this fuel will have to be carried out separately and in parallel with the KSTR operation, because the latter can only be operated at a maximum temperature of 250°C.

The tendency of the fuel suspension to segregate under certain conditions has as a consequence that the reactor kinetic behaviour might depend strongly on the hydrodynamic and rheological properties of the fuel system.

The technological aspects of such problems were studied very elaborately in high-temperature loops, whereas a first indication of the reactor kinetic behaviour could be derived from a low-temperature zero energy assembly, mainly by neutron multiplication measurements and by neutron noise analysis.

To extend these studies to a higher temperature (250°C) and to conditions in which, in addition, heat generation and radiolysis may change the reactor behaviour, the KEMA Suspension Test Reactor was constructed as a high-temperature facility which is now available for operation.

FIG. 4. Electromicrographs of 5-μm 15% UO_2-85% ThO_2 fuel microspheres: (a) unirradiated, (b) irradiated at 80°C, burn-up 100 MWd/t, (c) irradiated at 300°C, burn-up 1000 MWd/t.

Erosion problems, to a certain extent also arising from the fact that the suspension system is actually not fully homogeneous, have been studied in technological loops. Adequate methods have been developed to limit the phenomenon to an acceptable level.

The full evaluation comprises roughly three stages before a demonstration reactor can be built:

(1) Extensive studies concerning the colloid chemical and irradiation stability of the fuel, the rheology and the hydrodynamic behaviour of a suspension system, the fission product recoil, and first indications of the reactor kinetic behaviour at a low temperature. From the results it was decided to construct a small 1-MW(th) reactor operating under power reactor conditions, although, for technical reasons, relevant at the time of decision, the maximum temperature would be restricted to 250°C.

(2) In support of this decision, components and procedures were developed, process studies were made, technological characteristics were determined, fuel preparation processes were developed and a design of the reactor was made. In part parallel with these activities, the construction was carried out.

(3) With the facility completed, a new period of research will start to reveal the dynamic behaviour of a suspension reactor under various operating conditions and to understand the interaction between the technological behaviour of the suspension system and the nuclear behaviour of the reactor. Valuable operational experience will be collected at the same time. However, since the temperature and the fuel composition in the KSTR are different from those of a suspension power reactor, a parallel program of fuel irradiations is required to evaluate the fuel lifetime under actual conditions.

The first two stages of evaluation have now been completed and, after a short period of performance testing of the KSTR system, it will be operated with fuel, starting in mid-1972.

4. DESCRIPTION OF THE KEMA SUSPENSION TEST REACTOR

In this section, a very general description of the central part of the KSTR is presented.

The main component of the suspension system is a 20 litre, more or less spherical core tank with the inlet at the bottom and the outlet at the top. The required flow pattern of the suspension in the core is achieved through a rigid system of vanes and cones in the inlet (Fig. 5). To ensure a rather flat flux distribution, the wall thickness of the core is kept to a minimum and the core is therefore surrounded by a pressurized BeO-graphite reflector inside a pressure vessel. The fuel consists of spherical $(U, Th)O_2$ particles of about 5 μm in a concentration of 200-300 grams per litre in light water. A mean power density of 50 kW/l is envisaged for the core. Differing from the conditions of a power reactor are the temperature (250° versus 300°C), the uranium content of the fuel (about ten times higher for the KSTR) and the use of light instead of heavy water (Table I). Furthermore, the main system contains a pump, a heat exchanger and a gas injector and separator (Fig. 6).

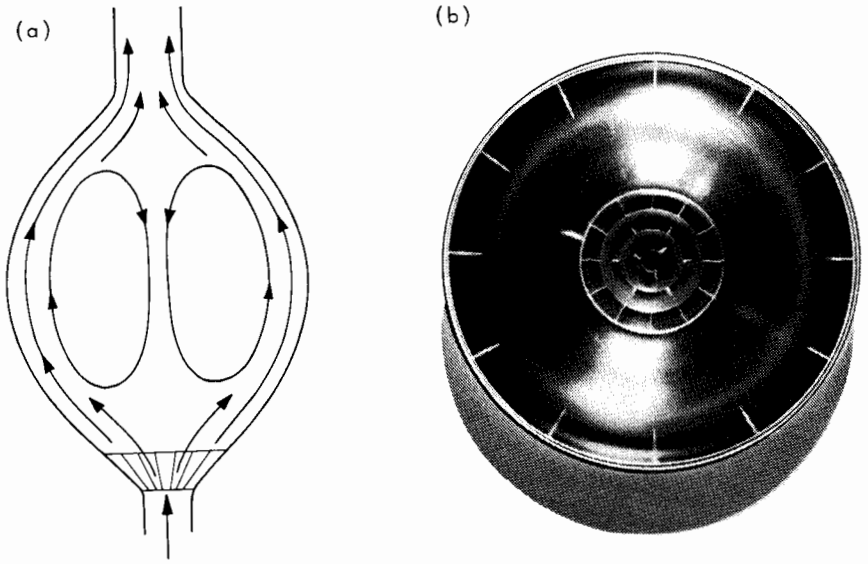

FIG. 5. (a) KSTR core vessel with (b) flow distributor.

In open connection with the suspension system is a gas system. The latter serves several purposes: pressurization of the reactor main system, recombination of radiolytic gases from the core, purge water production from the suspension and suppression of the xenon poisoning. The system is connected to the main suspension circuit through a gas-liquid contactor and a gas-separator unit.

The third important system is the cooling system connected to the main heat exchanger. In the sub-critical phase of operation, where the temperature must be maintained at a very constant level for accurate measurements, the same system is used for controlling the temperature of the suspension in the main loop.

Added to this, for the sake of safety, convenience, experimental purposes or maintenance, are a number of secondary systems, such as those for fuel concentration adjustment, dumping, reflector cooling, leak detection and prevention and freezing of lines to be blocked.

The whole reactor system is contained in a number of isolated steel tanks, embedded in concrete as a biological shield. These leak tight tanks are at an underpressure with respect to the reactor hall. The oxygen is removed from the containment before operation because pressurization of the suspension is effected with hydrogen. This enables a check on the air leakage into the compartments to be made at any time. Control measurements are performed by oxygen detection. The containments are connected with suitable systems to maintain an underpressure in the compartments and to remove radioactive gases.

Although the reactor is relatively small, the system is considerably more complicated than a power reactor of this type ever would be. First, certain provisions had to be included because the operational behaviour of such reactors is still unknown. Then , a high flexibility both in the mode of operation and in a remodification had to be specified in the design.

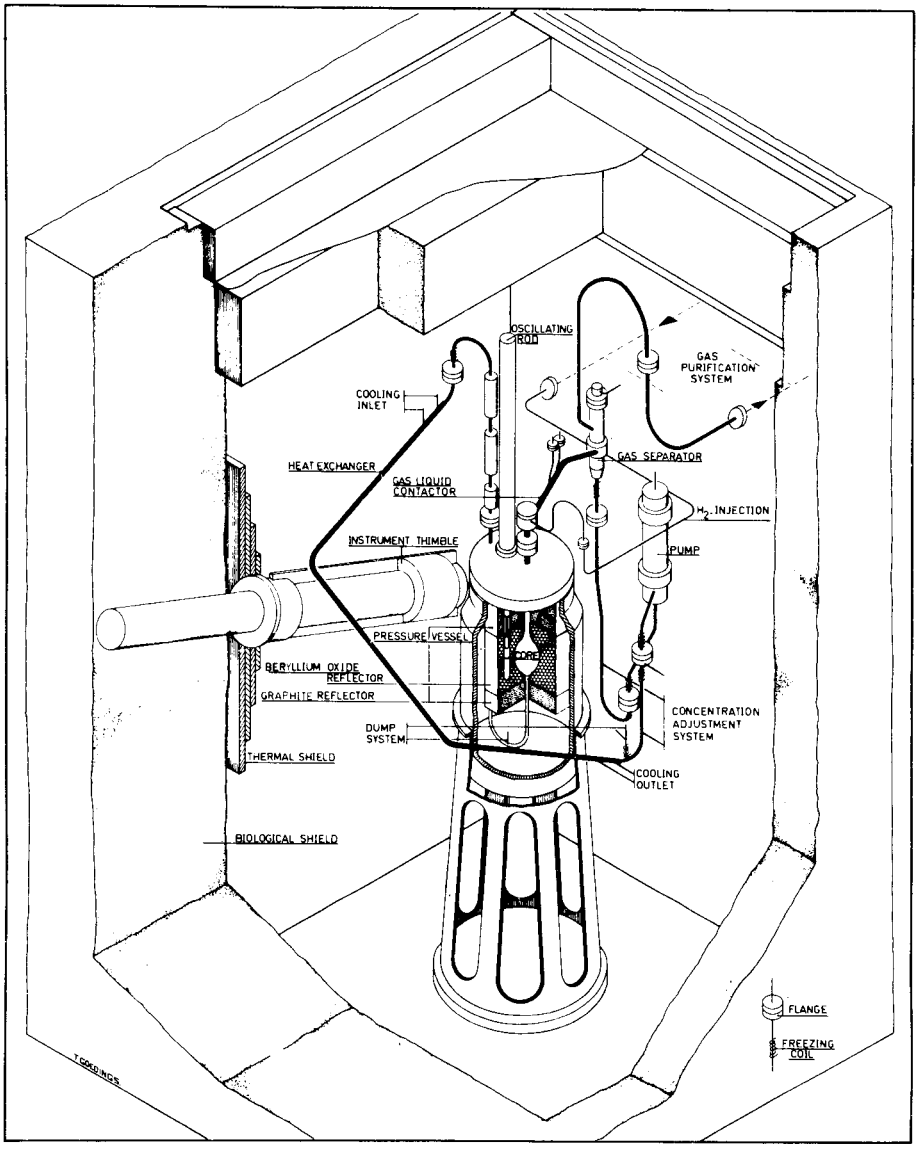

FIG. 6. Artist's view of KSTR main system.

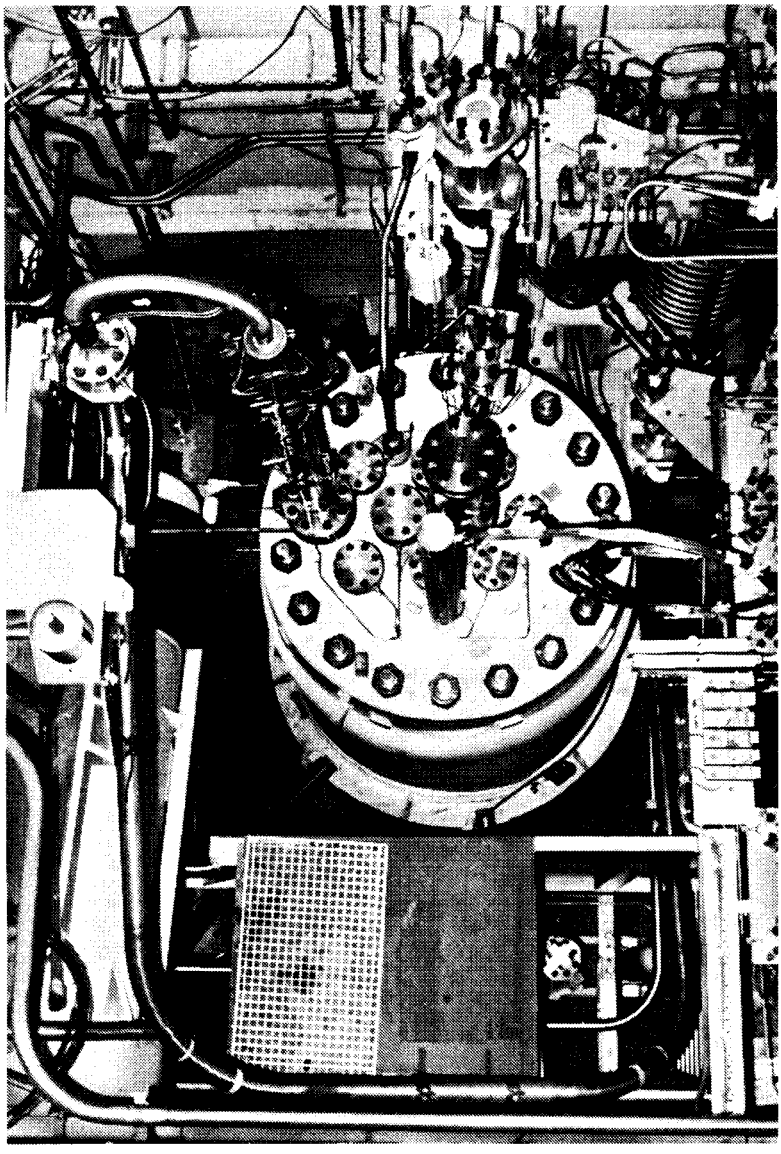

FIG. 7. Status of construction of the KSTR main system on 1 April 1971.

Account has been taken of the fact that different types of fuel with widely differing rheological properties could be used for evaluation in the event that such an extension of the program became desirable. Finally, the reactor has been instrumented abundantly to extract a maximum of information in the shortest possible time. As stated above, the facility has been completed and will soon start its performance tests as a whole. The individual systems have already been tested separately (Fig. 7).

A few remarks could be made here on the safety aspects. Power excursions can be avoided in the suspension reactor because of its prompt negative temperature coefficient. On the other hand, as with any LFR, it requires certain special measures because its fuel is a fluid which also circulates outside the reactor core. It may therefore spread readily from the system once the integrity of its containment has been violated. Therefore, very high leaktightness standards of such a system must be guaranteed. A complete, multiple containment is required.

5. RESEARCH WITH THE KSTR

5.1. Operation

After a period of performance testing of the integral reactor system with hot water and with a natural fuel suspension, the information to be derived from the system will be collected in two operational stages.

5.1.1. Sub-critical operation

In this stage the basic neutron physical data of the system will be collected. At the same time, the technological information derived from the previous performance tests can be further refined.

In contrast to the usual practice in sub-critical experiments, operation of the KSTR is envisaged at high source multiplication factors, very close to the point of criticality. This ensures high sensitivity of the measurements

A number of physical data, such as the concentration coefficient and the temperature coefficient, can be determined without the complication of radiolysis or temperature gradients which would occur under power conditions.

In general, the sub-critical phase of operation will show such characteristics of the system as the heat balance, the unperturbed dynamic behaviour of the reactor and its stability. With respect to the stability it must be borne in mind that, under these conditions, the system is still most sensitive towards reactivity changes which are not damped by a temperature feedback mechanism.

5.1.2. Power operation

Apart from collecting operational experience, the main objectives of this phase of operation are: the determination of the degree of internal stability of the reactor, of the rate of radiolysis and of the efficiency of the Xe purification. Compared with the preceding operational phase, additional phenomena present themselves which contribute largely to the stability of the system. The prompt negative temperature coefficient and

the void coefficient initiated by radiolytic gas present in the core may strongly influence the dynamic stability. It is expected that a power reactor of the same type could be operated on its prompt negative temperature coefficient at a constant fuel concentration, i.e. without control rods. Temperature variations would then take care of power variations. As this feature would be very attractive technically, it must be well established during power operation.

5.2. Methods of measurement

The required information will be obtained mainly by physical measurements which do not interfere with the flowing suspension. Temperature, flow velocity, pressure and flux indications are used for surveying the system technologically, whereas the specific reactor properties are derived from flux, temperature, flow velocity and concentration measurements. Stability studies will be made by analysing the reactor noise both with and without reactivity disturbances. An oscillating poison rod is available in the reflector for this purpose.

It is to be expected that certain phenomena will be observed in the reactor which require further technological investigation. For this purpose, the reactor experiments can be supported by extensive studies in a number of the present high-temperature loops. When an interaction of the hydrodynamic with the nuclear behaviour of the system can be studied at ambient temperature, a low temperature zero energy facility with circulating suspension is available.

5.3. Fuels

In specifying the design of the reactor, provision was made for the installation to be used with widely differing suspensions ranging from 5-μm fuel spheres in water to colloidal systems, although the system needs some modification, especially for parts which control the fuel concentration. The first fuel to be studied is a 5-μm fuel, the concentration of which can, in principle, be controlled rapidly with hydrocyclones. Depending on the results of further irradiations, fuels with smaller fuel particles might be introduced. Technological experience has shown the applicability of such fuels.

6. SUMMARY

In the view of the Netherlands electricity supply undertakings, a liquid fuel reactor operating on the ^{232}Th-^{233}U cycle could contribute largely to the solution of nuclear power production in the decades to come. A promising variant could be a suspension reactor. A critical 1-MW(th) facility has been developed, designed and constructed and will start operation shortly. The problems encountered so far in connection with this type of facility have been solved by elaborate and careful studies to the extent that this could be done without the actual KSTR. The KSTR will have to show whether a suspension power reactor is feasible and what problems still have to be solved for further development.

REFERENCES

[1] ORGANISATION FOR ECONOMIC CO-OPERATION AND DEVELOPMENT, Illustrative Power Reactor Programmes, ENEA Rep. (1968).

[2] WENT, J.J., WIECHERS, W.K., "The impact of fuel cycle economics on the future development of nuclear power", To be published in Advances in Nuclear Science and Technology 6, Academic Press, New York.

[3] ORGANISATION FOR ECONOMIC CO-OPERATION AND DEVELOPMENT – INTERNATIONAL ATOMIC ENERGY AGENCY, Uranium: Resources, Production and Demand, Joint ENEA-IAEA Rep. (1970).

[4] KREYGER, P.J., et al., UN Int. Conf. peaceful Uses atom. Energy (Proc. Conf. Geneva, 1958) 9, UN, Geneva (1958) 427.

[5] WENT, J.J., 3rd UN Int. Conf. peaceful Uses atom. Energy (Proc. Conf. Geneva, 1964) 11, UN, New York (1965) 266.

COMPARISON OF URANIUM FUEL CYCLES USING HIGH AND LOW ENRICHMENT IN HIGH-TEMPERATURE REACTORS

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Abstract–Résumé–Аннотация–Resumen

COMPARISON OF URANIUM FUEL CYCLES USING HIGH AND LOW ENRICHMENT IN HIGH-TEMPERATURE REACTORS.

The question of whether the thorium or the uranium fuel cycle is more favourable for a high-temperature reactor is of vital importance to further fuel-element development. The different approaches to this problem become obvious when it is realized that, in the United States and in the Federal Republic of Germany, the cycle with highly enriched uranium has been investigated by preference, whereas, in the United Kingdom, present reactor studies are based on the cycle with uranium of low enrichment. In this paper, the pertinent technological and economic problems are discussed. For a 600-MW(e) high-temperature reactor, the thorium fuel cycle with reprocessing of the breed elements and recycling of the bred fissile material, as well as the uranium cycle with reprocessing of the fuel and sale of the spent uranium and the bred plutonium, were optimized with regard to fuel cycle cost. In selecting the reference cycles, special emphasis was placed on a comparable status of fuel element development. Hence the design data were chosen so that the constraints regarding fast dose, burn-up, heavy metal density of the fuel matrix, maximum fuel and wall temperatures, and maximum power output per fuel element are not exceeded. For both reference designs, the costs of the outer fuel cycle for fabrication and reprocessing at an installed capacity of 5000 MW(e) were determined. Due to the influence of the specific costs on the fuel cycle parameters, a re-optimization proved to be necessary. It is shown that the thorium cycle leads ultimately to lower fuel cycle costs whereas the uranium cycle could be introduced with less development effort.

COMPARAISON DE CYCLES DE COMBUSTIBLE A URANIUM FORTEMENT ENRICHI ET A URANIUM FAIBLEMENT ENRICHI POUR LES REACTEURS A HAUTE TEMPERATURE.

La question de savoir quel est, de l'uranium ou du thorium, le combustible le plus approprié pour un réacteur à haute température a une importance capitale pour la réalisation d'éléments combustibles. Les différentes manières d'aborder ce problème apparaissent lorsqu'on constate qu'aux Etats-Unis et en République fédérale d'Allemagne on a étudié de préférence les cycles à l'uranium fortement enrichi, alors qu'au Royaume-Uni, les études actuelles sont fondées sur le cycle à l'uranium faiblement enrichi. Les auteurs étudient les aspects technologiques et économiques du problème. Pour le réacteur à haute température de 600 MW(e), le cycle au thorium avec retraitement des éléments surgénérateurs et recyclage de la matière fissile surgénérée, ainsi que le cycle à l'uranium avec retraitement du combustible et vente de l'uranium épuisé et du plutonium surgénéré sont optimisés par rapport au coût du cycle du combustible. Pour comparer les cycles les auteurs se sont efforcés, en particulier, de choisir ceux où la technologie de l'élément combustible était analogue. Par conséquent, ils ont choisi les données de l'étude de manière à respecter les limitations relatives à la dose de neutrons rapides, au taux de combustion, à la densité du métal lourd de la matrice de combustible, aux températures maximales du combustible et de la paroi et à la production maximale d'énergie par élément combustible. Pour les deux études de référence, ils ont déterminé les coûts du cycle de combustible extérieur pour la fabrication et le retraitement, correspondant à une capacité installée de 5000 MW(e). En raison de l'influence des coûts spécifiques sur les paramètres du cycle du combustible, il a fallu procéder à une réoptimisation. Les auteurs montrent que le thorium permet en définitive d'obtenir des coûts de cycle de combustible moins élevés, tandis que le combustible à l'uranium exige moins de travaux de mise au point.

СРАВНЕНИЕ УРАНОВЫХ ТОПЛИВНЫХ ЦИКЛОВ С ИСПОЛЬЗОВАНИЕМ ВЫСОКООБОГАЩЕННОГО И МАЛООБОГАЩЕННОГО УРАНА В ВЫСОКОТЕМПЕРАТУРНЫХ РЕАКТОРАХ.

Вопрос о том, какой топливный цикл, ториевый или урановый, больше всего подходит для высокотемпературного реактора, имеет жизненно важное значение для дальнейшей разработки тепловыделяющих элементов. Различный подход к этой проблеме подтверждается тем фактом, что в Соединенных Штатах Америки и Федеративной Республике Германии предпочтение отдавалось исследованиям топливного цикла с высокообогащенным ураном, тогда как в Англии современные реакторные исследования основываются на цикле с малообогащенным ураном. В представленном докладе рассматриваются соответствующие технологические и экономические проблемы. Для высокотемпературного реактора мощностью 600 МВт(эл) ториевый топливный цикл с регенерацией воспроизводящих элементов и повторным использованием воспроизведенного делящегося материала, а также урановый цикл с регенерацией топлива и продажей отработанного урана и воспроизведенного плутония оптимизировались в отношении стоимости топливного цикла. При выборе базисного цикла особое внимание уделялось сравнимому статусу разработки топливных элементов. Поэтому данные конструкции выбирались таким образом, чтобы не было чрезмерным требование в отношении дозы быстрых нейтронов, глубины выгорания, плотности тяжелых металлов топливной матрицы, максимальных температур топлива и стенок, а также максимального выхода энергии в расчете на топливный элемент. Для обоих базисных конструкций была определена стоимость внешнего топливного цикла для производства и регенерации топлива при установленной мощности в 5000 МВт(эл). Из-за влияния специфических расходов на параметры топливного цикла потребовалось провести реоптимизацию. Показано, что ториевый цикл ведет в конечном счете к более низкой стоимости топливного цикла, зато урановый цикл можно ввести при меньших усилиях по его разработке.

COMPARACION ENTRE CICLOS DE COMBUSTIBLE DE URANIO MUY ENRIQUECIDO Y POCO ENRIQUECIDO EN REACTORES DE ALTA TEMPERATURA.

Para el desarrollo ulterior de los elementos combustibles es de importancia capital averiguar cuál de los dos ciclos del combustible es el más favorable para los reactores de alta temperatura, si el de torio o el de uranio. Las diferentes maneras de enfocar este problema quedan de manifiesto en el hecho de que los Estados Unidos y la República Federal de Alemania han investigado con preferencia el ciclo de uranio muy enriquecido mientras que el Reino Unido basa el estudio actual de los reactores en el ciclo de uranio poco enriquecido. La memoria discute los problemas tecnológicos y económicos pertinentes. En un reactor de alta temperatura de 600 MW(e) se optimizaron, respecto del costo, el ciclo del combustible de torio con reelaboración de los elementos generadores y reciclado de los materiales fisionables generados, y el ciclo del uranio con reelaboración del combustible y venta del uranio agotado y del plutonio generado. Al seleccionar los ciclos de referencia se insistió especialmenre en el estado comparable del desarrollo de los elementos combustibles. Por tanto, los datos de diseño se escogieron de modo que no se sobrepasaran los requisitos referentes a dosis rápida, grado de quemado, densidad de metales pesados de la matriz del combustible, temperaturas máximas del combustible y de la pared, y máxima potencia generada por elemento combustible. Para los dos diseños de referencia se determinaron los costos del ciclo del combustible exterior para la fabricación y reelaboración en una instalación de 5 000 MW(e). Debido a la influencia de los costos específicos sobre los parámetros del ciclo del combustible, se demostró que era necesario volver a efectuar una optimización. La memoria indica que el ciclo del torio origina finalmente un abaratamiento del ciclo del combustible, mientras que el ciclo de uranio se puede implantar con menor esfuerzo de desarrollo.

1. INTRODUCTION

The decision to use either the thorium or the uranium fuel cycle for high-temperature reactors is of vital importance to further fuel-element development. The different approaches to this question are demonstrated by the fact that, in the United States and in the Federal Republic of Germany, preference has been given to the cycle with highly enriched uranium, whereas in Great Britain, current reactor studies are based on the cycle with uranium of low enrichment. In this paper, the pertinent technological and economic problems are discussed.

2. BASIS FOR COMPARISON

A 600 MW(e) pebble-bed reactor was used as the basis for the comparative evaluation of the low and highly enriched uranium fuel cycles. The design parameters – to the extent needed for the purpose of this paper – are given in Table I. They are in good agreement with known data from other HTR concepts throughout the world.

The core power density is the only parameter mentioned in Table I which could influence the comparison. In fact, the power density for the optimum fuel costs is different for the two cycles. More detailed calculations, however, have shown that, regarding the total generating costs, both the low and the highly enriched uranium fuel cycles are optimal at a core power density of about 8 W/cm^3. In view of the importance of the direct capital costs, and to favour a simplified analysis, a comparison based on this latter value is justified.

The core of the pebble-bed high temperature reactor is composed of 1.04 million "pebbles" of 60-mm diameter. The fuel elements are semi-hydrostatically pressed from graphite powder and binder. Their central region of 50-mm diameter consists of coated particles in a graphite matrix. The same type of fuel element is used in the AVR in Jülich and will be used in the 300 MW(e) prototype under construction at Schmehausen in the Federal Republic of Germany.

A once-through highly enriched uranium fuel cycle was chosen for the prototype reactor. For the commercial reactors of the future, a more elaborate fuel management is desirable. Alternative ways of combining the fissile isotopes ^{233}U, ^{235}U, ^{239}Pu and ^{241}Pu with the main fertile isotopes ^{232}Th and ^{238}U are discussed in the next section.

3. CHOICE OF FUEL MANAGEMENT

Many different fuel management schemes have been discussed in recent years [1, 2]. With highly enriched uranium, the feed-breed system appears to be particularly well-suited for the pebble-bed reactor, since different residence times for the two fuel element types can be easily arranged. The breed pebbles are reprocessed and the fissile isotopes, mainly ^{233}U, are

TABLE I. DESIGN PARAMETERS OF THE CORE

Thermal power	1540 MW
Average power density	8 W/cm^3
Diameter	700 cm
Height	500 cm
Gas inlet temperature	282°C
Mixed gas outlet temperature	775°C
Helium pressure	50 atm.
Radial/axial form factor	1.25/1.26

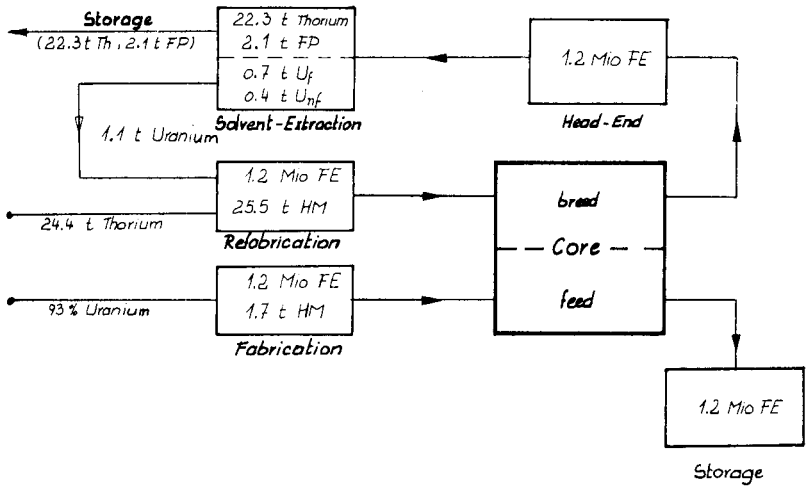

FIG. 1. Flow diagram for highly enriched uranium cycle (annual throughput for 5000 MW(e)).

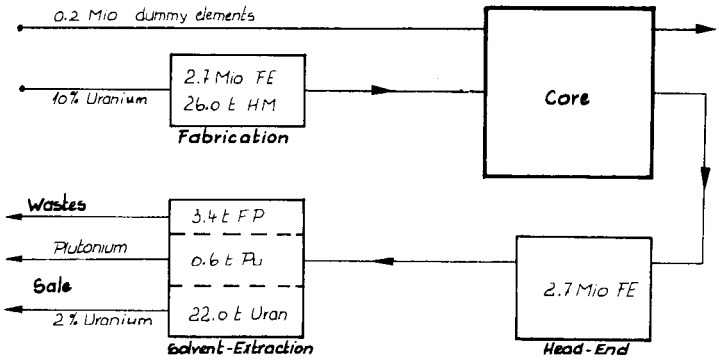

FIG. 2. Flow diagram for low-enriched uranium cycle (annual throughput for 5000 MW(e)).

recycled in the same type of fuel element. The conversion ratio is chosen slightly above unity to balance refabrication losses. In this way a closed cycle is obtained for the fissile isotopes of the breed elements. The feed elements maintain the criticality of the reactor. They contain highly enriched uranium and will not be reprocessed. A flow diagram is given in Fig. 1.

Such a procedure may seem to be wasteful, but, by recycling the feed elements, a considerable amount of ^{236}U would be built up, thereby impairing the overall neutron balance of the core. In fact, detailed calculations [3] have shown that the required ^{235}U make-up is higher if the feed elements are recycled.

For the low-enriched uranium fuel cycle, a heterogeneous fuel arrangement is desirable to lower the feed enrichment of the uranium. In the case of a pebble-bed reactor with random distribution of fuel elements, the

necessary degree of heterogeneity can be achieved by mixing the fuel elements with "dummy" graphite elements in a statistical manner. Due to the absence of structural material in the core, a high burn-up can be achieved in a HTR; thus most of the bred plutonium is burnt in the core. Fuel management calculations, however, have shown that it would be advantageous to reprocess the fuel; besides other considerations, the cost of heavy metal storage is avoided. The calculations presented in this paper were therefore based on reprocessing of the irradiated fuel elements and sale of recovered uranium and plutonium. A flow diagram is given in Fig. 2.

4. ASSUMPTIONS OF FUEL DEVELOPMENT

For a valid comparison of the two fuel cycles, special emphasis had to be placed on the comparable status of fuel element development. The design data and the limiting values of the fuel were therefore chosen accordingly.

The design parameters of the fuel are given in Table II. They differ for the feed and breed elements of the thorium cycle, as well as for the fuel element of the uranium cycle, due to the different performances of these types of element in the core.

The coated particle of the feed fuel is rather small so that its integrity up to the very high burn-up to which it is exposed is ensured. For the same reason, its maximum permissible fast dose has been reduced. On the other hand, the nominal fuel and surface temperatures of the element could be higher than for the other two fuel element types, because of the rapidly decreasing power output with time – there is hardly any conversion.

The power output of the breed element remains constant with time so that, even at the end of its service life, it has the same temperature as at the beginning of service. Since radiation-induced damage and diffusion

TABLE II. DESIGN PARAMETERS OF THE FUEL ELEMENT (PEBBLE OF 60-MM DIAMETER, 50-MM FUELLED)

	Fuel cycle / Parameters	U - Th		U - U
		Feed	Breed	
	Diameter of oxide kernel (μm)	250	600	600
	Coating thickness (μm)	165	185	185
	Conductivity of fuel element at 1000°C (kcal/mh°C)	25	20	20
Maximum	Surface temperature (nominal) (°C)	1050	1000	1000
	Fuel temperature (nominal) (°C)	1250	1100	1150
	Heavy metal density (g/cm^3)	0.08	0.38	0.38
	Fuel element output (W/cm^2)	50	27	50
	Fast dose (10^{21} nvt)	6.0	8.5	8.5
	Burn-up (GWd/tHM)	700	120	120

of fission products are strongly temperature-dependent, it is necessary to relieve the strain on the coated particles by reducing the maximum permissible fuel temperature and power output more than for the other types of fuel element.

The fuel temperature limits given in Table II are nominal; they are relatively low, as the use of SiC coatings was not considered.

It is felt that these design data represent a balanced view on what can be achieved in terms of fuel development for the uranium and thorium cycles in the Federal Republic of Germany in the next few years.

5. COST ASSUMPTIONS

In addition to the design data and technological limits of the three types of fuel element, the specific costs of the outer fuel cycle had to be determined. The more general data are summarized in Table III. They are generally accepted in fuel cycle investigations and will not be discussed further here.

The more specific costs of the HTR fuel cycle are given in Table IV. They are based on an installed capacity of 5000 MW(e) at an average load factor of 70%. It is shown later that this corresponds to an annual throughput of about 2.5 million pebbles and about 26 tons of heavy metal for either of the two fuel cycles.

The data were partly based on a detailed break-down of the outer fuel cycle costs of the 300 MW(e) THTR prototype with a throughput of 0.17 million pebbles annually. Although some degree of uncertainty is involved in such a procedure, it is assumed that any error which might arise will affect the costs of both cycles to about the same extent and thus will not distort the attempted comparison. In any case, a larger error would result by choosing too small a market size for this reactor type, since the thorium cycle would be penalized more than the uranium cycle owing to its more expensive solvent extraction and hot fabrication stages.

The "coated particle" costs (Table IV) include fabrication, testing, control of the coated particles, and all raw material except heavy metals. Since the low-enriched uranium particle is very similar to the THTR particle, its costs could be extrapolated directly. Higher specific costs will

TABLE III. FINANCIAL GROUND RULES

Uranium ore cost	\$7.0/lb U_3O_8
Conversion $U_3O_8 \rightarrow UF_6$	\$2.4/kg U
Separation cost	\$28.7/kg SW
Thorium cost	\$16.7/kg ^{232}Th
Shipping, conversion of UF_6	\$230/kg ^{235}U
Load factor	70%
Net thermal efficiency	41%
Interest/tax rate	7.0/1.9%
Amortization period	17 yr

TABLE IV. COST PARAMETERS FOR FUEL CYCLE ANALYSIS FOR AN ASSUMED INSTALLED CAPACITY OF 5000 MW(e)

Fuel cycle / Cost factors	U - Th cycle		U - U
	Feed	Breed	
Coated particle ($/kg HM)	480	140	120
Fuel element fabrication ($/kg C)	11.5	13.7	11.5
Dummy elements ($/kg C)	-	-	5.0
Shipping ($/kg C)	3.0	3.0	3.0
Head end process ($/kg C)	-	4.9	4.9
Solvent extraction process ($/kg HM)	-	70	24
Storage of thorium ($/kg HM)	-	6.2	-
Storage of feed elements ($/kg C)	4.9	-	-
Plutonium value (fissile) ($/kg)	-	-	10.0
Depreciation factor of uranium	-	-	0.8
Reprocessing time (years)	-	0.96	0.96
Reprocessing loss (%)	-	1.0	1.0

arise for the breed particle, since remotely controlled fabrication is envisaged due to the ^{232}U content of the recycled uranium; estimates indicate an increase of about 20%. The coated particle of the feed element – containing only highly enriched uranium – is even more expensive to fabricate, since only small batches can be coated in the fluidized beds for reasons of criticality.

The item "fuel element fabrication" includes the costs of graphite raw material, binder, different pressing stages, and the control of the finished fuel element. The same costs arise for the low-enriched uranium and the feed element, whereas the breed element is more expensive because of the remote fabrication; again a 20% rise was assumed. The dummy graphite elements are needed in the low-enriched uranium cycle only to increase the heterogeneity of the fuel arrangement.

The costs of "shipping" allow for the expenses incurred for the storage of fuel elements on the reactor site and their transport to and from a central refabrication plant. Given as cost per pebble or per kg graphite, they are independent of the selected fuel cycle. This is true also for the "head end process", whereby the graphite is burnt and the heavy metals are made available as oxides.

The costs of the "solvent extraction process" of the thorium cycle include the disposal of wastes. A Thorex process is envisaged by which uranium, thorium and the fission products are separated from each other. The accumulated thorium is stored at the reprocessing plant for 15 years to allow for the decay of ^{228}Th. The USAEC has guaranteed to reprocess thorium fuel at $125/kg of heavy metal (HM) [4] and a similar value is known from reprocessing cost assessments of GGA [5] and the Dragon Project [6]. A break-down into the different reprocessing steps yields 40% for head end, 55% for solvent extraction and waste storage, and 5% for thorium storage.

A different situation exists for the low-enriched uranium cycle. After the head end step, the heavy metals are available as oxides and can then be handled as LWR fuel, i.e. via the Purex reagent. Therefore a known technology and, equally important, solvent extraction plants built for LWRs can be utilized for the HTR. Fuel cycle cost calculations for LWRs are currently based on reprocessing costs of $30/kg HM. Allowing 20% for mechanical head end, $24/kg HM remain for the solvent extraction step. This latter figure is equally well applicable to the HTR fuel cycle with low-enriched uranium fuel.

In comparison with the specific cost of the Purex process, the corresponding cost of the Thorex process appears to be relatively high. It must, however, be considered that the low-enriched uranium cycle will share the capacity of large reprocessing plants which will be available for LWRs in the future. Thus a lower unit cost than for the Thorex process can be achieved. In addition, the Purex process can be expected to be somewhat cheaper as it is a simpler process.

The costs of "storage" are incurred by intermediate storage of the thorium of the breed elements and final storage of the feed elements of the highly enriched uranium cycle.

The specific costs quoted are valid for the throughputs of the different refabrication steps as given in Figs 1 and 2. The throughputs in turn depend on the data of the reference fuel cycle, the choice of which is influenced by the specific fabrication costs. It was therefore necessary to perform the optimization several times until the procedures had converged. This was facilitated by the fact that, at the assumed capacity of 5000 MW(e), the specific costs vary to a small degree.

6. REFERENCE FUEL CYCLES

The reference fuel cycles for the thorium and uranium version were optimized with regard to fuel cycle costs consistent with the core data of Table I, the design parameters and technological limits of Table II, and the cost assumptions of Tables III and IV. The results are given in Table V for the reference data and in Table VI for the fuel costs.

Comparing Tables II and V, we find that the critical design parameters are as follows:

for the feed element:	fuel temperature, power output, fast dose and burn-up
for the breed element:	fast dose
for the U/U element:	fuel temperature and burn-up.

It is interesting to note in this context that the highly enriched uranium cycle in the feed-breed system has six degrees of freedom, of which one is used to satisfy the condition of a critical core and a second the requirement of a closed breed cycle. With the remaining four free parameters, five technological limits are apparently reached. In fact, however, the power output of the feed element is only very close to its limit. The large number of limits reached leaves little freedom for choosing the optimum core composition.

TABLE V. DESIGN PARAMETERS OF OPTIMIZED FUEL ELEMENTS

Parameter \ Fuel cycle	U - Th		U - U
	Feed	Breed	
Fissile material/pebble (g)	1.25	0.57	1.01
Fertile material/pebble (g)	-	20.1	8.70
Heavy metal density (g/cm^3)	-	0.32	0.15
Residence time (full power) (yr)	2.18	2.54	1.74
Volume fraction in core (%)	46.6	53.4	81.0
Age factor	3.1	1.1	1.2
Maximum surface temperature (°C)	970	940	950
Maximum fuel temperature (°C)	1250*	1070	1150*
Maximum fuel element output (W/cm^2)	50*	25	40
Maximum fast dose (10^{21} nvt)	6.0*	8.5*	5.0
Maximum burn-up (GWd/tHM)	700*	82	120*

Note: * marks where technological limits are reached.

TABLE VI. COMPARISON OF OPTIMIZED FUEL CYCLE COSTS (mills/kWh)

Cost items \ Fuel cycle	U - Th	U - U
Heavy metal	1.03	1.07
Coated particle	0.19	0.13
Fuel element	0.25	0.24
Reprocessing	0.18	0.16
Taxes	0.06	0.05
Uranium recovery	0.27	0.10
Plutonium recovery	-	0.11
Fuel cycle costs	1.44	1.44

In the case of the low-enriched uranium cycle, four degrees of freedom are at one's disposal. Here, one free parameter is taken to fulfil the criticality condition of the core and two further variables are used to meet the two critical limits. One parameter is left to optimize the fuel cycle costs.

In conclusion, the thorium cycle is at present more restricted by technological limits than the uranium cycle. The feed element is particularly critical in its lay-out, whereas with the breed element a high dilution of the

fuel kernel with the fertile isotope results from the condition of a closed cycle. As a result, burn-up, power output, and temperatures are reduced below the present technological limits.

The throughputs through the different stages of the fabrication and reprocessing plants were calculated from the fissile and fertile content of the fuel elements, their relative distribution, and their residence times (Table V). An installed capacity of 5000 MW(e) at a 70% load factor was assumed.

The fuel cycle costs of the low and highly enriched uranium cycles turn out to be the same, both cycles having costs of 1.44 mills/kWh. Nevertheless, individual cost items are different; these are broken down in Table VI.

The item "coated particle" is considerably more expensive for the U/Th-cycle for two reasons: first, the separation of fissile material into breed and feed particles is uneconomical from a manufacturing point of view, as it demands small batches for the feed fuel, and second, remote fabrication of the breed particles is inherently more expensive.

The item "fuel element" is nearly the same for the two cycles; the higher specific costs of the remote fabrication are balanced by the overall lower throughput of the cycle with highly enriched uranium.

With regard to "reprocessing", the larger throughput of fuel elements for the low-enriched uranium cycle – 2.7 million against 2.4 million pebbles – conceals a considerable difference in costs of the solvent extraction process. Here, about 0.02 mills/kWh must be spent in comparison with 0.06 mills/kWh for the thorium cycle. Of the remaining costs, 40% go into shipping and 60% into the head-end process. Head end for the low-enriched uranium cycle is about four times more expensive than solvent extraction. The development effort should thus be aimed primarily at reducing the unit cost of this reprocessing step.

If the total costs of the outer fuel cycle are compared, one finds that, in the thorium cycle, 0.6 mills/kWh are to be spent for fuel handling and 0.5 mills/kWh for the uranium cycle. This difference is in turn balanced by the higher fissile material costs of the low-enriched uranium cycle.

7. COMPARISON OF THE TWO FUEL CYCLES

In the preceding section, reference cases for the low and highly enriched uranium fuel cycles were evaluated. The result is to be considered under the allowances made for future fuel development and the assumptions made of the specific fabrication costs. The extent to which they may influence the conclusion is discussed in this section.

Based on the technology of the THTR fuel with a coated particle kernel of 400 μm diameter, larger kernels (600 μm diameter) must be developed for both cycles. Their performance is comparable to that of THTR particles, since the lower maximum fuel temperature and burn-up of the breed particle is balanced by its higher fast dose and constant power output. In addition, a feed particle on which high demands on temperature and burn-up are made must be developed for the thorium cycle.

With regard to the element as such, the heavy metal content of the feed element and the low-enriched uranium element is lower than for the THTR

element, so that no problems will be encountered. Some development is, however, required for the breed element, because of the higher fast dose to which it is exposed, and the higher heavy metal content – 20.7 g compared with 10.7 g for THTR.

In general, more critical limitations are reached for the thorium than for the uranium cycle. Therefore, more effort must be put into fuel development and testing of the highly enriched uranium cycle. The same is obviously true for the development of fabrication and reprocessing methods. The solvent extraction process for fuel containing thorium must be promoted to a technical scale. In addition, it seems necessary to adapt the present techniques for kernel preparation, coating, and fuel element pressing to remote fabrication.

As there is considerable uncertainty about the costs of remote fabrication, the fuel cycle was reoptimized with unit costs increased by 50%. The fuel costs increased by 0.15 mills/kWh to 1.59 mills/kWh. On the other hand, considerable uncertainty exists about the cost of reprocessing, especially the solvent extraction step. Under the optimistic assumption that the cost of the thorium solvent extraction process drops to the level estimated for the equivalent uranium process, that is, from \$70 to \$24/kg HM, 0.04 mills/kWh would be saved.

On the other hand, the problem of the uranium fuel cycle lies mainly in the necessity to sell the discharged uranium and plutonium. The value depends on the market conditions, and thus on the requirements of other reactor types, especially light-water reactors and fast breeders. It was shown in Ref. [3] that the uranium cycle would only be competitive with the thorium cycle if the irradiated fuel were stored rather than reprocessed. In view of the uncertainty of the price of irradiated fuel, the optimization was repeated with a 20% reduction of the depreciation factor for irradiated uranium – from 0.80 to 0.64 – and of the plutonium value – from \$10 to \$8/g. In this case, fuel cycle costs amount to 1.48 mills/kWh instead of 1.44 mills/kWh.

A further point worth considering is the sensitivity of fuel cycle costs to uranium ore and separation costs. Owing to the better overall neutron economy of the thorium cycle, its natural uranium requirement and demand for separative work is smaller. From the throughputs of the two reference fuel cycles, an annual consumption per MW of 160 lb U_3O_8 and 80 separative work units follows for the highly enriched uranium cycle and of 190 lb U_3O_8 and 85 separative work units for the low-enriched uranium cycle, whereby the fissile plutonium was assumed to be equivalent to ^{235}U. Correspondingly, the influence on fuel cycle costs is smaller for the thorium cycle. A cost penalty of 0.10 mills/kWh arises if the natural uranium price increases by \$2.8/lb or if the price per separative work unit is increased by \$5.6/kg. The equivalent figures for the low-enriched uranium cycle are \$2.1/lb and \$4.8/kg separative work unit, respectively.

8. SUMMARY

A comparative study on the low and highly enriched uranium fuel cycles for high-temperature reactors was carried out. The main results can be summarized as follows:

(1) The introduction of the thorium cycle on a commercial basis demands more effort because of the higher requirements for fuel – fast dose and burn-up – and of the necessity to develop a remote fabrication technique and the thorium solvent extraction process. It should, however, be considered that, in the Federal Republic of Germany, much has already been undertaken in this direction.

(2) With the assumptions made in this paper, fuel cycle costs can be expected to range from 1.44 to 1.48 mills/kWh for the uranium cycle and from 1.40 to 1.59 mills/kWh for the thorium cycle.

(3) The thorium cycle is less sensitive to changes in the uranium ore and separative work costs. Fuel costs of the thorium (uranium) cycle change by 0.1 mills/kWh if the uranium ore price is changed by \$2.8 (2.1)/lb or if the costs per unit separative work are changed by \$5.7 (4.8)/kg.

In conclusion, at the present level of both uranium ore and separation costs, the low and highly enriched uranium fuel cycles appear to be equally attractive in view of the present uncertainties. The question arises as to whether the potential of the thorium cycle, at higher costs of enriched uranium in relation to the specific refabrication costs, justifies the greater development effort.

ACKNOWLEDGEMENT

The authors wish to thank Mr. W. Müller for his assistance in providing and evaluating most of the data presented in this paper.

REFERENCES

[1] SCHRÖDER, E., BLOMSTRAND, J.H., BRUNEDER, H., Fuel cycles for power reactors, J. Br. nucl. Energ. Soc. 5 (1966) 408.

[2] SCHLÖSSER, J., SCHRÖDER, E., "Economic aspects of high temperature reactors", Nuclear Energy Costs and Economic Development (Proc. Symp. Istanbul, 1969), IAEA, Vienna (1970) 305.

[3] SCHLÖSSER, J., "Fuel cycles in high temperature reactors", Economics of Nuclear Fuels (Proc. Symp. Gottwaldov, 1968), IAEA, Vienna (1968) 399.

[4] USAEC, Announcement reported in Nucleonics Week (18 June 1970) 6.

[5] SHEFCIK, J.J., STEYER, K.G., Cost of Reprocessing HTGR Fuels in Central Plants of Various Capacities, GAMD 8458 (1967).

[6] WHARTON, J., SHORT, G.D., HTR Fuel Reprocessing and Refabrication Study, Dragon Project Rep. 540 (1968).

DISCUSSION ON AGENDA ITEM 2.8

Developments in the thorium fuel cycle

DISCUSSION ON ALL THE PAPERS IN THIS AGENDA ITEM:

P/157	*Canada*	*Presented by W.Bennett Lewis*
P/186	*Italy*	*Presented by F. Doria*
P/020	*Netherlands*	*Presented by J.J. Went*
P/389	*FRG*	

G.H.B. LOVELL: Could Mr. Lewis indicate any fuel or material problems that will require investigation in the development of the reactor system he has described?

W.Bennett LEWIS: In the full paper we mention having irradiated $Th(+U)O_2$ fuel to 50 MWd/kg of heavy element, but I am sure we would wish to carry out long-term irradiations of the exact fuel design proposed.

I am also sure that decisions on materials to be used and the techniques for maintaining the organic coolant will be taken after more test results have been obtained. When differences are small the decision sometimes takes longer. I myself would be prepared to go ahead on the basis of present information but the fact that large expenditures are involved means that many other persons will have to be satisfied.

M.R. SRINIVASAN: I would like to ask Mr. Lewis whether the organic-cooled thorium reactor could be started with an initial enrichment of plutonium instead of ^{235}U and, if so, what the economic penalty might be.

W.Bennett LEWIS: Yes, it could. The use of plutonium was compared with the use of ^{235}U with thorium fuel in report AECL-2274 of July 1965 (Ref. [16] of our paper). There is some economic penalty if ^{235}U is readily available at about US $11/g.

H.B. STEWART: Mr. Lewis, you mentioned the two different ^{233}U values in your paper. What was your basis for suggesting a figure of US $15.30? For example, did this include penalties for fabrication and ^{236}U?

W.Bennett LEWIS: The value of $15.30/g ^{233}U was that value which would result in fuel combinations No. 18 and No. 4 having equal fuelling costs, based on $11/g for ^{235}U.

The penalty for fabricating fuel with ^{233}U is negligible provided the fuel is fabricated immediately after separating the ^{233}U and new thorium is used, as proposed at the 1967 IAEA Symposium on Heavy-Water Power Reactors (Ref. [11] of our paper).

G.B. ZORZOLI: The KEMA reactor concept (KSTR), like that of the molten salt reactor (MSR), differs from other reactors in one important respect – from the point of view of safety. Of the three classical barriers (cladding, pressure vessel or tubes-container), the first is not present. In the case of MSR this is compensated for by the low operational

pressure. But for a very high-pressure system like the KEMA reactor, what are the predictable consequences of a pressure vessel failure? One might reply that pressure vessels do not fail. This is an acceptable answer in the case of light-water reactors, but in that of a suspension reactor, in the presence of a fluid with potential erosion effects, I would not take this for granted. Can Mr. Went comment on this question?

J.J. WENT: As is normal for light-water reactors, the KSTR has three containment barriers, too, but the first of these is the liquid fuel itself. Of the two other barriers, the first is pressure-tight. As the reactor vessel is more than an order of magnitude smaller than the normal light-water reactors for the same power and as, apart from the inlet and outlet, no vessel penetrations are required, it is highly improbable that one could have a major break occurring so fast that dumping does not work. Erosion in the reactor vessel does not occur due to the flow conditions. The only place where some erosion is possible is in the impeller of the circulation pump.

Furthermore, even if serious failure occurred in the pressure vessel, there would be no possibility of over-reactivity or of overheating. The fuel would leave the vessel with the water and injection of a large amount of coolant into the compartment, without radioactivity risks, would always remain possible. In this respect the solution is simpler than in light-water reactors.

R.M. NUNN: Could Mr. Went comment on the magnitude of likely maintenance problems, particularly with respect to the valves and pumps in the primary circuit?

J.J. WENT: It is in the effort to avoid such problems that we have constructed the primary system without valves. However, in secondary systems, e.g. in the gas purification system, valves are present and therefore we have decided on a layout in which we can reach each valve from above. Moreover, each valve can be separated by freeze plugs. Replacement of the pump or the pump impeller, although this is a major operation, will be carried out in the same way. Oak Ridge experience supports this approach.

AGENDA ITEM 5.1

Safeguards systems analysis and safeguards objectives

Analyse de systèmes pour les garanties et objectifs des garanties

Анализ систем гарантий и цели гарантий

Análisis de sistemas de salvaguardia y objetivos de las salvaguardias

Chairman

W. Bennett LEWIS, Canada

Scientific Secretaries

V. SHMELEV, IAEA

R. SKJÖLDEBRAND, IAEA

GERMANY, FEDERAL REPUBLIC
INVITED REVIEW PAPER
A/CONF.49/P/771

SYSTEMS ANALYSIS IN SAFEGUARDS OF NUCLEAR MATERIAL

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Abstract–Résumé–Аннотация–Resumen

SYSTEMS ANALYSIS IN SAFEGUARDS OF NUCLEAR MATERIAL.

Traditional science deals with objective nature by applying mathematical models to it. Systems analysis applies models, if possible mathematical ones, to the system composed of nature, men and their interactions including the operations of men. For that the system is defined by its objective and constraints. In that approach, operations research is a subset of systems analysis. Having implicitly or explicitly this approach in mind, only a few papers exist that deal with the design of such an overall system. A logical scheme for a systems analysis, if complete, is elaborated and explained in detail. It places in the logical perspective of the safeguards systems designer the definition of the objective, the description of the fuel cycle, the collection of information, its effect on the inspector as well as the operator and the strategies and operations of these parties. Having clarified the long-range systems analysis task by this explanation, the scheme is then used to assess the function and position of the various contributions to systems analysis so far. Attention is being given in particular to the results of the IAEA panels on systems analysis and safeguards methods and techniques as well as to the partly implicit considerations for the establishment of the now proposed safeguards system. Finally, the large number of individual and more detailed contributions are reviewed against the above-mentioned logical scheme. It becomes apparent that a few areas are well covered and understood whereas other areas are only slightly or not at all covered. It is possible to identify a few topics that cover most of the analysis work done so far. In line with this, special attention is given to MUF (material unaccounted for).

L' ANALYSE DE SYSTEMES ET LE CONTROLE DES MATIERES NUCLEAIRES.

La science traditionnelle traite les problèmes naturels en leur appliquant des modèles mathématiques. L' analyse de systèmes applique des modèles, si possible mathématiques, au système constitué par la nature, l' homme et leur interaction, et s' étend aux activités humaines. A cette fin, le système est défini par ses objectifs et ses limitations. Dans cette optique, la recherche opérationnelle est un élément subsidiaire de l' analyse de systèmes. Compte tenu de cette considération, implicite ou explicite, on constate que peu de travaux étudient la conception d' un tel système d' ensemble. En effet, pour qu' un système soit complet, il faut qu' il soit élaboré et expliqué en détail. Le mémoire présente, dans la perspective logique de la conception du système de contrôle, la définition de l' objectif, la description du cycle de combustible, le rassemblement des données, leur importance pour l' inspecteur et pour l' opérateur et la politique générale ainsi que les activités de ces parties. Après avoir ainsi expliqué les buts à long terme de l' analyse de systèmes, le mémoire utilise alors ce schéma pour évaluer le rôle et l' importance des différentes contributions qui ont été faites jusqu' à présent à l' analyse de systèmes. Il analyse notamment les résultats des groupes d' étude de l' AIEA, sur l' analyse de systèmes et les méthodes d' application des garanties, ainsi que les considérations partiellement implicites qui militent en faveur de l' établissement du système de garanties maintenant proposé. Enfin, le mémoire étudie les nombreuses contributions individuelles et plus détaillées sous l' angle du schéma logique mentionné. Il apparaît que quelques domaines sont amplement analysés et bien compris tandis que d' autres ne sont qu' insuffisamment traités, voire pas du tout. Il est possible de déterminer quelques rubriques qui recouvrent la plupart des travaux d' analyse effectués jusqu' à présent. Suivant les considérations qui viennent d' être indiquées, les différences d' inventaires font l' objet d' une attention toute particulière.

СИСТЕМЫ АНАЛИЗА ПРИ ПОСТАНОВКЕ ПОД ГАРАНТИИ ЯДЕРНЫХ МАТЕРИАЛОВ.

Традиционная наука рассматривает объективную природу путем применения к ней математических моделей. Системы анализов применяют модели, если возможно – математические модели к системе, состоящей из природы, людей и их взаимодействий, включая действия людей. Поэтому эта система определяется ее целью и связями. При таком подходе иссле-

дования действий являются вспомогательными по отношению к анализам систем. Имеется лишь несколько докладов, в которых безоговорочно или определенно учитывается такой подход и в которых рассматривается проект такой всеобщей системы. Логическая схема анализа систем, если она полная, тщательно разрабатывается и объясняется в деталях. Она дает определение цели, описание топливного цикла, сбор информации, ее влияние на инспектора и на оператора, а также на их стратегию и действия в логической перспективе составителя систем гарантий. Внеся ясность этим объяснением в задачу анализа большого разнообразия систем, эта схема используется пока для оценки функций и положения различных видов вкладов в анализы систем. В частности, внимание уделяется результатам совещания МАГАТЭ по анализам систем, а также по методам и способам гарантий, до некоторой степени – полному рассмотрению вопроса создания предложенной в настоящее время системы гарантий. Наконец, рассматривается большое число индивидуальных и более подробных вкладов в вышеуказанную логическую схему. Становится очевидным, что несколько областей хорошо охвачены и известны, тогда как другие области охвачены лишь незначительно или совсем не охвачены. Можно установить несколько тем, которые охватывают большую часть работы по анализам, проделанной до сих пор. В соответствии с этим особое внимание уделено понятию MUF (неучтенный материал).

EL ANALISIS DE SISTEMAS EN LA SALVAGUARDIA DE LOS MATERIALES NUCLEARES.

La ciencia aborda tradicionalmente los problemas de la naturaleza con ayuda de modelos matemáticos. El análisis de sistemas aplica modelos, de ser posible, de tipo matemático, al sistema integrado por la naturaleza, el hombre y las relaciones recíprocas entre ambos. Por esta razón, el sistema queda definido por sus objetivos y sus condiciones límite. En tal supuesto, la investigación operacional constituye un subconjunto del análisis de sistemas. Pocos trabajos se encuentran que, teniendo en cuenta esta hipótesis, explícita o implícitamente, se preocupen de la elaboración de un sistema global de este tipo. Para ser completo, el esquema lógico de un análisis de sistemas debe elaborarse y explicarse en detalle. Ha de contener la definición de los objetivos, la descripción del ciclo del combustible, un conjunto de datos experimentales, su efecto tanto sobre el inspector como sobre el explotador, y las estrategias y operaciones de ambas partes en la perspectiva lógica del creador de los sistemas de salvaguardias. Después de haber explicado de este modo la misión a largo plazo del análisis de sistemas, se utiliza este esquema para definir la función y la posición de las diversas aportaciones al análisis de sistemas hasta ahora conocidas. Se concede especial atención a los resultados alcanzados por los grupos de expertos convocados por el OIEA sobre el análisis de sistemas y sobre los métodos y técnicas de salvaguardia, así como a las consideraciones parcialmente implícitas relativas al establecimiento del sistema de salvaguardias que ahora se propone. Finalmente, el elevado número de distintas aportaciones elaboradas en mayor detalle, se examinan en función del esquema anterior. Resulta entonces que algunos aspectos quedan bien delimitados y comprendidos, mientras que otros sólo lo son ligeramente o casi nada. Es posible identificar las líneas principales que abarcan la mayor parte del trabajo de análisis realizado hasta ahora. En consecuencia, se prestará atención a la DI (diferencia inexplicada).

1. THE MEANING OF SYSTEMS ANALYSIS

Science in its traditional sense deals with objective nature, which does not include decision-making and action-taking man. Traditional science's approach to nature is measurement. By the use of yardsticks numbers are obtained and mathematics can be applied. In traditional science, say physics, the otherwise difficult task of quantification is accomplished by measurement. The application of mathematics, made possible by this quantification, leads to the detection of laws of nature. A more careful and conscious position is to talk of models for the description of nature. The underlying problem is recognized, but this paper is not the place to elaborate on the difficult question of the ontological meaning of mathematical models in traditional science. The systems approach, in contrast to traditional

science, embraces not only objective nature but also on purpose includes decision-making and action-taking man as well as interactions between men and particularly between man and objective nature. In that sense the area of operations research is also covered by this general systems approach [1]. Constitutive to this approach is that systems are defined by an objective. All components, objective nature, men and the interactions necessary for meeting this objective make up the system. Then it is the systems approach to develop to the largest possible extent a mathematical model for that system. Forecastings, operation aspects, optimizations, statistics in particular, and decision theory are frequent aspects of such mathematical models. The application of decision theory allows for instance to some extent the nondeterministic description of conflicts between human partners, an aspect that never occurs in traditional science.

In a more pragmatic style of description, this aspect of employing a mathematical model becomes the dominant one [2] and systems analysis there becomes common sense expressed in terms of a mathematical model. This aspect is probably too narrow.

Whatever the right answer is, it is useful to reflect on the following. Establishing and using a mathematical model accomplishes two things: it forces one to organize one's thoughts and it helps one to get specific results. In many cases of systems analysis, the explicit and complete formulation of a mathematical model has not been accomplished. But it has been possible to conceive the general structure or logical flow sheet of such a model. I understand there was once a saying in Princeton that it is better to have an approximate solution to an exact problem than to have an exact solution to an approximate problem [3]. An effort to formulate the exact and therefore right problem can be strongly supported by spelling out at least the general structure or logical flow sheet of the relevant mathematical model. After having done so, it is possible in most cases to give mathematical models for at least various sections of the system. If one is aware of the principal situation, such models, if only for parts of the overall problem, can be very helpful on a somewhat pragmatic basis.

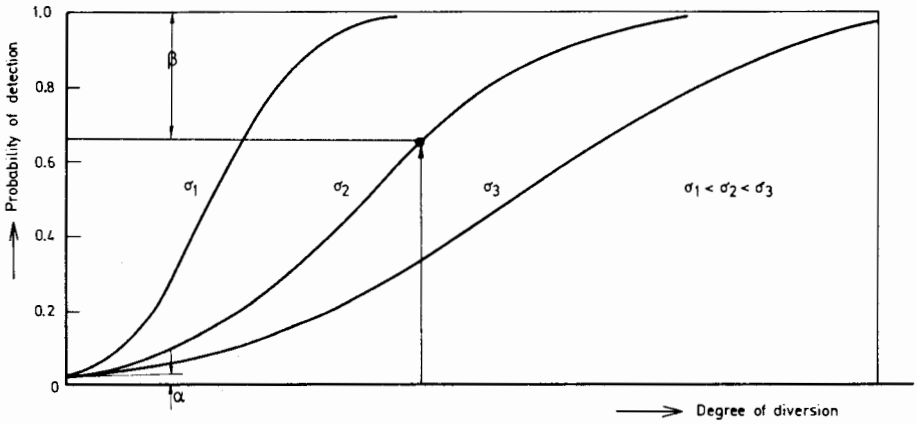
2. EARLY RECOGNITION OF INTERNATIONAL SAFEGUARDS AS A SYSTEMS PROBLEM

Systems analysis for international safeguards of nuclear material has by its very nature to take into account the boundary conditions of politics. Overall policy decisions have to be made and it is one of the tasks of the systems analyst to formulate the issues in such a way that the policy-making bodies can understand them [4]. But this goes both ways. Along these lines one has to postulate that international safeguards has to be objective, rational and formalized [5, 6].

It has to be objective in order to be universally applicable throughout political camps that oppose each other. Related to that, it has to be rational because of the size and complexity of the job. And it has to be formalized as formalization strongly helps to eliminate the open-endedness that may be otherwise inherent in an inspection process: when will the inspector be satisfied?

One realizes that the prevailing boundary conditions are different for safeguards on the international level, on the regional or national level or on the plant management level. In the past years the problem was and to some extent still is to recognize these differences and still make safeguards on the various levels mutually consistent. This paper deals with safeguards of nuclear material and by that the sum of international and regional/national safeguards is meant. Where necessary, specialities of either international or regional/national safeguards will be indicated [7].

Systems analysis for safeguards started on a somewhat broader basis around 1967. This is indicated by a number of activities, among them the following: In August 1967 the IAEA held a panel on Safeguards Techniques in Vienna [8]. The USAEC had held practically at the same time a symposium on Safeguards R & D [9]. In the United Kingdom relevant activities were given greater attention [10] and in the Federal Republic of Germany the project "Spaltstoffflusskontrolle" including the relevant systems analysis was started [5, 11]. There was one diagram, known as the power curve, that was independently produced by quite a number of groups or authors [5, 12-14]. It is given in Fig. 1.



α Probability of concluding there is diversion when in fact there has been none (type I error).

β Probability of concluding there is no diversion when in fact there has been one (type II error).

σ Inaccuracy of procedures involved.

FIG. 1. Typical power curves (based on Refs [5, 12-14]).

This diagram revealed in the early days of international systems analysis a number of important features of safeguards when errors or uncertainties, say of measurements, are implied:

(a) For a finite degree of diversion and a given inaccuracy σ of a measurement there is no 100% certainty of detection. Safeguards is of a probabilistic nature and not a go/no-go thing.

(b) The error β can be steadily decreased if the inaccuracy σ is steadily decreased. This usually requires a higher effort (cost). Therefore, cost effectiveness comes into the picture: What are reasonable costs

and what is a reasonable probability? The reasonability of the probability of detection can refer to both: the threat and the costs. Further: The level of probability by which one is satisfied must be pre-established, otherwise inspection could go on and on, the inspection would be because of β inherently open-ended.

(c) There is a finite chance of a false alarm, the error α.

In the more general context of safeguards, one is precluded by (a) from using safeguards for unlimited political objectives while applying limited resources (money, effort, accepting intrusions, etc.). By (b) it becomes necessary to clarify at least in qualitative terms what amount of resources is available for ultimately political reasons and what benefit/cost ratios are acceptable. Also, to cut the above-mentioned inherent open-endedness, the conditions beyond which the inspection or verification process is considered to be satisfactory must be predetermined. This is accomplished not only by assessing the numerical values of technical parameters but also by formally assessing methods and procedures. This illustrates the requirement for objectivity and formalization. And by (c) the aspect of objectivity and formalization again is emphasized: if there is a false alarm it must be readily and formally possible to identify and renormalize the situation.

Therefore, one is lead by contemplating Fig. 1 to consider the following type of statement as a desirable form for the result of safeguards [5, 15]: "The inspectorate is confident with the x% confidence level that the material balance is closed within y%".

This leads to the problem of statements and is thereby already an illustration of what has been more generally said on systems: by the character of this statement the acting (operating) man is already explicitly included in the analysis.

It is remarkable to what extent the features of systems analysis and thereby of the underlying reality become apparent by making use of such a simple diagram.

The safeguards system as a whole, the problem of defining the relevant model and the problem of designing and dealing with this system have been, to various degrees, the subject of a number of papers. The first paper I would like to refer to is that of Bennett, "Progress in Systems Analysis", as presented during the Karlsruhe symposium, 1970. The realm and mechanism of the safeguards system as a whole comes into the picture there although no explicit treatment of the problem is given. Specifically I would like to cite a broad definition of the safeguards system:

"The purpose of the 'system' to which we are directing our attention is to detect, and so deter by the threat of detection, the diversion of nuclear materials to non-peaceful uses. The input to this system is a combination of activities to be required and/or carried out by operating personnel, governments, or international agencies in order to detect or deter such diversion. The ultimate output of this system is 'assurance that significant diversion has not taken place'. The basic task of the systems analyst is to establish in some fashion a quantitative, or at least a logical relationship between the inputs and the outputs, and determine the interrelationship between the elements of the system in such a fashion as to enable us to characterize the effectiveness of the system for a given cost and determine those features of the system which limit its effectiveness. Implicit in this

process is the recognition of the need for new or improved techniques or procedures".

Bennett's work aims at an overall mathematical model. This could give, for instance, figures for confidence levels, benefit/cost ratios and others. As mentioned before, a full mathematical overall model is not yet really at hand today; only sections of the problem can be more systematically and formally dealt with. Therefore, methods and procedures are still a somewhat separate problem. There are two papers that deal more directly with the methods and procedures problem. One is the paper of Brown: The Design of a Safeguards Material Control System [16]. Material balance areas, accounting, reporting, surveillance, inspection, containment and material unaccounted for (MUF) are terms that are explicitly put together to make up for a logical system. The systems analysis approach is implicit but not explicit. Methods and procedures are also in the foreground of the paper of Hough and Solem [17]. They stress the definition of the measurement capability and desired limits of a material balance control, the definition of an index to quantify surveillance and containment and the development for a logic to process information and to connect it with a decision structure.

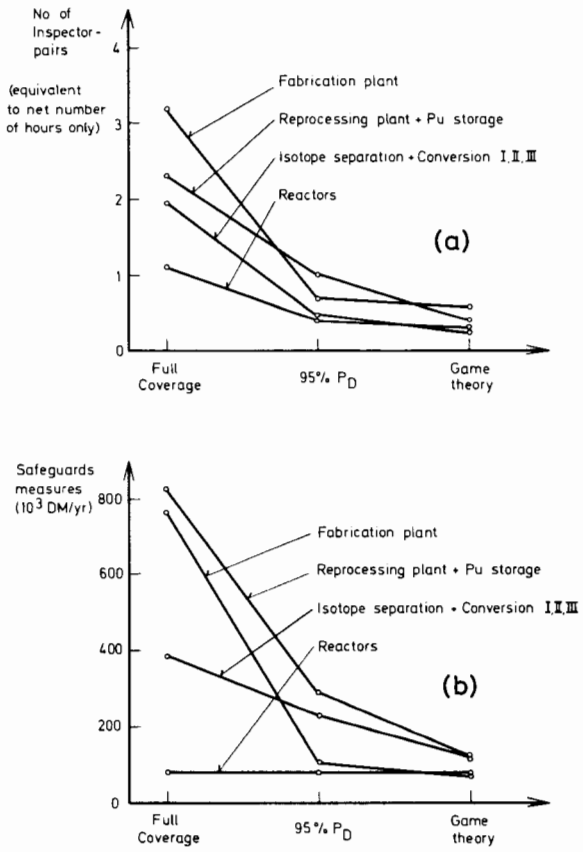


FIG. 2. Distribution of safeguards efforts among important nuclear facilities in the reference fuel cycle for different inspection intensities: (a) number of inspectors-pairs, (b) safeguards measures in 10^3 DM/yr.

Both papers should be seen against the background of existing safeguards systems, namely that of Euratom, which is based on the Treaty of Rome [18] and that of the IAEA in the pre-NPT era [19]. The above-mentioned elements of methods and procedures are there already to a large extent although the stringent systems approach is not yet fully pronounced. The whole system and a relevant mathematical model for it is the subject of a paper by Avenhaus and Gupta [20]. A whole fuel cycle is modelled, including reactors, separation plants and other facilities, safeguards measures, its efficiencies, interplays and costs are assessed and the optimization of manpower and other efforts is studied. Naturally and admittedly, the model is still somewhat simplified and reality is still more complex, but it has been possible to follow through the main safeguards questions quantitatively. One of the particularly interesting results within the constraints of the model are the curves on the distribution of safeguards efforts for various facilities and inspection intensities as given in Fig. 2. If full coverage is required, safeguarding fabrication and reprocessing plants is roughly ten times more expensive than safeguarding reactors. This factor changes rapidly to lower values, if only 95% confidence level in the inspectors statement is required and it becomes quantitatively apparent how insensitive reactors are as far as safeguards are concerned. It has been also possible to evaluate quantitatively operational aspects such as the mode of inspection and others. Methods and procedures are not in the foreground of this paper. Thus it complements the two foregoing papers [16, 17]. The papers of Oshima, Imai and others [21, 22] also consider principally the whole system but stress the aspect of interaction between the national and the international system and thereby study the introduction of formal action levels.

Together with the sequence of the very successful IAEA Panels and Technical Working Groups on Safeguards [7, 8, 23-28] which shall be considered somewhat later in this paper, these studies have more or less unanimously led to a number of important conclusions:

(a) The objective of international safeguards is the timely detection of diversion of significant quantities of nuclear material from safeguarded uses to the manufacture of nuclear explosive devices or for purposes unknown, and deterrence of such diversion by the risk of early detection [7, 16, 17].

The salient point of this definition is that the objective is the detection, not the prevention or physical protection. Another point is the word significant. By that a vast number of academic considerations on milligrams which one loses track of are discarded. This definition also has the word timely in it, and thus refers to some sort of effectiveness.

(b) The major components for building up a safeguards system are accountancy, containment and surveillance [4, 6, 16, 17, 29].

This helps to optimize, at least in principle, safeguards procedures in various cases, as one can partly substitute one for the other and there is mutual influence.

(c) The hazard which international safeguarding is to safeguard against is up to now assessed in terms of two parameters: the amount of effective

kg [30] and the critical time [31] for which a significant diversion is expected to exist without detection.

These two parameters are the first attempt to make the hazard tangible and ultimately, along with certain improvements, perhaps even quantifiable.

(d) The principal steps for safeguards methods and procedures are the following [16-19]: design review, records and reports, inspection.

3. RIGOROUS SYSTEMS ANALYSIS

Systems analysis in a more rigorous sense does not exhaust safeguards as such. General systems analysis for safeguards probably has in most cases much informed common sense and judgement in it, whereas it is the more long-range principal purpose of a rigorous systems analysis to employ mathematics. Thereby systems analysis becomes a tool, only a part of the overall approach. It is quite often of help to elaborate a logical scheme and to organize thoughts this way. In Fig. 3 such an elaborate logical scheme is presented. We shall discuss it now. (Note: round units indicate definitions or assessments, boxes indicate steps that ultimately should be part of a formal mathematical model or procedure. Further, figures in round brackets refer to the various units of the scheme.)

The key to the flow sheet is a definition of the objective (I). This is obvious already in general terms but more specifically so if we recall that systems are defined this way. The definition of the objective is principally subject to iterative improvement. This is what has happened already in the past. The definition that has been given above was an improvement of an earlier, less specific definition [32] and has since been slightly improved [30]. With the objective in mind the systems designer conceives a set of methods and procedures (II). For that he has to describe the reality of the fuel cycle (1). Data collection on the fuel cycle and its facilities, together with forecasts on future developments, are an essential part of the task of systems analysis. It refers to objective fuel-cycle reality. The systems designer needs further to take into account a number of restraints and conditions (III): non-intrusiveness with respect to the operation of the facility, a given amount of effort or money to spend for safeguards, possibilities to influence the design of containments, etc. A major input of information to the systems designer comes from integral experiments, that is, safeguards exercises (2). Objective nature is involved there, for instance, unknown operative losses in processing facilities can be experienced this way. But also man is included. Did the inspectors do their job well? Were there frictions with the operators or was there lack of cooperation on the part of the operators?

Having established in the first order (iterative step) such a set of methods and procedures the systems designer reflects on their application. Having the objective reality in mind he evaluates measurements and its statistics (3). For instance, in a mathematical model, the objective flow of the nuclear material could be described by differential and integral equations. Measurement and its statistics can be represented by probability distributions for the outcome of the measurements and random numbers to simulate an actual measurement. Or in short, in (3) the whole area of

accountancy is represented. The result is information. As outlined before, accounting can be complemented by surveillance. So the information obtained by surveillance is to be described in a model in (4). This has problems in it but should be possible [33], but further work on this is necessary. Information is also produced by reporting (and recording). This must be described in (5) using the same terms as in (3) and (4), resulting in a comprehensive and unified representation of this information (6). Related to the representation of accountancy, surveillance and reporting as parts of a system is their respective development (7), (8), (9). Being related to the system means that the targets for these developments are derived from (3), (4) and (5) and, the other way around: possible developments help to complete the system.

Information becomes communication if the receiver and/or the sender is taken into account. The information described in (6) is of different operational meaning for the inspector and the operator of the plant. This transformation from information to communication is to be described in (10). This communication is among other things the input for the inspector in setting up his strategy for detecting a diversion (IV). Modes of diversion are to be considered one input to strategy (II). Other components are: his reflection (or communication value) on the rules of procedures (12), the available budget (13), an assessment on what his opponent, the operator, thinks (V) and the fact that he is to make a statement (14). This leads to an assessment by the systems designer as to what his inspector might feel to be the best strategy (IV).

Similarly, the transformation of information into communication of the operator is described in (15), his additional inputs are: his reflection (or communication value) on the rules of procedures (16), modes of diversion (11), the penalty for diversion (17), his incentives for a diversion (18), and his assessment on what his opponent, the inspector, thinks (VI). This leads to an assessment by the systems designer as to what his operator might feel the best strategy to be (VII). For completeness: the cost of safeguards (13) also influences the thinking of the operator and the penalty for diversion (17) that of the inspector.

Now the systems designer executes the model: his operator uses the chosen strategy of diversion which his inspector does not know (19) and in turn his inspector, to make a statement, uses the chosen strategy for detecting the diversion which the operator does or does not know (20). The chosen rules for methods and procedures are applied by evaluating the probability of detection and the involved cost efficiency and benefit-cost ratios (21). This is judged against the threat of diversion (VIII). If the result is satisfactory, the system is ready for implementation, if not, the rules and possibly the objective or the restraints and conditions must be adjusted (22). In any event, however, this evaluation (21) makes it possible to assess eventually figures in the definition of the objective (I) if so required. If the system is implemented, experience will also tell whether the system is satisfactory (23); a further iteration is then possible.

Let us come back to the possible modes of diversion (11). These will be strongly influenced by the existence and type of containments. Therefore, the function of the containment is to be a conduit for action in the same way as it is a conduit for the actual flow of nuclear material. The conduit function is described in (24). Note: surveillance refers among other things also to the conditions of containment. For instance: are all seals unbroken? But

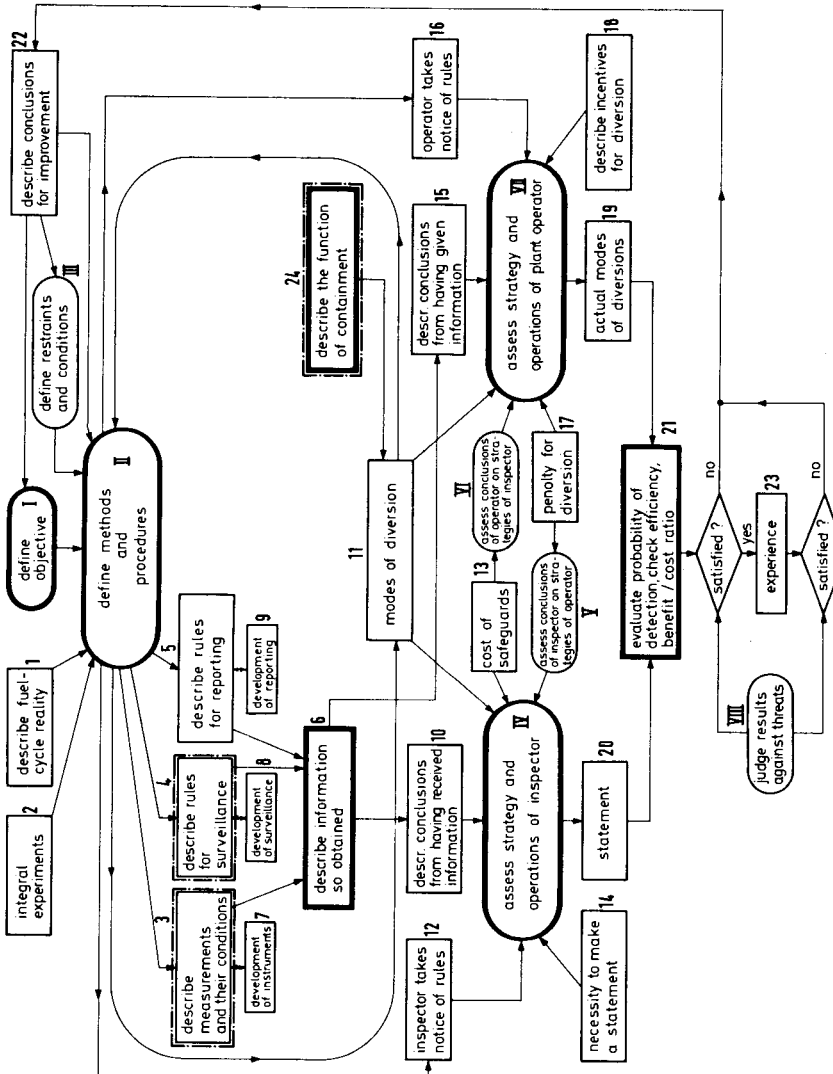


FIG. 3. Structure of the model for the design of methods and procedures for safeguards from a systems point of view.

this results in information. It is worthwhile to note that besides this information component there is the conduit component of the function of the containment. It influences not information but possible actions or, more clearly, operations, by influencing the assessment of the respective strategies. The modes of diversions feed back to the definition of methods and procedures (II) and vice versa. The combination on the one hand of information of the conduit function of the containment for possible actions and on the other hand of the assessment of strategies by men, namely the operator and the inspector, is an aspect of decision theory. It is fully in line with the systems approach, described in the beginning of the paper, that embraces objective nature and men. In this context it is natural that information has to be transformed into communication to become operational.

It is worthwhile to stop here for a second and to realize that in most past approaches, information has been considered a part of an objective picture. But still it was necessary to argue about the use of information, actions, etc. So this is an indication for the operational component of the system, that should have been more fully taken into account. Descriptions of possible operations are part of the systems approach presented here. Along these lines, the above-mentioned claim for formalization of safeguards procedures and in particular for a formal statement from the inspector as the result of the inspection activities becomes natural. It is believed that the systems approach outlined in this paper, together with some terms taken from decision theory, describe the system more smoothly. One further remark: the necessity or at least the desire has been expressed quite often to quantify the function not only of accountability but also of surveillance and containment [5, 17]. Accountability leads directly into figures, surveillance would lead to figures only with some difficulty [29]. The quantification of the containment function seemed to be exceedingly difficult [34]. By feeding the containment function into the models of diversion and letting them influence the respective strategies, which in turn ultimately result in a quantitative evaluation of safeguards efficiency, it may be possible to quantify the conduit function of containment in a backward fashion.

This ends the description and discussion of the logical flow sheet for the analysis and design of the system. Note: for the design of the system and its model, it must be assumed that the operator tries to have a diversion. By that no judgements on real cases are implied.

It is against this background that special attention should be given to the two IAEA Panels on Systems Analysis, one in Vienna, August 1969 [26] and the other in Tokyo, December 1969 [7]. Among other things they resulted in a list of tasks (1 through 5) for a thorough systems analysis. Task 1 is the description of nuclear materials system, it refers to (1) of the logical scheme; task 2 is the definition of safeguards objectives, concepts and criteria, it refers to (I), (II), and (III); task 3 is the requirement and use of safeguards information and refers to (6) as well as to (15) and (17); task 4 is on safeguards methods and effectiveness, it refers to (II), (13), (20), (24), and especially (21); task 5 is the evaluation of integral experiments, it refers to (2). Also many if not most of the subtasks can be found in this logical scheme. Generally, these two IAEA Panels on Systems Analysis, together with the other IAEA Panels and Technical Working Groups on Safeguards [7, 8, 23-28] and also the work of the Consultants to the Director General of the IAEA [31, 35, 36], reflect the process by which the thinking on safeguards became more explicit step by step and by which the systems approach

became more and more predominant in spite of the fact that the explicit application of mathematical models even for sub-aspects has been limited. The setting up of more and more mathematical models and the resultant possible detailed optimization in quantitative terms remains to be done. However, as mentioned before, the paper of Avenhaus and Gupta [20] can be considered to be a step in this direction.

4. SPECIAL CONTRIBUTIONS

Now, in the context of systems analysis, a number of special contributions will be reviewed. These special contributions deal with detailed aspects of systems analysis which also are important and highly necessary. Against the background of the overall diagram it becomes apparent what the interactions of the following subjects are:

(a) Fuel cycle and facility data, forecasts
(b) Integral experiments
(c) Accountability studies
(d) Correlations
(e) Material unaccounted for (MUF)
(f) Simulation
(g) Decision theory
(h) Cost effectiveness
(i) Design criteria for future plants.

These subjects shall now be reviewed.

(a) Fuel cycle and facility data, forecasts

The data of facilities that are relevant for safeguards were identified during the IAEA Working Group on Design Information [27]. They are relevant for the flow of nuclear material through these facilities. A realistic fuel-cycle model, its time evolution, source data for the involved facilities, the distribution of the nuclear materials in a fuel cycle and in particular the computation of staff requirements and staff deployment has been evaluated by Shmelev [37]. A very remarkable result of that study is that the expected cost of safeguards is well below 1% of the cost of electricity. There the safeguards costs are characterized by the figure of $150 per year and MW(e). In a similar paper Shmelev again evaluates the distribution of nuclear material in a realistic fuel cycle and the forecast for the fuel cycle [38]. A salient feature of the paper is the result that, in terms of effective kilograms, the share of reactors in the overall fuel-cycle inventory is comparatively small; the major share is with reprocessing plants while, in terms of straightforward linear fissile material content, the share of reactors is of course the largest.

(b) Integral experiments

Integral experiments are safeguards exercises in an existing nuclear facility for a certain production campaign. The principal objective is to close for these actual campaigns the material balance, to evaluate the measurement errors, the operating losses, the safeguards effort and to test

instruments and procedures. In Europe there have been seven integral experiments altogether:

The ALKEM I and ALKEM II experiments at Karlsruhe [39-41]
The Mol I and Mol II experiments at Eurochemic, Mol [41-42]
The Mol III experiment at Eurochemic, Mol as a joint large-scale international venture [43-44]
The integral reactor experiment at Novovoronesh 2, USSR, conducted for the IAEA under a research contract (IAEA Research Contract No. RC994-USSR)
The Euratom integral experiment conducted in a fuel fabrication plant [45].

In the United States of America there have been three integral experiments:

The Yankee reactor experiment
The NFS experiment at the reprocessing facility in West Valley [46]
The IAEA experiment at the Westinghouse fuel fabrication plant at Columbia, S. C.

The United Kingdom has contributed to this subject of integral experiments by conducting two theoretical studies on the application of safeguards techniques to a large zero-energy fast-reactor facility and also to a fuel store of a zero-energy fast reactor [47, 48].

The recently published report on the Mol III experiment is probably the most elaborate one [43]. The technique of taking samples at the entrance of the reprocessing plant (see also [49]), an extended interlaboratory test, and the simulation of the material flow in the plant by a mathematical model [50] have been major areas of interest. Special attention was given in Ref. [50] to the in-process inventory determination by the so-called MIST technique. The other major point was the evaluation of MUF and systematic errors of the involved measurements. MIST and MUF have also been the major aspects for the two experiments at the fabrication facility ALKEM. Mol I, II and III especially have confirmed the MIST technique to be a remarkable tool. Later in the paper more attention will be given to this. The concentrated attention on the evaluation of MUF by closing the material balance has resulted in a considerable increase in detailed and reliable information on this complex subject. Reports on the other integral experiments have not yet been published.

(c) Accountability studies

The United States compilation in 1969 of papers on accountability should first be mentioned here [51]. This compilation deals with the statistical and other aspects of closing a material balance in general, and in special facilities such as chemical reprocessing, fuel fabrication and conversion plants. Other contributions came from Terrey [52], Brown, Good and Parker [14, 53], all from the UKAEA, from Nakajima et al. [54] as well as from the Karlsruhe group [55].

(d) Correlations

Correlations refer to isotopic compositions of nuclear material at various points of the nuclear fuel cycle and their mutual dependences. These isotopic

compositions and their correlations are additional information that can be used for safeguards purposes. Starting from the idea to look in particular for minor isotopes and their correlations with the hope of arriving at some sort of a fingerprinting of the nuclear material, say by concentrating on its ^{234}U or ^{238}Pu content, all sorts of isotope correlations are now covered. If, for instance, say a LWR fuel element with say 3% ^{235}U initial enrichment is burned in a power reactor, the Pu build-up produced in this way is certainly related to the depletion of the ^{235}U content [56]. This can, to some extent, be predicted in principle by careful, detailed and large-scale burnup codes. The surprising thing is that by averaging these correlations over a certain portion of the core, which is achieved by dissolving a number of fuel elements in the dissolver at the entrance of the reprocessing plant, these correlations become fairly simple in structure. In particular Schneider and the whole Battelle North West Group at Hanford have been pursuing this matter [57]. But also the contributions of Moeken and Bokelund of the Eurochemic group [58, 59] must be considered. Häfele and Nentwich considered the problem of accountability at the entrance of a reprocessing plant in its relation to the isotope correlations [6]. Isotopic changes can also help to determine the inventory of a plant while in operation. For that it is necessary to have a step in the isotopic composition of the process stream that passes then as an interface through the plant. This has been mainly investigated and experimentally proven by the Karlsruhe group [43, 60]. By confronting a deterministic description with a stochastic description, Larisse and Winter developed methods for independent determination of process inventories in a fabrication plant [61]. An extension of these investigations was presented at the Karlsruhe symposium [62].

(e) Material unaccounted for (MUF)

Material unaccounted for (MUF) has five components: the statistical measurement error, the systematic measurement error, unknown operating losses, shipper/receiver differences, and finally, the losses, if any, by diversion. In a more straightforward way, MUF is the result of a material balance, being the difference between the book and the physical inventory. Such material balance refers to a certain area, the material balance area (MBA) and a certain time interval, that is the time between two material balances. To establish a material balance, beginning and ending inventories must be taken and input and output flows must be measured. So MUF is the most direct quantitative result of accountability, being the most important among the three safeguards measures, and it offers itself directly as a basis for an inspector's statement. But here the complexities of the matter become immediately obvious: is it MUF or the time integrated value of MUF that should be the basis? Integrated MUF cancels the random measurement errors and, for safeguards, it is not so much the MUF of a particular campaign which is of importance but rather this integrated value of MUF because operating losses may happen once but should not accumulate. It should be noted that steady streams of waste and scraps must be explicitly subject to the material balance and cannot go unidentified. But one can equally well postulate that it is the tendency of MUF that must be judged, because it assures that all waste and scrap streams are identified. Also, combinations of MUF, integrated MUF and its tendencies can be considered and made the index upon which to base a statement. Surprisingly enough,

there is only a comparatively small number of sources that report explicitly on experimental values of MUF.

A major source is the contribution of Rowen, Murphey and Smith of the National Bureau of Standards [63]. Another source are the papers of Crowson [64] and of Wischow et al. [65] of the USAEC. From the Karlsruhe group, Gmelin and Kraemer have reported on MUF [66], referring to the result of the integral experiments carried out by the Karlsruhe group. From Eurochemic, Meoken et al. reported MUF values [67]. If one considers these results together and checks whether these data are narmally distributed, these checks are surprisingly positive. Singh was able to prove that all these results have an expectation value of MUF that is in a narrow interval of 0.2 to 0.5% of the total amount of material in these campaigns. The evaluation of the variance does not lead to such a narrow interval. But it is a well-known statistical fact that for obtaining a good estimate for the variance, much more input data is required than is the case for the expectation value. Even so, it is obvious that the derived value for σ is clearly larger than the expectation value μ. And, once more, these available data on MUF are indeed normally distributed.

Although Morgan, in his contribution to the Karlsruhe symposium, had stated that the absolute values of MUF as obtained from UK plant data are also normally distributed, he found a fairly large number of different outliers, depending on the time sequence he took [68]. A rigorous analysis of these data led Singh, however, to the conclusion that these absolute values were not really normally distributed. However, when he normalized the UK data with respect to the absolute values of the amounts of nuclear material involved, as he did with all other MUF data, then they also were found to be normally distributed. Surprisingly enough much fewer outliers were found for this distribution and they were the same for all ranges. These findings of Singh [69] are probably a first important step towards a more thorough understanding of MUF.

(f) Simulation

Simulating the process of nuclear material in nuclear facilities or even the whole fuel cycle is an important step for any optimization, for evaluating targets for the development of safeguards instruments and for evaluating cost effectiveness figures. The already cited papers of Oshima et al. [21, 22], and Nakajima et al. [54] must be mentioned again. The same applies to the already cited papers of Rowen, Murphy and Smith [63], Larisse [61, 62, 70], Christensen et al. [56] as well as the studies at Battelle Memorial Institute [44]. In addition to these papers the paper of Drosselmeyer and Rota must be mentioned [50]. The material flow in the Eurochemic reprocessing plant has been simulated there by a mathematical model with the purpose of investigating the possibilities and limits of in-process inventory determination by isotope correlations, as mentioned earlier. An isotopic step function is allowed to pass through the process and the purpose of this paper is to determine the degree of dissipation and distortion of that function. Beets, Goossens and Mostin have simulated part of the fuel cycle belonging to the Belgian high-flux test reactor BR2 by making a model of the fuel fabrication plant and the test reactor. The specific aspect there is that the fuel cycle uses highly enriched uranium [71]. A major simulation study of the properties of the weighted Pu/U estimate of the amount of Pu input to a processing

facility has been performed by Stewart and Schneider [72]. In the paper of Marcuse and Williams [73], a model is developed for the planning of an integral experiment for providing feedback to the plant instrument program and for assessing and extrapolating experimental results. In particular, the connection to the development of instruments by setting targets should be noted. Finally the work of Avenhaus, Gmelin, Gupta and Winter should be mentioned [74]. They simulate the material flow in single plants and evaluate parameters that are relevant for safeguards, such as the mean detection time, the probability of detection and other relevant measurement data. Work of this type will be important in a later stage of safeguards development for the implementation of a more thorough systems analysis and for optimization. This becomes obvious if one considers the logical scheme of Fig. 3 that has been discussed before.

(g) Decision theory

From looking at the logical flow sheet presented earlier in the paper, it becomes apparent that a rigorous systems analysis also embraces decisions and operations of the involved human partners of the system. The three round, big units indicate those parts of the system while being designed: there is the safeguards systems designer, the inspector and the operator. For the assessment of the strategy of his inspector and his operator the systems designer has to employ, at least ultimately, some sort of mathematical model. It is along these lines that a number of papers on decision theory have already been written. Bierlein of Karlsruhe University was the first to do so [75]; he established a model for direct surveillance: at n places of a process facility a possibility for diversion is assumed to be possible. There are k inspectors < n. Minimax strategies for the inspector are evaluated. A remarkable result is that there is a distinct number of inspectors, k_0, beyond which the operator is induced to behave legally. Admittedly, the model is simplified, but it helps to clarify the thinking. Bierlein and Beinhauer continued by incorporating the aspect of inventory-taking by surprise [76, 77]. Another aspect is the question of where thresholds of significance for certain safeguards parameters such as MUF shall be placed. This question has been investigated by Avenhaus and Höpfinger [78]. Similarly, optimum sampling procedures have been considered as a decision theoretical model by Avenhaus and Gupta [20]. Höpfinger has considered the problem of action levels as related to these thresholds of significance [79]. It is obvious that the models for making decisions or assessments need further improvement before they can be applied fully to existing problems, which is not yet the case. But, as mentioned earlier, these aspects are an integral part of a rigorous systems analysis.

(h) Cost effectiveness

Cost effectiveness is one of the constituents of systems analysis. This has already been discussed several times in this paper (see, for instance, [20, 78]). A major paper, however, that specifically focuses on cost effectiveness in the context of inventory taking is that of Stewart [80]. Techniques for giving shape to the planners' sampling-plan objectives are given by the

manner of stating the alternative hypothesis, optimizing the allocation of effort and determining a sample size which gives adequate protection.

(i) Design criteria for future plants

In practically all cases, existing nuclear facilities have not been designed with safeguards in mind. During our considerations on the logical scheme for a rigorous systems analysis, attention was given to the conduit function of containment. The containment design influences the modes of diversion that must be taken into account by the systems designer. Consequently they influence the rules on methods and procedures as well as the thinking of the inspector and the operator. The definition of strategic points [5, 30] for the existing inspection efforts and in particular the measurement of the flow of nuclear material and inventory is influenced by the design of the containment and the more general layout of the plant. Future plants can be designed to be specifically compatible with safeguards requirements [81-84]. It is a special task for the safeguards system designer to develop design criteria for future nuclear facilities. Hagen et al. of Karlsruhe have developed a first step for a reprocessing plant [85, 86]. Brown et al. in the UKAEA have also investigated that matter [87, 88].

With this paragraph on design criteria, the review of special contributions comes to an end. As mentioned earlier, these special contributions must be seen against the overall situation, both generally and as outlined in the chapter on rigorous systems analysis.

5. CONCLUDING REMARKS

Safeguards analysis, research and development is still a young discipline. It came up by challenge and has been pursued internationally on a somewhat larger scale since about 1967. Much needs to be done, specifically in the area of decisions and operations. But it was already possible to build up an international safeguards system by having the terms of systems analysis in mind, that is, the design was done from a systems point of view. This major accomplishment was possible by intense international discussion and cooperation [30].

And finally, a more practical remark: the reader who wants to familiarize himself more intimately with this young discipline still has a fairly easy situation, since the clear majority of relevant papers that exist today were presented during the first IAEA International Symposium on Progress in Safeguards Techniques that was held in Karlsruhe, Federal Republic of Germany in July 1970, and, in conclusion, I would like to emphasize the importance of this Symposium.

ACKNOWLEDGEMENTS

The author wishes to thank Dr. Gupta and Dr. Avenhaus for valuable discussions and the whole Karlsruhe group for their help in evaluating the literature.

REFERENCES

[1] SEETZEN, J., "Entwicklung zu einer allgemeinen Systemtechnik", Wissenschaft und Praxis in Kirche und Gesellschaft, 56 4 (1967) 160.

[2] CHURCHMAN, C.W., ACKOFF, R.L., ARNOFF, E.L., Introduction to Operations Research, John Wiley and Sons, New York (1957).

[3] INTERNATIONAL ATOMIC ENERGY AGENCY, Safeguards Techniques (Proc. Symp. Karlsruhe, 1970) II, IAEA, Vienna (1970) 286, remark of C.A. BENNETT.

[4] BENNETT, C.A., "Progress in systems analysis", Ibid. p.247.

[5] GMELIN, W., GUPTA, D., HÄFELE, W., On Modern Safeguard in the Field of Peaceful Application of Nuclear Energy, Rep. KFK 800 (1968).

[6] HÄFELE, W., NENTWICH, D., "Modern safeguards at reprocessing plants and reactors", see Ref. 3, I 3.

[7] INTERNATIONAL ATOMIC ENERGY AGENCY, Panel on Safeguards Methods and Techniques, Tokyo, 1969, IAEA PL-368 (1969).

[8] INTERNATIONAL ATOMIC ENERGY AGENCY, Panel on Safeguards Techniques, Vienna, 1967, IAEA PL-256 (1967).

[9] ANL, Proc. Symp. Safeguards Research and Development, Argonne National Laboratory, 1967, Rep. WASH-1076 (1967).

[10] MORGAN, F., JANISCH, D., Presentation at the IAEA Panel on Safeguards Techniques, Vienna, 1967, see Ref. 8, 17-22.

[11] HÄFELE, W., GMELIN, W., GUPTA, D., LARISSE, J., WINTER, H., Safeguards System Studies and Fuel Cycle Analysis, Rep. KFK 900 (1968).

[12] SUZUKI, G. et al., "Safeguards accountability analysis in a nuclear fuel reprocessing plant", see Refs 51, 29.

[13] STEWART, K.B., JAECH, J.L., "The role of statistics in safeguards", see Ref. 51, 197.

[14] GOOD, P.T., BROWN, F., A Statistical Approach to the Problem of Verifying the Inventories of Stores of Declared Nuclear Materials for Safeguards Purposes, UKAEA Rep. COS 2.

[15] KOUTS, H.J., WILLIAMS, J.M., "U.S. safeguards systems studies", see Ref. 51, 1.

[16] BROWN, F., "The design of a safeguards material control system", see Ref. 3, I 273.

[17] HOUGH, C.G., SOLEM, L.C., "Verification of nuclear materials", see Ref. 3, I 421.

[18] Vertrag zur Gründung der Europäischen Atomgemeinschaft (Euratom), 25 March 1957, Chapter VII, Article 77-85, see, e.g., Bundesgesetzblatt 1957, II 1014.

[19] INTERNATIONAL ATOMIC ENERGY AGENCY, The Agency's Safeguards System (1965, as provisionally extended in 1966 and 1968), IAEA, Vienna, INF/CIRC/66/Rev.2 (1968).

[20] AVENHAUS, R., GUPTA, D., "Effective application of safeguards manpower and other techniques in nuclear fuel cycles", see Ref. 3, I 345.

[21] OSHIMA, K., IMAI, R., NAKAJIMA, N., WATANABE, S., "Dynamic safeguards system for multiple action levels", see Ref. 3, I 375.

[22] OSHIMA, K., NAKAJIMA, N., WATANABE, S., IMAI, R., "An approach to international safeguards system", see Ref. 7, IAEA PL-368.

[23] INTERNATIONAL ATOMIC ENERGY AGENCY, Panel on Safeguards Technical Practices for Irradiated Fuel Plants, Vienna, 1968, IAEA PL-279 (1968).

[24] INTERNATIONAL ATOMIC ENERGY AGENCY, Panel on Safeguards Methods for Reactors, Vienna, 1968, IAEA PL-315 (1968).

[25] INTERNATIONAL ATOMIC ENERGY AGENCY, Panel on Safeguards Methods for Conversion and Fabrication Plants, Vienna, 1969, IAEA PL-332 (1969).

[26] INTERNATIONAL ATOMIC ENERGY AGENCY, Panel on Safeguards Systems Analysis of Nuclear Fuel Cycles, Vienna, 1969, IAEA PL-352/353 (1969).

[27] INTERNATIONAL ATOMIC ENERGY AGENCY, Working Group on Interim Determination of Design Information Requirements, Vienna, 1970, IAEA Rep. (1970).

[28] INTERNATIONAL ATOMIC ENERGY AGENCY, Safeguards Technical Working Group on the Verification of Nuclear Material, Vienna, 1970, IAEA Rep. (1970).

[29] BENNETT, C.A., GRANQUIST, D.P., "Safeguards Systems Studies", see Ref. 9, 51.

[30] INTERNATIONAL ATOMIC ENERGY AGENCY, The Structure and Content of Agreements between the Agency and States Required in Connection with the Treaty on the Non-Proliferation of Nuclear Weapons, IAEA, Vienna, INF/CIRC/153 (1971).

[31] MORGAN, F., Report to the Director-General of the IAEA by the Consultants on Criteria for Safeguards Procedures (Topic 1), IAEA (1969).

[32] INTERNATIONAL ATOMIC ENERGY AGENCY, Statute, as amended up to 31 January 1963, Art. III A.5, IAEA (1967).
[33] see Ref.29, pp.53.
[34] see Ref.26, pp.10 and 21.
[35] FREDERIKSEN, P., JANISCH, D.B.B., JENNEKENS, J.H., Report to the Director General of the IAEA by the Consultants on Criteria for Safeguards Procedures (Topic 2), IAEA (1969).
[36] BENNETT, C.A., HOUGH, C.G., LENDVAI, O., PUSHKOV, A., Report to the Director General of the IAEA by the Consultants on Criteria for Safeguards Procedures (Topic 3), IAEA (1969).
[37] SHMELEV, V., "Progress in safeguards systems analysis", Atom. Energy Rev. 8 2 (1970) 417.
[38] SHMELEV, V., "Fuel cycle study for safeguards systems analysis", see Ref.3, II 373.
[39] GMELIN, W., NENTWICH, D., OTTO, H.E. et al., Safeguards Exercise at the Fabrication Plant ALKEM, Rep. KFK 901 (1969).
[40] GMELIN, W., to be published.
[41] KRAEMER, R. et al., "Integral safeguards exercises in a fabricating and a reprocessing plant", see Ref.3, I 83.
[42] KRAEMER, R., VON BAECKMANN, A., HAGEN, A., NENTWICH, D., Beschreibung eines Kontroll-experiments in der Wiederaufarbeitungsanlage EUROCHEMIC, Rep. KFK 907 (1969).
[43] KRAEMER, R., BEYRICH, W. (Eds) et al., Joint Integral Safeguards Experiment (JEX-70) in the Reprocessing Plant EUROCHEMIC, Mol (BELGIUM), Rep. KFK 1100, EUR 4576e (1971).
[44] EWING, R.A., SCHNEIDER, R.A., Supporting Studies for the Mol III Integral Experiment, Battelle Memorial Institute, Columbus Laboratories, ACTIAC Rep.24 (1970).
[45] MIRANDA, U. et al., "Contribution to safeguards studies in fabrication plants", see Ref.3, I 105.
[46] SHARPE, B.W., FRENZEL, W., FRITTUM, H., RUBINSTEIN, G.R., "IAEA safeguards operations in USA", see Ref.3, I 167.
[47] BROWN, F. et al., "The application of safeguards techniques to a large zero-energy reactor facility", see Ref.3, I 125.
[48] BROWN, F. et al., Progress in the Development of a System for the Safeguards Inspection of the Fuel Store of a Zero-Energy Fast Reactor, UKAEA Rep. COS 20.
[49] COSTELLO, J.M. et al., Procedures used in the Calibration of Accounting Tanks, UKAEA Rep. COS 22.
[50] DROSSELMEYER, E., KRAEMER, R., ROTA, A., "Application of digital simulation techniques for process inventory estimation", see Ref.3, II 311.
[51] UNITED STATES ATOMIC ENERGY COMMISSION, Safeguards Systems Analysis of Nuclear Fuel Cycles, Rep. WASH-1140 (1969).
[52] TERREY, D.R., PLANTER, A Computer Program which Simulates Error Propagation in Nuclear Materials Processing Plants, UKAEA Rep. COS 3 (1969).
[53] BROWN, F., GOOD, P.T., PARKER, J.B., Sampling of Shares for Safeguards Purposes, UKAEA Rep. COS 4A.
[54] NAKAJIMA, K. et al., "Analysis of accountability system in a reprocessing plant and a plutonium fuel facility", see Ref.3, I 139.
[55] GMELIN, W., GUPTA, D., HÄFELE, W., Use of Statistical Analysis for the Establishment of Material Balance in a Reprocessing Plant, Rep. KFK 802 (1968).
[56] CHRISTENSEN, D.E. et al., "A summary of results obtained from the first MIST experiment at NFS, West Valley, New York", see Ref.3, I 563.
[57] SCHNEIDER, R.A., The Use of Isotope Correlations in Nuclear Materials Safeguards, Rep. BNWL-B-100 (1970).
[58] MOEKEN, H.H.Ph., BOKELUND, H., Verification of the Uranium and Plutonium Measurements on a Batch of Dissolved Reactor Fuel, EUROCHEMIC Rep. ETR-235 (1969).
[59] MOEKEN, H.H.Ph., BOKELUND, H., "Some developments in input accountability at EUROCHEMIC", see Ref.3, I 551.
[60] WINTER, H., AVENHAUS, R., GUPTA, D., KATZ, F., KRAEMER, R., Determination of In-Process Inventory in a Reprocessing Plant by Means of Isotope Analysis, Rep. KFK 904 (1969).
[61] LARISSE, J., WINTER, H., Methods for Independent Determination of Process Inventory in Nuclear Facilities – Fabrication Plant –, Rep. KFK 903 (1968).
[62] LARISSE, J., MATTHES, W., MÜLLER, K.-H., ROTA, A., "Contribution to systems analysis of nuclear plants", see Ref.3, II 333.
[63] ROWEN, J.W., MURPHEY, W.M., SMITH, C.N., "Assessment of 'Material unaccounted for' control criteria", see Ref.3, II 291.
[64] CROWSON, D.L., "Progress and prospects for nuclear materials safeguards", see Ref.3, I 23.
[65] WISCHOW, R.P., D'AMICO, V.J., PAGE, R.G., ONG, L.D.Y., "US safeguards experience in regulation and inspection of the private nuclear industry", see Ref.3, I 435..

[66] GMELIN, W., KRAEMER, R., "Analysis of components of material unaccounted for", see Ref. 3, I 451.
[67] MOEKEN, H.H.Ph., FRANSSEN, F., BECKERS, J., "Study of the MUF values calculated daily over three balance areas during a reprocessing run for highly enriched uranium fuel", see Ref. 3, I 473.
[68] MORGAN, F., "The usefulness of systems analysis", see Ref. 3, II 265.
[69] SINGH, H., Analysis of some Available Data on Material Unaccounted For (MUF), Rep. KFK 1106 (1971).
[70] LARISSE, J., Compartement d'un processus de fabrication dans lequel interviennent des tests de contrôle, Revue Française d'Informatique et de Recherche Opérationnelle, Série Rouge, R 3, 91 (1970).
[71] BEETS, C., GOOSSENS, H., MOSTIN, N., "Estimation de l'efficacité des mesures de contrôle pour les usines de fabrication et les reacteurs", see Ref. 3, I 251.
[72] STEWART, K.B., SCHNEIDER, R.A., "Properties of the Pu estimate based on weighted Pu/U values", see Ref. 3, I 583.
[73] MARCUSE, W., WILLIAMS, J.M., "Application of a nuclear process simulation model to an integrated safeguards experiment", see Ref. 3, II 351.
[74] AVENHAUS, R., GMELIN, W., GUPTA, D., WINTER, H., Relations between Relevant Parameters for Inspection Procedures, Rep. KFK 908 (1970).
[75] BIERLEIN, D., "Direkte Überwachungssysteme", Op. Res. Verf. 6 (1969) 57.
[76] BIERLEIN, D., "Indirekte Überwachungssysteme", Op. Res. Verf. 7 (1970) 36.
[77] BEINHAUER, R., BIERLEIN, D., "Games theoretical models for inspection procedures", see Ref. 3, II 425.
[78] AVENHAUS, R., HÖPFINGER, E., "Optimal inspection procedures in a nuclear facility for a sequence of inventory periods: a decision theoretical model for an inspection system based on material measurements", see Ref. 3, II 411.
[79] HÖPFINGER, E., Criteria for Action Levels for Inspection Based on a Game Theoretical Model, Rep. KFK 909 (1969).
[80] STEWART, K.B., "A cost effectiveness approach to inventory verification", see Ref. 3, II 387.
[81] VON BAECKMANN, A., GMELIN, W., GUPTA, D., HÄFELE, W., Fissile Material Flow Control at Strategic Points in a Reprocessing Plant, Rep. KFK 801 (1968).
[82] GUPTA, D. et al., Safeguarding Fissile Material Flow at Strategic Points in Power Reactors, Rep. KFK 803 (1969).
[83] SCHRÖDER, R., WINTER, H., GUPTA, D., HÄFELE, W., Development of Safeguards Procedures for Heavy Water Moderated, Cooled and Reflected Pressurized Water Type Reactors, Rep. KFK 804 (1970).
[84] GUPTA, D., GMELIN, W., KRAEMER, R., RICHTER, K., SCHNEIDER, V., Safeguards Measures and Efforts in Conceptual Fabrication Plants for Uranium and Plutonium Containing Fuel Elements, Rep. KFK 910 (1969).
[85] HAGEN, A., NENTWICH, D., KRAEMER, R., GUPTA, D., HÄFELE, W., Development of Safeguards Procedures for a Reprocessing Plant Similar to the WAK Type, Rep. KFK 1102 (1970).
[86] HAGEN, A., HERRE, F., GUPTA, D., Design Criteria for Reprocessing Plants, Rep. KFK 1107 (1970).
[87] BROWN, F. et al., Design Criteria for Nuclear Installations to Facilitate the Application of Safeguards, UKAEA Rep. COS 12 (1970).
[88] BROWN, F. et al., "Criteria for the design of nuclear installations to facilitate the application of safeguards", see Ref. 3, I 389.

A REVIEW OF THE GOALS, METHODS AND TECHNIQUES OF NUCLEAR MATERIALS SAFEGUARDS*

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Abstract–Résumé–Аннотация–Resumen

A REVIEW OF THE GOALS, METHODS AND TECHNIQUES OF NUCLEAR MATERIALS SAFEGUARDS.

The purpose of a safeguards system is to detect, or determine through threat of detection, the diversion of nuclear material for unauthorized uses. The input is the definition of activities required and/or carried out to achieve this purpose, and the output is "assurance that significant diversion has not taken place". Analysis and quantification of existing systems; present and future research and development needed to improve the present system or provide equally effective alternatives; and implications of the safeguards system with respect to the design and operation of nuclear facilities are discussed. A decade of study of the nature of safeguards systems and analysis of their operating characteristics has led to a broad degree of international agreement on the best alternatives and their capabilities. However, there is less agreement on the extent to which implementation is feasible and desirable, since this may be highly dependent on the nature of the facilities and operations to be safeguarded and the definition of safeguards objectives. Present systems work is being directed towards (1) determining the consequences of imposed constraints and (2) examining possibilities for improving or modifying the system. Review of experience has led to the conclusion that research and development activities should concentrate on providing and improving methods of non-destructive assay for fissionable material. This program should consist of three parts: (1) development of techniques for assay; (2) definition of equipment requirements for application of these techniques, both for use in plants and by inspection staff; and (3) determination of the errors, costs, and throughputs of measurement devices using these techniques. Preliminary estimates have been made of the accuracy of a material balance over segments of the nuclear industry when these methods are used. One area of recent emphasis has been the development of design criteria for industrial nuclear facilities of several kinds. The first results have been a number of recommended criteria to be used during the initial design stage. These criteria would ensure that facilities embody structural and equipment features that might be expensive or time-consuming to add later. The second stage of this activity provides a list of physical and procedural characteristics considered necessary for safeguarding specific kinds of nuclear facilities. This set of characteristics is constructed to meet the needs of United States domestic safeguards as they arise in connection with future plants. As the International Atomic Energy Agency's safeguards methods become better defined, the list of characteristics will, if necessary, be modified to embody Agency needs.

EXAMEN DES OBJECTIFS, DES METHODES ET DES TECHNIQUES DU CONTROLE DES MATIERES NUCLEAIRES.

Le but d' un «système» de contrôle est de détecter ou de déterminer, en menaçant de détection, le détournement de matières nucléaires à des fins non autorisées. «Les données d' entrée» sont constituées par la définition des activités nécessaires ou mises en œuvre à cette fin et les «données de sortie» par «l'assurance qu' aucun détournement important n' a eu lieu». Le mémoire décrit l' analyse et la quantification des systèmes existants, les travaux de recherche et de mise au point qui sont ou seront nécessaires pour améliorer le système actuel ou fournir d' autres solutions également efficaces et les incidences du système de garanties pour l' étude et l' exploitation des installations nucléaires. Après dix ans d' étude sur la nature des systèmes de garanties

* Work performed under the auspices of the U.S. Atomic Energy Commission.

et d' analyse de leurs caractéristiques opérationnelles, la communauté internationale est parvenue à un large degré d' accord quant aux meilleures solutions et à leurs possibilités. Toutefois, l' accord est moindre quant à la mesure dans laquelle la mise en œuvre de ces systèmes est possible et souhaitable, car ceci peut dépendre dans une large mesure de la nature des installations et des opérations soumises aux garanties et de la définition des objectifs des garanties. Les travaux actuels en la matière visent à 1) déterminer les conséquences des limitations imposées et 2) examiner les possibilités d' amélioration ou de modification du système. A la lumière de l' expérience acquise, on est parvenu à la conclusion que les travaux de recherche et de mise au point devraient viser à instaurer et améliorer des méthodes d' analyse non destructive des produits fissiles. Le programme devrait comporter trois parties: 1) mise au point de méthodes d' analyse; 2) définition du matériel nécessaire à l' application de ces méthodes, à la fois dans les installations et par les services d' inspection; 3) détermination des erreurs, et des coûts des dispositifs de mesure utilisant ces techniques, ainsi que des quantités de produits mesurés. On a procédé à des évaluations préliminaires de l' exactitude d' un bilan matières dans les secteurs de l' industrie nucléaire où ces méthodes sont utilisées. On a particulièrement mis l' accent récemment sur l' étude des caractéristiques à donner à différents types d' installations nucléaires. On est d' abord parvenu à établir un certain nombre de critères à appliqner au cours de l' étude initiale. Ces critères assureraient que les installations comportent des structures et des appareils dont l' adjonction pourrait ultérieurement être onéreuse ou demander beaucoup de temps. Au deuxième stade, on a établi une liste de caractéristiqnes physiques et opérationnelles jugées nécessaires pour le contrôle de certaines catégories d' installations nucléaires. Cet ensemble de caractéristiques est destiné à répondre aux besoins du contrôle qui devrait être exercé sur le plan intérieur aux Etats-Unis du fait de l' aménagement des futures installations. A mesure que les méthodes de garanties de l' Agence internationale de l' énergie atomique seront mieux définies, on modifiera le cas échéant la liste des caractéristiques pour mieux répondre aux besoins de l' Agence.

РАССМОТРЕНИЕ ЦЕЛЕЙ, МЕТОДОВ И ТЕХНИЧЕСКИХ ПРИЕМОВ ПРИМЕНЕНИЯ ГАРАНТИЙ К ЯДЕРНЫМ МАТЕРИАЛАМ.

Цель "системы" гарантий состоит в том, чтобы обнаружить или определить, под угрозой обнаружения, переключение ядерных материалов на неразрешенные цели. Проверка ввода ядерного материала в установку – это определение деятельности, необходимой и(или) проводимой для осуществления данной цели, проверка выводов материала подтверждает "уверенность в том, что не произошло значительного переключения материала". В данном докладе обсуждаются вопросы анализа и количественного определения существующих систем; современные и будущие научные исследования и разработки, необходимые для улучшения существующей системы или для предоставления таких же эффективных альтернатив; применение системы гарантий в области проектирования и эксплуатации ядерных установок. Десятилетнее изучение характера систем гарантий и анализ их рабочих характеристик привели к определенной международной договоренности относительно наилучших альтернатив и их возможностей. Однако не имеется полной договоренности относительно того, насколько желательно и возможно это применение, поскольку это в значительной степени может зависеть от характера установок и операций, к которым должны применяться гарантии, и от определения целей гарантий. Работа существующих систем направлена на: 1) определение влияния введенных ограничений, 2) изучение возможностей для улучшения или изменения системы. Изучение накопленного опыта привело к выводу о том, что научные исследования и разработки должны сосредоточиться на предоставлении и улучшении методов анализа делящегося материала без разрушения образца. Данная программа должна состоять из трех частей: 1) разработка методов анализа; 2) определение потребностей в оборудовании для применения этих методов как для использования на установках, так и инспекторами; 3) определение ошибок, расходов и производительности измерительных устройств, в которых применяются эти методы. Проведено предварительное определение точности баланса материалов по различным отраслям ядерной промышленности, где используются указанные методы. Одной из областей, которой недавно уделялось особое внимание, была разработка критериев для проектирования промышленных ядерных установок нескольких типов. Первыми результатами был ряд рекомендованных критериев для использования во время первоначального проектирования. Эти критерии обеспечат воплощение в установках структурных особенностей оборудования, которые могут оказаться дорогостоящими или требующими много времени, если их дополнять позднее. Второй этап этой работы предусматривает список физических и процедурных характеристик, которые считаются необходимыми для применения гарантий к особым видам ядерных установок. Эти характеристики составлены таким образом, чтобы отвечать требованиям внутренних гарантий США по мере их возникновения в связи с созданием будущих установок. По мере более точного определения методов применения гарантий Международного агентства по атомной энергии мы будем при необходимости изменять список характеристик, для того чтобы отражать требования Агентства.

EXAMEN DE LOS FINES, METODOS Y TECNICAS DE SALVAGUARDIAS DE MATERIALES NUCLEARES.

La finalidad de un «sistema» de salvaguardias consiste en detectar o determinar por medio de la posibilidad de detección la diversión de materiales nucleares para empleos no autorizados. El principio consiste en la determinación de las actividades requeridas y/o realizadas para alcanzar esa finalidad, y el resultado es «la seguridad de que no ha tenido lugar una diversión significativa ». En la memoria se estudia el análisis y cuantificación de los sistemas existentes; las investigaciones y desarrollos, actuales y futuros, necesarios para mejorar el sistema actual o proporcionar alternativas igualmente efectivas; y las implicaciones del sistema de salvaguardias con respecto al diseño y funcionamiento de instalaciones nucleares. Una década de estudio del carácter de los sistemas de salvaguardias y de análisis de sus características de funcionamiento ha llevado a un amplio acuerdo internacional sobre las mejores alternativas y posibilidades. No obstante, el acuerdo es menor sobre la extensión en que es posible y deseable la aplicación de tales sistemas, puesto que ello puede depender de la naturaleza de las instalaciones y de las operaciones que se sometan a salvaguardias, así como de la determinación de los objetivos de las mismas. Los actuales trabajos sobre los sistemas se orientan hacia: 1) la determinación de las consecuencias de las limitaciones impuestas; 2) el examen de las posibilidades de mejorar o modificar el sistema. La consideración de la experiencia ganada ya ha llevado a la conclusión de que las actividades de investigación y desarrollo deberían concentrarse en proporcionar y mejorar los métodos de ensayo no destructivos de materiales fisionables. Este programa consistiría en tres partes: 1) desarrollo de técnicas de ensayo; 2) determinación de las necesidades de equipo para la aplicación de esas técnicas, para su empleo tanto en las plantas como por el personal de inspección; 3) determinación de los errores, costos y rendimientos totales de los aparatos de medida que emplean esas técnicas. Se han preparado cálculos preliminares de la precisión de un balance de materiales referente a sectores de la industria nuclear cuando se emplean dichos métodos. Recientemente se ha prestado especial atención al desarrollo de criterios de diseño para varias clases de instalaciones nucleares industriales. Los primeros resultados han producido cierto número de criterios recomendados para su aplicación durante la fase inicial de diseño. Esos criterios asegurarían la incorporación en las instalaciones de características estructurales y de equipo que, si hubieran de introducirse más tarde podrían ser costosas o causar pérdidas de tiempo. La segunda fase de estos estudios la servido para proporcionar una lista de características físicas y de procedimiento necesarios para cierto tipo específico de salvaguardias de las instalaciones nucleares. Este conjunto de características tiene como fin satisfacer las necesidades referentes a las salvaguardias internas en los Estados Unidos que se hayan de aplicar a futuras plantas. A medida que vayan perfeccionándose los métodos de salvaguardias del Organismo Internacional de Energía Atómica se irá modificando, en los casos necesarios, la lista de características para que incorpore los requisitos exigidos por el Organismo.

Safeguards on nuclear materials are undergoing evolution in the United States, as well as in the international community. The changes under way are largely the result of analytical studies of the goals of safeguards and of the methods and techniques available for achieving the goals. We shall summarize here some of the reasoning that has evolved in American analysis, and some of the new directions being explored.

General Objectives

There is generally broad agreement on the basic elements of safeguards systems and the purposes served by these elements. The basic elements of any safeguards system are surveillance, a component called either containment or physical protection, depending on the context, and material balance accounting. Containment can involve the use of fences, substantial walls and enclosures, locks, tags, seals, and similar measures available for international as well as national safeguards. A national safeguards system can also make use of guards and security devices. The mixture of containment and protective measures in national safeguards is usually called physical protection. Surveillance includes direct observation and monitoring, and, equally importantly, verification of plant operation and plant data. Material balance accounting is concerned with records, reports, and source data from the facility being inspected, and independent measurement as needed to form independent conclusions on materials accountability. While surveillance and physical measures provide direct and timely information on safeguarded material, accounting procedures provide indirect or circumstantial evidence.

A safeguards system achieves effectiveness through its capability to deter or prevent diversion attempts and to detect diversion if it occurs. Physical protection, including containment, provides a clear-cut capability for timely evaluation; for if a diversion were attempted, protective barriers would have to be breached, or broken locks or seals could ensue. However, effective containment and protection can sometimes restrict the mobility of material and plant personnel, and this may be incompatible with the operation of the process.

Surveillance can both detect and deter diversion, but its effectiveness is very difficult to quantify. Surveillance is certainly more timely and effective if it is continuous. Anyone intent on performing a diversion that might be detected by surveillance would obviously make his attempt when surveillance is minimal or absent. Any non-random or announced periodic surveillance would be largely ineffective in revealing a diversion attempt, unless measures were included to ensure that evidence is left afterwards.

Procedures such as those discussed above are designed primarily for timely evidence regarding a possible diversion. Material accounting procedures are used to analyze the possibility that diversion may have taken place over some past period of time. Thus, it is possible through material accounting procedures to "catch up" with a diversion that has taken place in the past. In this way, the material balance portion of the safeguards system can set an upper limit to the amount which could have been diverted without detection. On the other hand, evidence from this type of inspection activity is always circumstantial unless it leads to after-the-fact investigation providing direct evidence regarding possible diversion. There is also a threshold to the sensitivity of material balance accounting, in that small (and perhaps not so small) diversions could occur with little effect on the accounts. Both the deterrent effect and the sensitivity of material accounting depend on the amount and type of material.

The preceding framework is useful in characterizing the input to a safeguards system. Characterizing the output is much more difficult. The amount of effort required for a given effectiveness or the effectiveness from a given effort will vary with the size of the facility and the throughput and complexity of the process. Because they contain immobile nuclear material, reactors present safeguards problems that are generally different from those either of chemical-processing plants or fuel-fabrication plants. Likewise, the high radioactivity of the materials encountered in chemical processing plants contributes to causing differences between safeguards at these plants and at fuel fabrication plants.

The effectiveness of inspection will of course depend on the intensity of effort. It will depend on the technical ability of inspectors, and the technical facilities available to them. The effectiveness will vary with the number of inspection personnel.

Intuitive considerations of this kind show in what ways the effectiveness of safeguards is a function of the size, or complexity, of the facility inspected and the magnitude of the inspection effort. These considerations can also be subjected to approximate quantification. Figure 1 shows an example of the relationship between these factors, as developed several years ago in connection with a study of inspection requirements for one plutonium production complex. The general relationships in Figure 1 should always be valid in most safeguards applications, although such attempts to quantify them are always subject to question.

No system can absolutely guarantee 100 percent effectiveness. No purely technical means exists for deciding just what combination of effort and ef-

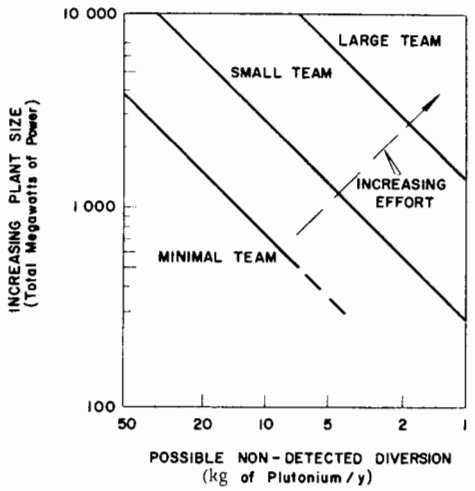

FIG.1. Dependence of effectiveness on complexity and effort.

fectiveness is right for a given facility. One difficulty in analyzing the needs for safeguards activities and for research and development has been this lack of a good definition of safeguards effectiveness, and a quantitative basis for evaluating how good safeguards must be. Usually we have only been able to determine approximate relationships of the type shown in Figure 1.

The above studies to date indicate that among the fruitful areas for further research and development leading to systems improvement are:

1. improved measurement procedures, particularly nondestructive methods that are on-line, accurate, and fast. These methods are needed to ensure that only measured data are used in material balance accounting.

2. methods of containment, physical protection, and surveillance that simplify accounting, improve assurance, and do not impede normal operation of processes. An example is the use of secure seals on shipments of fissionable material to ensure that any differences between shippers' and receivers' measured values of the amounts shipped are not indicative of theft in transit.

3. a system of reporting and analyzing accountability data that provides timely assurance with respect to a complete system, as well as to each facility in the system.

4. criteria for the design, construction, and operation of facilities of different kinds that define operational boundaries for safeguards purposes. Operation of a facility within the boundaries and according to the criteria should normally provide adequate assurance regarding safeguards.

5. continued study of the definition and evaluation of effectiveness.

These areas of research and development fall in two classes: study of methods of improvement of safeguards by facilities possessing and using fissionable material, and studies of methods of improving and quantifying assurance derived from inspection. In the first area, there has been an emphasis on the methods of safeguards and definition of the steps required for effective implementation of the method. Progress in the second area has led to recognition that:

1. analysis of inspection strategy depends strongly on the definition of the threat involved, not only in terms of the economic or strategic value of the material but also the possible methods and motives for diversion that must be considered, and

2. the feasibility and effectiveness of a given technique will vary with its level of application, so that the inspection strategy should vary qualitatively and quantitatively at various levels.

The first of these says that at the start of technical analysis, certain policy decisions are essential to define the required system. The second implies that the balance among the elements of the system, if not the elements themselves, should be expected to vary with the output required. It should be expected that a system developed by the IAEA to guard against nuclear proliferation, particularly at the national level, would differ in many important respects from the system used by a plant operator to insure against individual thefts.

We shall now focus on two aspects of the analytical studies: (1) the features and goals of design criteria that are being developed, and (2) estimates of the accuracy of a material balance over segments of the industry when new trends in design criteria and procedural criteria are effected.

Design Criteria

It has been recognized for some time[1] that certain plant design features are important to the safeguarding of strategically significant quantities of special nuclear material. Under the sponsorship of the AEC's Office of Safeguards and Materials Management, a programmatic study has been initiated to develop design criteria which, when implemented, should provide assurance that the design and construction of facilities are compatible with good safeguards practice. Initial studies are directed at producing criteria for future, privately-owned nuclear facilities, specifically, power and research reactors, fuel reprocessing plants, and fuel conversion and fabrication plants. Similar studies are to be conducted for AEC-owned and contractor-operated facilities to develop criteria equivalent to those for privately-owned facilities. Problems from any need for retrofitting of existing plants will be the subject of a later study.

The draft criteria which have been developed are still undergoing review and modification, and it would therefore be premature to state them in this paper. It is more appropriate to review the aspects of plant design and operations that have been considered. Because of the general acceptance and use of the health- and safety-oriented General Design Criteria for Reactors used for reactor safety analysis in the U.S.,[2] the safeguards-oriented criteria have borrowed several general concepts from the reactor criteria with appropriate changes to reflect the safeguards implications.

An important aspect of the draft design criteria is a provision that those systems and components which involve features important to safeguarding will be designed, fabricated, erected, tested, and maintained to quality standards that reflect the importance of, and assure the ability to implement, the safeguards functions to be performed. In order to facilitate safeguards inspection and confirmation, appropriate surveillance equipment will be installed, and adequate records will be kept of the design, fabrication, tests and maintenance of items important to safeguards.

In order to assure adequate physical protection and security of the facilities, the criteria include requirements for physical barriers, facility

isolation and lighting, restrictions on personnel access, secure methods of storage of special nuclear materials, and communication and intrusion alarm systems. There are also recommended design criteria as regards sampling capability, facilitation of the cleanout of equipment, and capability of the plant with its equipment and laboratories to measure with acceptable accuracy the amount and composition of all special nuclear materials in the plant, where and when such measurements are deemed important to nuclear safeguards.

A major consideration has been the intent to allow the plant designer as much latitude as possible in the design of his equipment. It should be unarguable that satisfactory safeguards requires a sure knowledge of the SNM content of any material entering or leaving the plant. This includes the special nuclear material in product, in waste (including all effluents), and in any scrap shipped off site. In addition, equipment should be designed to accommodate physical inventory, both for material in process and for material in storage.

Design criteria of this kind are related to elements of design information for safeguards administered by the International Atomic Energy Agency[3], but there are also fundamental differences. The U. S. studies on design criteria are intended to develop information helpful to plant design at an early stage while the design is flexible and the difficulty and expense of design changes are small. It is hoped that use of these criteria will largely avert any need for structural change to provide adequate safeguards after construction is finished. The design information sought by the International Atomic Energy Agency includes aspects of design of structures and equipment and also design of procedures. These are to be used in developing details of an agreement between the Agency and the State on administration of safeguards.

Procedural Safeguards

The above study of design criteria has been concerned more with aspects of physical protection and automated surveillance and only to a lesser extent with materials accounting. The reverse is true of the study of procedural safeguards. We shall consider here only that part of procedural safeguards related to accounting and only that part of safeguards that might be administered by the operator of a facility.

The study begins from the following assumptions:

1. Materials accounting by the facility must be sensitive to the possibility of diversion of special nuclear material containing amounts of U-235, plutonium, or U-233 exceeding certain values. These values, which may be different for different fissionable materials, we shall call the threshold values related to these materials. While specific threshold values have been chosen for the purpose of this study, other and in fact smaller values might be appropriate for other studies.

2. Materials accounting by the facility must provide assurance against the possibility of diversion over a critical period of time. The critical time is taken to be a period not longer than the minimum time between a postulated diversion and the construction of a nuclear explosive device from diverted material. This is the critical time as has been defined by Morgan[4] and Brown[5], although the times used in the present study differ to some extent from the ones suggested by Brown.

3. Indices that must be evaluated in terms of the threshold value and the critical time are required on the following quantities:

a. material unaccounted for (MUF)

b. the limit of error on MUF (LEMUF), as deduced from propagation of random and systematic errors.

Additional indices are required for:

c. measured discards

d. the accuracy of measurement of discards

e. shipper-receiver differences, both on single shipments and on sets of related shipments between the same shipper and the same receiver.

4. The analysis related to evaluation of the indices must be based entirely on measured quantities. The data used in the materials accounts must then all be measured as well. This requirement calls for a fully measured material balance, and it leads to a subsidiary set of requirements for measurements, programs of measurement calibration, records, and records analysis.

5. Secure methods of physical protection, containment, and surveillance can be used in special cases to extend beyond the critical time the period of time within which assurance is needed. For instance, highly reliable seals on containers or vaults can be used for this purpose.

Specific assumptions have been made with respect to threshold values and critical times for the purposes of this study. Other assumptions might be appropriate as well. The present assumptions are listed in Table I. The analysis has explored the capability of a safeguards system, administered by a facility possessing and using special nuclear material, to provide assurance consistent with Table I through the use of measured material balances using certain assumed measurement methods. These are not necessarily the best measurement methods that could be used.

The requirement for use only of measured data in materials accounting has been discussed by Suzuki[6] and Marcuse[7], who have shown that data obtained using by-difference methods instead of complete measurement tend to destroy the usefulness of accounting in analysing the possibility of diversion. The complete use of measured data also has the interesting and valuable property that it permits an almost continuous estimate to be formed on the closing of the material balance. This estimate is only as timely as the measurements used in the accounting. The basis of the estimate is the difference between the Book Inventory (Receipts less Withdrawals for the facility or the Material Balance Area) and the Tag Inventory (the sum of the tag values of

TABLE I. THRESHOLD VALUES AND CRITICAL TIMES ASSUMED FOR THIS STUDY

Material	Threshold values	Critical time
Plutonium	2 kg Pu	10 d
Low-enriched uranium	5 kg ^{235}U	1 month
High-enriched uranium	5 kg ^{235}U	10 d
^{233}U	2 kg ^{233}U	10 d

special nuclear material in containers or otherwise identified).[8] This MUF-like quantity is equal to the sum of MUF and the amount of special nuclear material in process, if the accounts are formed in such a way as to reflect the entry of material into processing. The amount of SNM in process can go to zero either through intent or as the result of normal fluctuation in manufacturing. At such a time, the MUF-like quantity is equal to the MUF. Accounts based on use of the measured tag values to define Tag Inventories provide an ultimate in timeliness of materials accounting.

Materials accounting based on fully measured tag values leads to considerable simplification and clarification of the procedures for physical inventory. In a sense, the Tag Inventory already has some of the properties of a physical inventory, because all tag values are measured values. Physical inventory then consists of go- no-go tests of the gross presence of the material listed on all tags, supplemented by verification of attributes listed on tags, using statistical sampling methods. The go- no-go tests considered so far are simple verifications of gross weights; the attributes considered are chemical and isotopic compositions. Studies until now have considered only conventional chemical and isotopic methods in most process and inventory measurements. It is recognized, however, that nondestructive methods of testing and assaying can simplify and speed up these activities, as well as provide them with a new dimension. Extension of the analysis to include use of nondestructive methods has a high priority.

Studies based on these points of view have been made for facilities fabricating low enriched uranium oxide fuel for power reactors, facilities fabricating reactor fuel from mixed oxides of plutonium and uranium, and chemical reprocessing plants for spent reactor fuel. These constitute most

TABLE II. CALCULATED PERFORMANCES OF DIFFERENT FACILITIES

Type of facility	Critical time	LEMUF
Low-enriched uranium oxide fuel fabrication[a]	1 month	9.72 kg ^{235}U[f]
Low-enriched uranium oxide fuel fabrication[b]	1 month	2.59 kg ^{235}U[f]
Mixed plutonium-uranium oxide fuel fabrication[c]	10 d	2.97 kg Pu[f]
Mixed plutonium-uranium oxide fuel fabrication[d]	10 d	1.59 kg Pu[f]
Spent fuel reprocessing[e]	1 month for Pu (based on secure process MBA)	2.15 kg Pu

[a] 2.2 tonne/d production rate, starting with hydrolysis of UF_6, 2.12% ^{235}U.
[b] 0.5 tonne/d production rate, starting with hydrolysis of UF_6, 2.5% ^{235}U.
[c] 0.5 tonne/d production rate, co-precipitation, 2% plutonium oxide.
[d] 0.25 tonne/d production rate, dry blending, 2% plutonium oxide.
[e] 2 tonne/d processing rate, 7 kg plutonium/tonne uranium, item accounting used up to fuel dissolution.
[f] Allowance made for some covariances among receipts, product, and inventories.

of the relevant facilities in the power reactor cycle, inasmuch as the power reactors themselves are best dealt with on the basis of item accounting for identifiable or countable fuel assemblies and fuel pins.

Typical facility-wide values of LEMUF at the 95% confidence level for different kinds of facility and different sizes of facility are shown in Table II. These have been calculated using specific assumptions as to error limits of individual measurements. These are not necessarily the best measurement methods that might be specified, but they are currently used or currently available methods. It is seen that in most cases values of LEMUF would meet criteria based on closure of the material balance to an accuracy compatible with the threshold values in Table I over the critical times in Table I at least for the smaller facilities. As the facility size is increased, several courses of action are available. Better methods of measurement and calibration could be used than those assumed for the calculations, to reduce the estimated value of LEMUF to below criteria levels. Alternatively, the facility can be divided into several material balance areas. This possibility is less helpful for reprocessing plants than for inventory-dominated facilities such as those for fabricating fuel.

Subdivision of facilities into smaller material balance areas is facilitated by the use of the classification shown in Figure 2, which differs somewhat from the classification proposed by the IAEA for international use. The basic material balance area is the process area. A process area receives feed, and it experiences removals as product, intermediate product, or waste. A secure process area has an added feature: secure containment is used to assure that special nuclear material only enters and leaves the area in monitored and completely measured streams. MUF can be generated in either a process area or a secure process area. A measured discard area receives discards, the input being measured. The discard area can be the discard point, or it can be a staging area on the way to discard. In either case, the values of SNM recorded for discard are based on the same measured values as are used to book the SNM into the area. A storage area is used for feed, product, or intermediate product, and receipts and removals are based on the same measured values. No MUF is possible from a measured discard area or a storage area

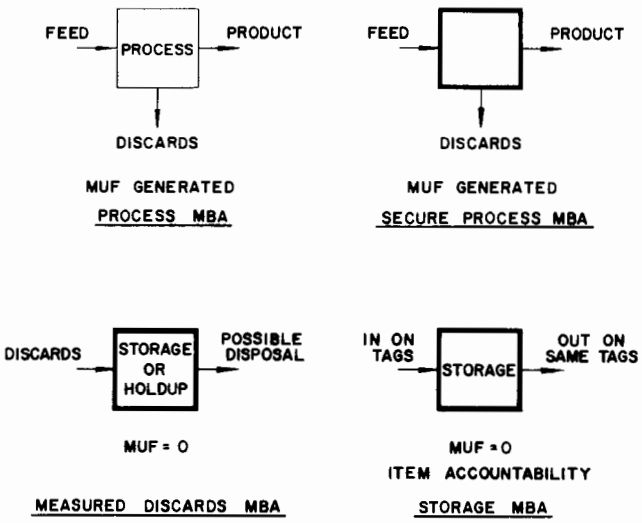

FIG. 2. Types of MBA.

unless a diversion has taken place. The difference between this scheme for MBAs and that used by the IAEA is simply the result of the difference between procedures proposed for use by the IAEA and those that can be used by the safeguards staff of a facility.

The use of physical protection and containment for secure process areas and secure storage provide a basis for extending the period between physical inventories of these areas, as indicated earlier.

Values of LEMUF have been calculated for a subdivision of the larger low enriched oxide fabrication facility listed in Table II. The results are shown in Table III.

TABLE III. LEMUF FOR PARTITIONED UO_2 FUEL FABRICATION FACILITY[a]

MBA	Type	Critical time (months)	LEMUF (kg ^{235}U)
UF_6 receiving	Storage	1	0[c]
Co-precipitation	Secure process	2[b]	5.10
Ceramic	Process	1	5.04
Product storage	Storage	1	0[d]

[a] 2.2 tonnes/d production rate, 2.12% ^{235}U (averaged).

[b] Critical time extended because of assumed secure area.

[c] LEMUF = 0; LE of inventory = 0.77 kg ^{235}U.

[d] LEMUF = 0; LE of inventory = 2.21 kg ^{235}U, based on assumed four-month residence of uranium in the plant.

The analysis described above has focused on calculations of LEMUF over relatively short balance periods. This quantity is important as representative of the size of diversion or loss that could lead to an excessive level of MUF over the accounting period. Excessive MUF would be a condition indicating a need for investigation. The value of LEMUF formed over a short period of time has large contributions from inventory terms and random errors. As the period for forming LEMUF is lengthened, the effects of inventory terms and random errors become small compared to the effects of flow terms and systematic errors. The net result is that tests for long-term MUF are much more sensitive than might be expected simply from extending the results in Tables II and III over longer periods. Propagation of anticipated systematic errors over long periods of time leads in most cases to a sensitivity per unit time substantially better than that over short periods.

These studies of procedural safeguards have demonstrated that a structuring of the system along the lines of threshold values and critical times is workable. It has been concluded that while criteria based on the assumed threshold values cannot be met in all cases now, especially for some larger facilities, introduction of the better measurement methods now available would permit meeting the criteria in a reasonably short time. Better measurement methods would, in fact, permit using more stringent criteria.

Future Studies

Future analytical studies will concentrate on the following areas:

1. Extension of the analysis to other facilities, particularly AEC-owned, contractor-operated ones.

2. Re-analysis to take into account the advantages offered by the rapid methods of nondestructive assay that have been generated by the research and development program. These methods should improve timeliness of measured data. They should also lead to reducing the systematic errors attached to some measured data, at the minor disadvantage of some increase in the random error.

3. Improved inspection strategies, and the interaction between facility safeguards and inspector activities.

4. The development of methods of information transmittal, storage, and analysis, to take advantage of the possibilities offered by safeguards structured on the lines described in this paper.

5. Extension of analysis to include effects of IAEA activities under the Presidential Offer.

6. Other possible choices of threshold values and critical times.

REFERENCES

[1] CROWSON, D.L., "The U.S. program – its relationship to international safeguards", presented to the 9th ANL-AUA Faculty-Student Conf., 21 August 1970.

[2] USAEC rules and regulations, 10 CFR 50 Appendix A.

[3] INTERNATIONAL ATOMIC ENERGY AGENCY, Final Report of IAEA Safeguards Technical Working Group on Interim Determination of Design Information Requirements, IAEA, Vienna, 13-17 April 1970.

[4] MORGAN, F., Report to the Director General of the IAEA by the Consultants on Criteria for Safeguards Procedures, IAEA, Vienna, Topic 1 (1969).

[5] BROWN, F., "The design of a safeguards material control system", Safeguards Techniques (Proc. Symp. Karlsruhe, 1970) 1, IAEA, Vienna (1970) 273.

[6] SUZUKI, G. et al., "Safeguards accountability analysis in a nuclear fuel processing plant", Safeguards Systems Analysis of Nuclear Fuel Cycles, USAEC Rep. 1 October 1969.

[7] MARCUSE, W., WILLIAMS, J.M., "Application of a nuclear process simulation model to an integrated safeguards experiment", Safeguards Techniques (Proc. Symp. Karlsruhe, 1970) 2, IAEA, Vienna (1970) 351.

[8] MARCUSE, W., "Preliminary conceptual design for safeguards", Internal TSO study, 3 March 1971.

A REVIEW OF THE NATIONAL SAFEGUARDS SYSTEM IN JAPAN AND ITS SYSTEMS STUDY

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Abstract–Résumé–Аннотация–Resumen

A REVIEW OF THE NATIONAL SAFEGUARDS SYSTEM IN JAPAN AND ITS SYSTEMS STUDY.

The first part of this paper summarizes the present situation on the implementation of the national safeguards system in Japan and on the safeguards system of the International Atomic Energy Agency based on bilateral agreements. Since the establishment of the Atomic Energy Basic Law in 1955, the Japanese government has succeeded in ensuring that nuclear material in Japan is used only for peaceful purposes. For achieving this purpose, the Law for Regulation of Nuclear Material and Reactors was established. According to that Law, several control measures were carried out on all nuclear material in Japan, including the design review of the nuclear facilities, the records and reports system as well as verification by the Japan Atomic Energy Bureau. Also, the IAEA is safeguarding some nuclear material and facilities transferred from the United States of America, the United Kingdom and Canada in accordance with the provisions of bilateral cooperation agreements for the peaceful uses of atomic energy. This paper also summarizes Japanese policy on the rationalization of the safeguards system for Japan with relation to the NPT, and outlines the research and development activities on safeguards technology in Japan.

The second part of this paper summarizes the systems study on the national safeguards system. The aim of this study is to evaluate the control system for nuclear material from the standpoint of cost effectiveness. As a basis for the systems approach, a macro-model of the entire national fuel cycle in Japan from 1970 to 1980 was constructed. Then a micro-model on the flow of nuclear material in the individual facilities was constructed. Using the output data of this model, the flow of materials through strategic points and the inventory in the facility were constructed. Based on the output of these models, the costs and effects of various safeguards measures were analysed. Combining the above results, a cost-effectiveness analysis was studied for the proposed national control system of nuclear material with special consideration given to a consistent level of effectiveness in different facilities.

EXAMEN DU SYSTEME NATIONAL DE GARANTIES APPLIQUE AU JAPON ET DE L'ETUDE DE SYSTEMES SUR LEQUEL IL SE FONDE.

La première partie du mémoire expose succinctement la situation actuelle en ce qui concerne la mise en œuvre du système national de garanties au Japon et celle du système de garanties de l'Agence internationale de l'énergie atomique, sur la base d'accords bilatéraux. Depuis l'entrée en vigueur de la loi fondamentale sur l'énergie atomique de 1955, le Gouvernement japonais est parvenu à assurer l'utilisation exclusivement pacifique des matières nucléaires existant au Japon. C'est à cette fin qu'a été promulguée la loi sur le contrôle des matières et des réacteurs nucléaires. En vertu de cette loi, toutes les matières nucléaires se trouvant au Japon sont soumises à plusieurs mesures de contrôle, qui comprennent l'examen des plans des

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installations nucléaires, un système de comptabilité, la présentation de relevés et de rapports ainsi que des inspections par le Bureau de l'énergie atomique du Japon. L'Agence exerce aussi un contrôle sur certaines matières et installations nucléaires qui ont été transférées des Etats-Unis d'Amérique, du Royaume-Uni et du Canada au Japon, conformément aux dispositions d'accords de coopération bilatéraux relatifs aux applications pacifiques de l'énergie atomique. Le mémoire expose aussi succinctement la politique générale suivie par le Japon pour rationaliser le système de garanties appliqué par le Japon dans le cadre du Traité sur la non-prolifération des armes nucléaires et décrit les études faites au Japon dans le domaine de la technologie des garanties.

La deuxième partie du mémoire est consacrée à un exposé de l'étude de systèmes sur l'application des garanties à l'échelon national. L'objet de cette étude est d'évaluer l'efficacité du système de contrôle des matières nucléaires par rapport au coût. Pour l'étude de systèmes, on a mis au point un macro-modèle de tout le cycle national du combustible au Japon de 1970 à 1980. On a ensuite établi un micro-modèle du courant des matières nucléaires dans les différentes installations. En utilisant les données de sortie de ce modèle, on a établi le courant de matières aux points stratégiques, ainsi que l'inventaire de l'installation. Sur le base des résultats obtenus à l'aide de ces modèles, on a analysé les coûts et les effets de diverses mesures de garanties. En combinant les résultats susmentionnés, on a procédé à une analyse des coûts du système de contrôle des matières nucléaires national proposé, en tenant particulièrement compte de la nécessité d'assurer un degré constant d'efficacité dans diverses installations.

НАЦИОНАЛЬНАЯ СИСТЕМА ГАРАНТИЙ В ЯПОНИИ И ЕЕ ИССЛЕДОВАНИЕ.

В первой части доклада разъясняется современное положение дел в области национальной системы гарантий Японии и возможностей распространения гарантий Международного агентства по атомной энергии (МАГАТЭ), которые основываются на двусторонних соглашениях. Приняв в 1955 году Основной закон об атомной энергии, наше правительство добилось того, что ядерные материалы в нашей стране используются только в мирных целях. Чтобы обеспечить достижение этих целей, был принят Закон о наблюдении за ядерными материалами и реакторами. В соответствии с последним законом были проведены многочисленные меры контроля, включающие изучение проектов ядерных установок, систему регистрации, отчетности и проверки в Бюро по атомной энергии Японии. Более того, МАГАТЭ дает гарантию на ядерные материалы и установки, которые были получены из США, Англии и Канады в соответствии с условиями соглашений о двустороннем сотрудничестве по мирному использованию атомной энергии. Далее разъясняется политика Японии по рационализации системы гарантий страны в связи с Договором о нераспространении ядерного оружия, очерчены контуры исследований и описаны предпринятые меры по развитию методологии гарантий в Японии.

Во второй части доклада приведены итоги исследования национальной системы гарантий, цель которого состояла в оценке системы наблюдения за делящимися материалами с точки зрения затрат на ее содержание и эффективности. Для общего изучения методов системы гарантий была создана макромодель всего топливного цикла Японии с 1970 года по 1980 год. Затем была построена микромодель движения ядерных материалов в отдельных установках. С помощью выходных данных этой модели рассчитано движение ядерных материалов как со стратегической точки зрения, так и с точки зрения учета на установках. Основываясь на указанных моделях, проанализировали издержки и эффективность различных мер, встречающихся в системе гарантий. В результате совместного анализа вышеописанных поисков издержек и эффективности предлагаемой национальной системы контроля ядерных материалов найдено, что она вполне эффективна для контроля отдельных установок.

REVISION DEL SISTEMA NACIONAL DE SALVAGUARDIAS EN EL JAPON Y APLICACION AL MISMO DEL ESTUDIO DE SISTEMAS.

Esta memoria resume, en su primera parte, la situación actual en relación con la aplicación del sistema nacional de salvaguardias en el Japón y del sistema de salvaguardias del Organismo Internacional de Energía Atómica, basada en acuerdos bilaterales. Desde la promulgación de la Ley Básica sobre Energía Nuclear en 1955, el Gobierno japonés ha garantizado satisfactoriamente que los materiales nucleares se utilizan en el Japón solamente para fines pacíficos. Para asegurar su objetivo, se promulgó la Ley sobre Regulación de los Materiales Nucleares y los Reactores. Según esa Ley, se aplicaron a todos los materiales nucleares del Japón diversas medidas de control, incluyendo la revisión del diseño de las instalaciones nucleares, de los sistemas de registros e informes y una verificación adecuada a cargo del Japan Atomic Energy Bureau. Además el OIEA se encarga de las salvaguardias de ciertos materiales e instalaciones nucleares procedentes de los Estados Unidos, el Reino Unido y el Canadá, de acuerdo con las cláusulas de los acuerdos de cooperación bilateral para la utilización de la energía atómica con fines pacíficos. En esta memoria se resumen también las medidas adoptadas sobre la racionalización del sistema de salvaguardias para el Japón.

en relación con el Tratado sobre la no proliferación, y se resaltan las actividades de investigación y desarrollo realizadas en el Japón sobre la tecnología de las salvaguardias.

La parte segunda de esta memoria resume el estudio de sistemas aplicado al sistema nacional de salvaguardias. El objeto de este estudio es evaluar el sistema de control de los materiales nucleares desde el punto de vista del costo-eficacia. Como base para enfocar los estudios de sistemas, se contruyó un macromodelo de todo el ciclo nacional del combustible en el Japón entre 1970 y 1980. Como paso siguiente, se construyó un micromodelo del flujo de los materiales nucleares en las diversas instalaciones. Utilizando los datos de salida de este modelo, se establecieron el flujo de los materiales a través de puntos estratégicos y el inventario de la instalación. Con los datos de salida de estos modelos, se analizaron los costos y efectos de diversas medidas de salvaguardia. Combinando los resultados anteriores, se estudió el análisis costo-eficacia para el propuesto sistema nacional de control de materiales nucleares, prestándose especial consideración a la consecución de un grado uniforme de eficacia en las diversas instalaciones.

INTRODUCTION

Recently, the execution of safeguards for ascertaining the use of nuclear materials for peaceful purposes has become a serious problem because of the increasing movement and transfer of nuclear materials necessitated by the wide-spread peaceful utilization of atomic energy. Moreover, the International Atomic Energy Agency's (IAEA) safeguards to be carried out under Article III of the Treaty on the Non-Proliferation of Nuclear Weapons (NPT) have come to be of great world-wide importance since the NPT came into force. Japan enacted the Atomic Energy Basic Law as the fundamental law in this area at the start of its development of nuclear energy, the use of which has been restricted to peaceful purposes only. Because Japan does not have uranium resources, it relies mainly on importing from overseas the uranium required for the Japanese nuclear power stations, which are increasing rapidly. Up to now, imported nuclear materials were safeguarded under bilateral agreements with the United States of America, the United Kingdom and Canada, but the right of safeguarding has now been transferred to the IAEA.

Japan has had long experience in implementing its national safeguards system as well as the IAEA safeguards, has improved the safeguards system considerably and will also establish national policy for future modification of the system.

The first part of this paper describes this experience and policy in general, while the second part reports results of a systems study which has been conducted in Japan on the importance of applying the systems approach to a study of safeguards.

1. THE HISTORY AND PRESENT SITUATION OF THE SAFEGUARDS SYSTEM IN JAPAN

Japan established its fundamental policy on the development and utilization of atomic energy in Japan by the enactment of the Atomic Energy Basic Law in December 1955. According to this Law, fundamental policy is defined so that "the development and utilization of nuclear energy must be performed for peaceful purposes only, in a democratic way and independently, and its achievement must be open to promote international cooperation". Based on this Basic Law, the Law for the Regulation of Nuclear Raw

Material, Nuclear Fuel Material and Nuclear Reactors (Regulation Law) has been legislated. This Regulation Law includes provisions on the safeguards of nuclear materials, health and safety and the prevention of accidents.

1.1. The present situation of the national safeguards system under the Regulation Law

1.1.1. Introduction and design review

The quantity of nuclear materials in Japan increased rapidly in recent years, as shown in Table I. Especially noteworthy is the enormous increase of plutonium, enriched uranium and depleted uranium; for instance, plutonium increased 210 000 times and enriched uranium 1200 times in the past 10 years. Consequently, the number of facilities which hold nuclear materials also increased, as well as the frequency of the import or export of nuclear materials. Therefore, it is increasingly necessary that the effectiveness of national safeguards be improved.

The first step to be taken in carrying out the national safeguards system is the licensing of nuclear enterprises, nuclear facilities construction and/or use of nuclear materials. When an applicant wants to obtain such a licence, he must submit the necessary design information concerning the facilities using the nuclear materials. The Atomic Energy Commission of Japan reviews this information.

TABLE I. TOTAL AMOUNT OF NUCLEAR MATERIALS IN JAPAN

Year	Natural uranium (t)	Depleted uranium (t)	Total enriched uranium (t)	Enriched ^{235}U (t)	Plutonium (kg)	Thorium (t)
1961	57	0.1	0.2	0.0	0	1
1962	75	0.1	5	0.1	0.1	4
1963	89	0.2	13	0.3	0.5	5
1964	301	2	16	0.3	0.6	5
1965	300	2	17	0.4	5	5
1966	304	2	18	0.5	13	4
1967	355	31	18	0.6	165	6
1968	379	71	21	0.9	240	5
1969	386	95	121	3	321	10
1970	358	202	224	5	644	22

The Regulation Law governs ore-processing, fuel fabrication, reactors, reprocessing and other facilities using nuclear materials. Examination of design information is done as follows.

Design information must be examined from the point of view of ascertaining the peaceful uses of nuclear material, i.e. safeguards, health and safety as well as accident prevention. Design information concerning ore-processing plants, fabrication plants and reactors must be submitted twice, once for the business licence and once for the approval of the design and construction methods.

The construction of reprocessing plants is prohibited except for the Japan Atomic Energy Research Institute (JAERI) and the Power Reactor & Nuclear Fuel Development Corporation (PNC), so, legally, licences to operate a business do not exist and the examination of design information only takes place at the time of approval of the design and construction methods.

For other nuclear materials handling facilities there is no procedure for approval of design and construction methods, but design information has to be submitted for examination upon requesting a licence to use nuclear materials.

1.1.2. Records and reports

The system for safeguards records and reports is laid down in the Regulation Law. The requirements for records are established in the Prime Minister's Order under the Regulation Law, and records must be kept in the facility on the following: (1) nuclear material accounting, (2) radiation control and waste disposal, (3) operation, (4) maintenance, (5) accidents, (6) inspection of facilities and (7) other items (for example, meteorological records). The user of nuclear materials must send to the Science & Technology Agency various reports based on such records. The nuclear-material accounting reports for each facility are periodical and ordinarily submitted twice a year, once at the end of June and again at the end of December, but for facilities such as power reactors and fabrication plants reports are submitted monthly. Data in these reports include the input and output of the facility, nuclear production in the facility, losses, and material unaccounted for (MUF) for various nuclear materials (enriched uranium, natural uranium, depleted uranium, plutonium, thorium, and ^{233}U). The maximum time delay permitted between taking a physical inventory, with closing of the material balance, and submitting the material balance report to the Science & Technology Agency is 30 days.

The most important of the non-periodical reports is that dealing with the receipt or delivery of nuclear materials; this report deals with the input to or output of nuclear materials from the facility on a batch basis. Its maximum time delay is 15 days. The national control authority, which is the Atomic Energy Bureau of the Science & Technology Agency, can control the movement and flow of nuclear materials in Japan by the 15 days' time delay of these reports.

Also, there are several kinds of irregular reports. Some of them are accident reports, reports concerning the theft, loss or MUF within certain permissible limits, and radioactive waste disposal in the sea. The reports concerning loss and MUF are very important from the standpoint of safeguards implementation.

1.1.3. Verification

Verification, including inspection, is the most important procedure from the point of view of the effective execution of safeguards. Verification includes inspection at the facilities and checking the consistency of reports at the administrative centres.

It is important to make inspection effective by taking into consideration the characteristics of the fuel cycle in the country, as well as the characteristics of the facilities in the fuel cycle, social conditions, etc.

In Japan's case, the utilization and development of nuclear energy have been restricted from the start to peaceful purposes only, and this idea has been firmly established throughout Japan. Accordingly, under present social conditions, it is hardly likely that the diversion of nuclear materials into unauthorized purposes would occur in Japan.

The first verification step under the Regulation Law is the verifying of design information. For the facilities important for safeguards, such as reactors, fabrication plants and so on, the inspection of the facility is necessary for the approval of the design and construction method. Thereafter, regular inspection takes place once a year in the case of reactors. and the Science & Technology Agency has 15 nuclear facility inspectors for performing inspections. Moreover, based on the Regulation Law, the right of inspection is given to the inspectors of the Science & Technology Agency, including the right of access at all times to all places. In March 1969, the Science & Technology Agency established a Safeguards Office in its Atomic Energy Bureau for implementing the safeguards procedures and improving the safeguards system.

1.2. The IAEA safeguards based on bilateral agreements

Since 1955, Japan has concluded various bilateral cooperation agreements with several countries for the peaceful uses of atomic energy. Japan has had two reasons for concluding these agreements: one is to have access to uranium resources, because Japan has nearly no uranium, the other is to obtain technical information as well as research reactor services at the initial stage of development. The first cooperation agreement was the Japan-United States Cooperation Agreement for Research in Atomic Energy of 1955, the next was the first Japan-United States Cooperation Agreement for Peaceful Uses of Atomic Energy of 1958, followed by the Japan-Canada Cooperation Agreement of 1959, the Second Japan-United States Cooperation Agreement of 1968 and the Japan-United Kingdom Cooperation Agreement of the same year. The nuclear materials which were imported into Japan under such bilateral agreements are under the safeguards of the suppliers' country, but such rights were transferred to the IAEA by the Trilateral Safeguards Agreements between the Agency, Japan and the United States of America, etc. When the IAEA was established, Japan concluded the first project agreement with the Agency for the supply of nuclear materials, because Japan wished to help promote the IAEA's activities through such an agreement. By this agreement, Japan obtained for a research reactor, "JRR-3", 3 tonnes of natural uranium, which were safeguarded by the IAEA. Japan has thus had long experience with IAEA safeguards. When the Japan-United Kingdom-IAEA Transfer Agreement came into force in 1968, the

TABLE II. NUMBER OF FACILITIES UNDER IAEA SAFEGUARDS
(Situation in 1970)

Category	In the world (A)	In Japan (B)	(B/A) (%)
Power reactors	9	3	33
Reprocessing plants	3	0	0
Fuel fabrication plants	5	3	60
Experimental and research reactors	54	12	22
Critical assemblies	9	9	100
Research and development facilities	19	13	68
Other locations	57	57	100
Total	156	97	62

IAEA applied its safeguards to a commercial power reactor in Japan, the first of such safeguards in the world. This reactor is the Tokai Power Reactor of the Japan Atomic Power Co., the output of which is 166 MW(e). Thereafter, the number of facilities which were subjected to IAEA safeguards increased gradually with the addition of several power reactors and fuel fabrication plants in Japan.

At present, the number of facilities safeguarded by the IAEA is as follows:

Reactors:	24
Fabrication plants:	3
Research and development facilities:	13
Miscellaneous installations:	57
Total:	97

From this and Table II it can be seen that Japanese safeguarded facilities represent 62% of the facilities which the IAEA is safeguarding all over the world, as the miscellaneous installations are all located in Japan. Thus the IAEA safeguards have been applied on such a broad basis in Japan that all plutonium and enriched uranium in Japan are safeguarded by the IAEA, as shown in Table III. Therefore, Japan has to be most careful that safeguards does not hamper industrial activities. The reports which have to be sent to the IAEA from Japan are material accounting reports and operating reports (twice a year, but monthly in the case of power reactors and fabrication plants), joint notifications of international transfer of nuclear materials, special reports, including the transfer reports of large amounts of nuclear materials in Japan, and reports on the loss and MUF over a certain permissible limit. Japan now has 8 facilities for which the material accounting and operating reports have to be made monthly. Besides

TABLE III. THE PRESENT SITUATION OF IAEA SAFEGUARDS OF NUCLEAR MATERIALS IN JAPAN (as of Dec. 1970)

Category	Amount in Japan (A)	Amount under IAEA safeguards in Japan (B)	(B/A) (%)
Natural uranium (kg)	358 149	346 972	97
Depleted uranium (kg)	202 388	201 603	100
Enriched uranium (kg)	223 677	223 677	100
Plutonium (g)	644 371	644 391	100
Thorium (kg)	21 889	269	1

TABLE IV. THE FREQUENCY AND INTENSITY OF IAEA INSPECTION FOR JAPAN

Year	Frequency of visits to Japan	Man-days in Japan	Number of inspections at facilities
1964	2	23	7
1965	2	48	7
1966	2	28	6
1967	3	50	12
1968	5	111	15
1969	6	186	34
1970	6	286	55
1971 (up to the end of Feb.)	2	76	24
Total	28	808	160

these, Japan has many facilities for which reports must be made semi-annually and, since the nuclear materials are administered according to the country of origin, the reports reach an enormous volume. The number of joint notifications required for the international transfer of nuclear materials was 68 in 1970 and there were 11 advance notices for the international transfer of large quantities, making the total number 79. Meanwhile, the frequency of inspections conducted by IAEA inspectors has increased every year since May 1964, when IAEA inspectors came to Japan for the first time, making a total of 28 visits to Japan by the end

of March 1971. Their total number of man-days in Japan has been about 800 days and the total number of inspections at facilities so far has been 160 (see Table IV).

1.3. Japan's effort for the rationalization of the safeguards system

1.3.1. National policy

As mentioned above, it is necessary for Japan that the safeguards system be rationalized so that safeguards do not hamper the peaceful uses of atomic energy, which are increasing rapidly. This applies especially to Japan, which has its own effectively operating safeguards system. Therefore, the IAEA safeguards system has to be based on and utilize such existing national systems as an important factor to optimize the cost effectiveness of safeguards by the IAEA. From this point of view, Japan has had deep concern over the way safeguards are applied under the NPT. When Japan signed the NPT on 3 February 1970, the Japanese Government made a statement which included the following opinions.

(1) The content of the Safeguards Agreement which will be concluded between Japan and the IAEA must not be unadvantageous compared with Safeguards Agreements concluded between the IAEA and other party or parties.
(2) The safeguards under the NPT should be based on the principle that safeguards are applied at the strategic points in the fuel cycle and their procedures should be rational considering the principle of cost vis-à-vis effect and be as simplified as possible, utilizing each country's own system, if any, as far as possible. Moreover, careful considerations should be given so that leakage of industrial secrets and know-how will be prevented and that industrial activities will not be hampered.

Comment on four other subjects was also given in this statement by the Japanese Government. Following up the policy summarized above, Japan has actively participated in the discussions of the Safeguards Committee of the IAEA since June 1970, and sent to the Director General of the IAEA in May 1970, in response to his invitation, the views of the Japanese Government, which were almost the same as those of the statement in February 1970. On the other hand, the Japan Atomic Energy Bureau set up a special committee for safeguards matters, consisting of members from government departments and private industries, and of academic and experienced experts for studying these matters. At the IAEA, the Safeguards Committee finished its task by submitting the report to the Board of Governors in April 1971. Japan appreciates the efforts put into the formulation of the Committee's report, including the model Safeguards Agreement under the NPT, to be used as the basis for future more rationalized and effective safeguards based on the new concepts.

1.3.2. The implementation of research and development of a safeguards technique

It is clear that the research and development of a safeguards technique is very effective for the rationalization of safeguards. Japan has been engaging in such research and development work since 1968. As the technological development work of safeguards is in the interest of every country,

the Government and governmental organizations are financially supporting such development work. In 1968, three subjects were studied under such Government financing. Two of these subjects were concerned with research aimed at the improvement of material accounting methods in the fuel fabrication plants; these were (1) the production process control of highly-enriched plate fuels and (2) the management system of the fabrication process of dioxide uranium pellets. The results of these two works were submitted to the IAEA free of charge as a Japanese contribution. Another subject dealt with the assessment method of nuclear fuel materials in gas reactors for comparison of the assessment by burnup with the results of destructive analysis. Since 1968, there have been two types of research in Japan, namely, the development of software and of hardware. In research on software, the study of the accounting system in fabrication plants has been going on since 1968. Also PNC carried out a study on material accounting and control systems in reprocessing plants. Since 1970, a systems analysis study has been carried out on the inspection model under the sponsorship of the Government. The results of this study are given in Section 2 of this paper. In hardware research, the main efforts have been directed to the development of methods and equipment for non-destructive inspection. PNC and JAERI are also conducting a study of a non-destructive analysis method. Since such technological development is being pursued vigorously not only in Japan but also abroad, Japan considers it very important to exchange technical information with foreign countries as much as possible and to carry out research and development work on selected subjects which would contribute to the rationalization of safeguards.

2. SYSTEMS ANALYSIS APPROACH TO SAFEGUARDS

2.1. Macro-model

2.1.1. Objectives

It is essential for designing an optimal safeguards system applied to various nuclear facilities to understand the scale and characteristics of the country's nuclear fuel cycle. First of all, the intensity of a safeguards operation for each facility will be decided by the accessibility of the material for production of nuclear explosives. Secondly, when one takes into consideration the expected rapid increase in the scale of nuclear fuel cycle systems, it is important to identify clearly the flow of nuclear materials in the fuel cycle. Thirdly, safeguards operations should be applied with a rational scheme which gives consistency in each facility. For these objectives, a computerized "macro-model" of the total fuel cycle was constructed which will give the characteristics and the scale of the fuel cycle in Japan for the next ten years.

2.1.2. Procedure

Nuclear facilities are classified into three categories: conversion and fabrication plants, nuclear power plants, and reprocessing plants. The fabrication plants are classified according to the kind of fuels treated,

uranium or plutonium. Power plants are classified by reactor types. The plants which have been taken into account are limited to fabrication plants, power plants, and reprocessing plants, which are operating or included in the present construction program.

First, typical operation specifications are decided for each plant, and then the amount of materials treated per month in the plant is calculated. Next, a material balance is taken for the three categories of plants. The import and export of materials are estimated from the excess and deficit of the balance. Some computer calculations have been performed using this model (see Figs 1 a and b).

2.2. Micro-model

2.2.1. Objectives

It is essential to have information on the operations and flow of materials in each facility in order to study safeguards inspection modes and patterns. For this, a computerized model which will simulate the operation of a nuclear facility and provide the necessary data is constructed as a "micro-model". As a first attempt, a simulator of a reprocessing plant, which is not yet in existence in Japan, has been constructed.

2.2.2. Procedure

The reprocessing plant is of the Purex-type, and the operation can be simulated with a schedule corresponding to the various amounts, compositions, and enrichments of the nuclear fuels treated. The process flow-chart and measuring points are described in detail in Ref. [1].

The calculation procedure is as follows: The output of the macro-model is stored; the input consists of, among other things, the data specifying the name of the facility (two facilities are expected to operate by 1980) and the date of operation. The flow data at each measuring point is calculated as the output by the procedure shown in the flow-chart in Fig. 2. Random errors and systematic errors are included in this output data. This micro-model will also be useful in designing a computerized data-processing system in each facility.

2.3. Inspection pattern

2.3.1. Parameters representing inspection pattern

The following parameters are considered in characterizing the inspection pattern to make a quantitative evaluation of the effectiveness and cost associated with it:

(a) Inspection authority
(b) Frequency
(c) Mode of routine inspection
(d) Timing
(e) Number of strategic points
(f) Number of inspectors
(g) Duration

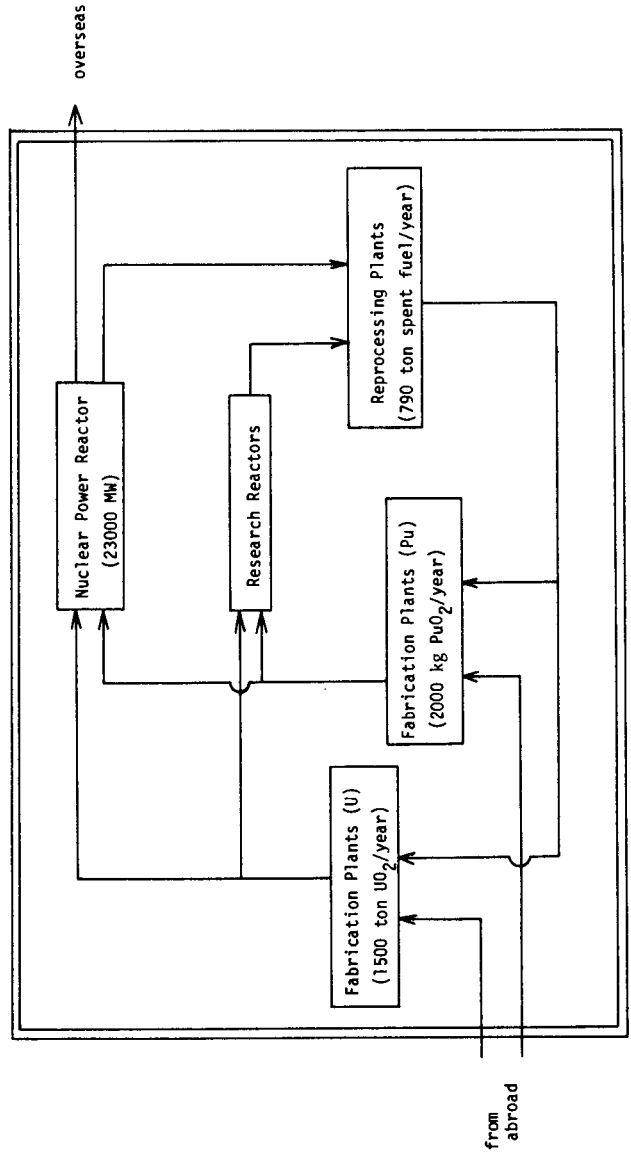
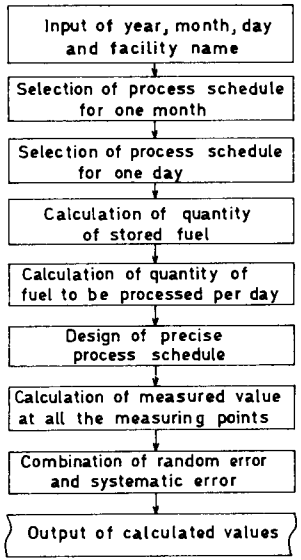


FIG. 1a. Fuel cycle of Japan in 1980.

Item			Calendar Year										
			70	71	72	73	74	75	76	77	78	79	80
Amount of Fuel (ton)	Input to Reactors		158	88	156	287	454	692	658	793	1133	1314	1391
	Output from Reactors	UO_2	53	87	87	110	169	262	403	477	587	765	945
		PuO_2	0.34	0.56	0.56	0.71	1.09	1.58	2.60	3.08	3.78	4.93	6.10
		UO_2 +PuO_2	53	88	88	111	170	264	406	480	591	770	951
	Contained in Reactors		181	181	250	426	709	1135	1385	1686	2225	2766	3204
Capacity of Electric Power (MW)			1323	1323	1823	3107	5177	8287	10113	12313	16239	20189	23389

FIG. 1b. Amount of fuel in Japan.



(a)

Input	1975 (year), 6 (month), 5 (day)				
	Measuring point	Batch / d	Volume, weight or piece [b]	U concentration	Pu concentration
Output (Example)	From power reactor	--	0 P	--	--
	Spent fuel storage pond	--	45 P	--	--
	From spent fuel storage fond	--	5 P	--	--
	Adjusted feed	2	2395 1/B	200 g/1	1.34 g/1
	From U concentration & denitration (UO_3)	2	474 kg/B	0.86 g/g [c]	--
	From Pu concentration	1	25 1/d	--	250 g/1
	U product	--	961 kg	0.86 g/g	
	Pu product	--	6.4 kg		250 g/1
	U product storage	--	4740 kg	0.86 g/g	--
	Pu product storage	--	31.25 kg	--	250 g/1
	U product shipping	--	0 kg	0.86 g/g	--
	Pu product shipping after storage	--	0 kg	--	250 g/1
	Pu product shipping before storage	--	0 kg	--	250 g/1
	Waste [a]	--	12.1 kg(U) 90 g (Pu)	--	--

[a] For convenience, weight of waste is shown by summing up various waste flows.

[b] P and B represent piece and batch, respectively

[c] g/g is grams of U per gram compound

(b)

FIG. 2. (a) Basic flow chart of the micro-model for a reprocessing plant.
(b) Example of the micro-model output.

Item (a) varies by the way the national authorities take part in the inspection. With item (c), various levels are considered corresponding to the degree the inspection relies on visual inspection or inventory-taking, especially when material balancing is considered.

2.3.2. Effectiveness

According to the objective of safeguards, the effectiveness can be defined as the degree, or probability, that nuclear material is prevented from diversion to non-peaceful uses. This has two aspects: technical effectiveness and psychological effects.

The basic concept for quantifying the technical effectiveness to deter diversion is as follows: the technical effectiveness is the degree of deterrence which results from the variation of technical measures in an inspection pattern. This corresponds to the degree that an increase of inspection frequency or accuracy of mode of routine inspection can increase the probability to detect the MUF which can be related to diversion.

The accuracy can be represented by γ, which can be set equal to σ_{MUF}. On the other hand, when the frequency of inspection is n times/yr and the annual throughput of a facility in effective kg is a, a/n describes the amount in kg of nuclear material treated in that MBA (material balance area) during the period between two inspections. Then,

$$E_t = 1\Big/\left(\frac{a}{n}\cdot\gamma\right); \quad a = A/A_0 \tag{1}$$

E_t, the technical effectiveness of safeguards, can be defined as the above evaluation equation (1).

Regarding the psychological effect, it can be considered to be closely related to the intensity of inspection associated with each facility. The required intensity of inspection can be related to the accessibility of the nuclear material in a facility to nuclear explosives and is expressed in terms of a "critical time", T_c, as follows:

$$I^+ = 1\Big/\left(\frac{k_1}{a} + T_c\right) \tag{2}$$

where I^+ is the required intensity of inspection and k_1 is a constant.

Then, the psychological effectiveness of inspection will be expressed by the following evaluation equation:

$$E_p = k_2 \cdot n/I^+ \tag{3}$$

where k_2 is a constant.

2.3.3. Costs

According to the definitions of effectiveness given above, the technical effectiveness depends on the accuracy γ and the frequency n, while the

psychological effectiveness depends on the frequency n. In fact, n corresponds to parameter (b) and γ to parameters (c), (d) and (e) in Section 2.3.1. Then, the effectiveness of inspection is decided by parameters (b), (c), (d) and (e). On the other hand, if E_t and E_p given by equations (1) and (3) are to be maintained at some predetermined levels, it is necessary to carry out some corresponding amount of effort. The amount of effort corresponds to parameters (f) and (g), and the costs of inspection can be calculated by them.

When the accuracy which has to be achieved by one series of inspections is γ, the mode of routine inspection, timing, and number of strategic points which are necessary for the realization of γ will be decided. Then the amount of effort needed will depend mainly on the product of the number of inspectors and the duration. Therefore, the necessary cost $c(\gamma)$ can also be specified.

2.4. Optimization of safeguards

2.4.1. Concept of optimization

The following concepts are considered in optimizing the safeguards system in this study: cost effectiveness and consistency. Cost effectiveness is considered as a criterion for optimizing the inspection mode of each inspection when the inspection frequency is already determined. Consistency is considered as a criterion for deciding the inspection frequency at facilities to maintain the psychological effectiveness consistent with the total fuel cycle in each facility.

Although, in principle, the cost effectiveness should be considered in optimizing the inspection frequency, it is difficult to define an inspection accuracy γ applied to the total fuel cycle, because it is almost impractical, for example, to consider a MUF at a reactor MBA other than on an item accountability basis.

2.4.2. Cost effectiveness

In optimizing the inspection mode, only the technical effectiveness is used as a criterion. Then f, which is the ratio of effectiveness/cost, is expressed as

$$f = E_t/\{n \cdot c(\gamma)\} = 1/\{a \cdot \gamma \cdot c(\gamma)\} \qquad (4)$$

The inspection mode corresponding to the γ which maximizes f is the optimal mode. In Fig. 3 an example of cost effectiveness is shown for a fabrication facility, taking the accuracy of the MUF determination as 2σ, limiting inventory-taking to normal operations, and taking the number of measurements as an optimization parameter. The cost is a direct cost estimated from the inspection data in Japan in 1970, and further work is being done on this estimate by introducing parameters such as a quantity, the "embarrassment factor".

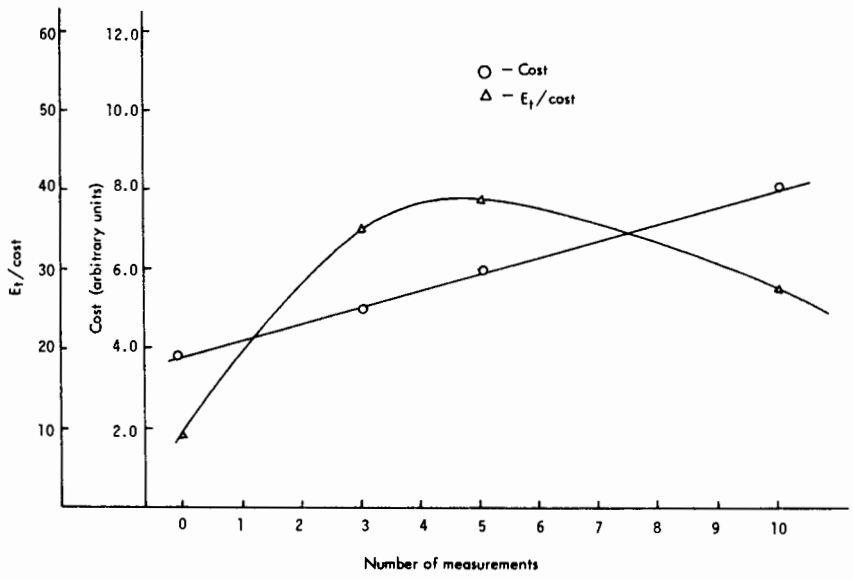


FIG. 3. Example of cost effectiveness [E(t)/cost] for a fabrication plant with ten fabrication processes.

2.4.3. Consistency

E_p, as defined by equation (3), has to be maintained at the same value for all facilities. Namely, when i represents a suffix for each facility,

$$\frac{k_2 n_i}{I_i^+} = \alpha, \qquad \alpha = \text{constant for all } i \tag{5}$$

Then, the inspection frequency is specified by

$$n_i = I_i^+(\alpha/k)$$

Here, if the value of α/k is not given, only the "relative" value of the inspection frequency for different facilities can be defined. In order to decide the value of α/k, one must know the total cost C_0 required for inspections of all facilities.

$$\sum_i n_i \cdot c_i(\gamma) = \frac{\alpha}{k} \sum_i I_i^+ c_i(\gamma) \leq C_0$$

2.5. Conceptual design of a safeguards system

There are several cases where a data-processing system is introduced for accounting and control of materials in nuclear facilities. This is expected to increase in the future, and the utilization of such a data-processing system in facilities as a part of the safeguards system will be effective in increasing safeguards efficiency and simplifying procedures.

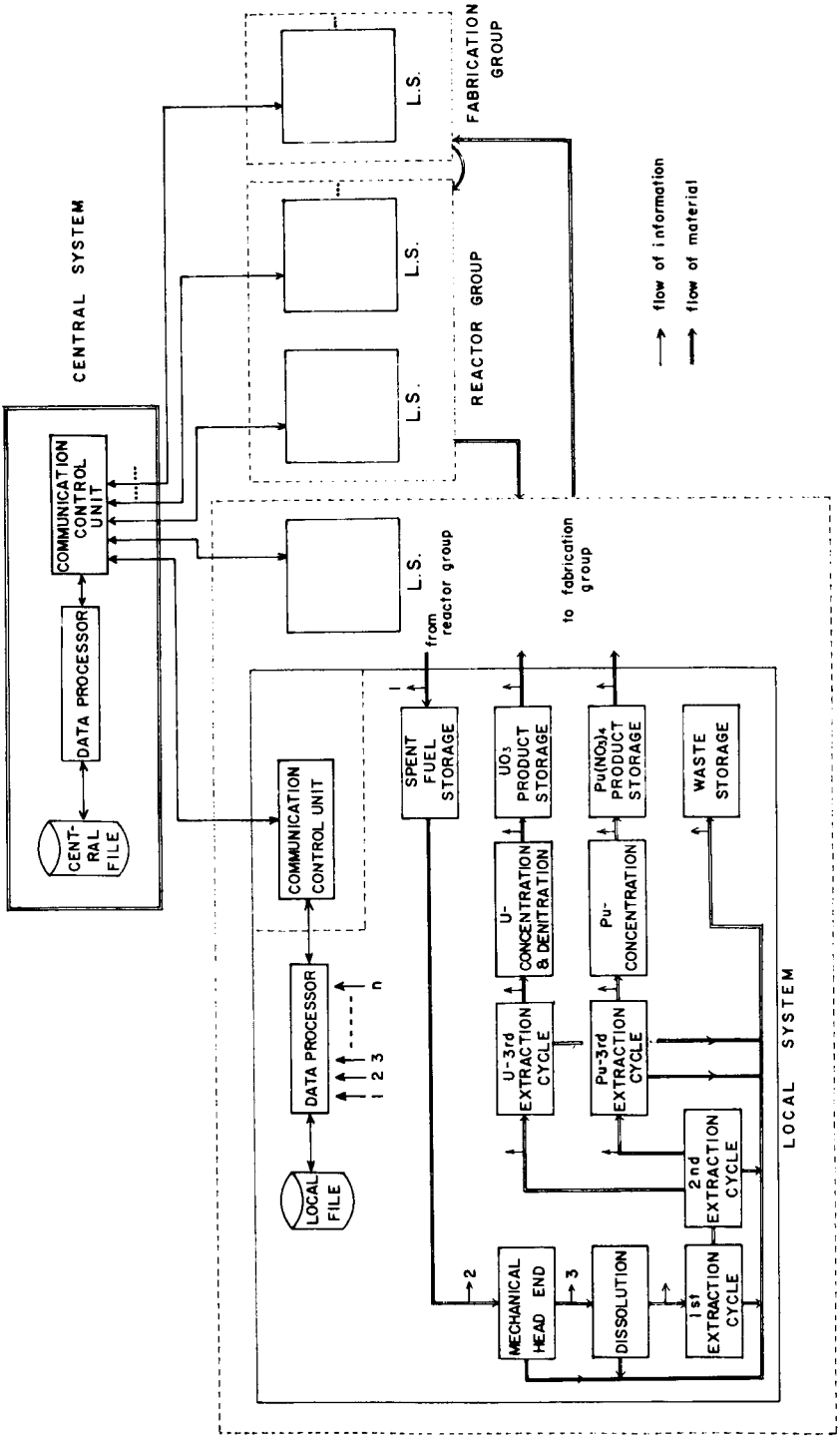


FIG. 4. Conceptual design of a data-processing system.

One conceptual design of a utilization of the data-processing system of a facility, namely a local system, is shown in Fig. 4. The local system shown in detail is that of a reprocessing plant as described in the micro-model in Section 2.2.; however, other facilities can be treated in the same manner. Data at each measuring point in the facility is transferred on-line or off-line as inputs to the data processor and stored (for example on magnetic tape or discs, etc.) in a local file. A communication control unit, added to the local system, transfers information necessary for safeguards to the central system from the local system. This unit has to be newly added to the data-processing system of the facility and will be controlled by the central system for the transfer of information. The central system will be operated by the national government or authorities working on its behalf, and will be the primary system for international safeguards in Japan.

Information transferred to the central system from the local file will be classified into several levels according to its contents. This multi-level system is decided mainly by the MBA, and the control level is improved when the MBA is divided into smaller sub-areas. When the suspicion is increased, the level will be enhanced to clarify the facts.

The advantages of such a system are: the additional work of safeguards reporting will be eliminated, delay in reporting to the safeguards authority will be reduced, and costs will decrease.

REFERENCES

[1] NAKAJIMA, K., et al., "Analysis of accountability system in a reprocessing plant and a plutonium fuel facility", Safeguards Techniques(Proc. Symp. Karlsruhe, 1970) 1, IAEA, Vienna (1970) 139.

[2] OSHIMA, K., et al., "Dynamic safeguards system for multiple action levels", Ibid., p.375.

ЧЕХОСЛОВАЦКАЯ СОЦИАЛИСТИЧЕСКАЯ
РЕСПУБЛИКА
A/CONF.49/P/558

ОПЫТ НАЦИОНАЛЬНОЙ СИСТЕМЫ УЧЕТА И КОНТРОЛЯ ЯДЕРНЫХ МАТЕРИАЛОВ В ЧССР

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Abstract–Résumé–Аннотация–Resumen

EXPERIENCE IN CZECHOSLOVAKIA OF A NATIONAL SYSTEM OF NUCLEAR MATERIAL ACCOUNTING AND CONTROL.

Czechoslovakia is a non-nuclear-weapon State, but has a relatively highly developed peaceful nuclear energy program. Since the ratification by Czechoslovakia of the Treaty on the Non-Proliferation of Nuclear Weapons a wide range of administrative and technical measures has been taken in order to adapt the Czechoslovak nuclear material control system to the requirements of the International Atomic Energy Agency. The paper describes the national system for the accounting and control of nuclear material as applied to different types of nuclear facilities in Czechoslovakia, and quotes the experience gained in drawing up an initial inventory of all nuclear materials and in delimiting the individual material balance areas. A description is also given of the reporting system and of the data processing and verification activities of the Nuclear Safety and Safeguards Department of the Czechoslovak Atomic Energy Commission. A central analytical laboratory for the precise determination of the state of nuclear material constitutes an important element in the national safeguards system. The analytical control methods applied and the types of equipment used in safeguards are described. Finally, mention is made of the way in which the national safeguards system is expected to develop in the light of the program for the expansion of nuclear power generation in Czechoslovakia.

EXPERIENCE DU SYSTEME TCHECOSLOVAQUE DE COMPTABILITE ET DE CONTROLE DES MATIERES NUCLEAIRES.

La Tchécoslovaquie est un pays qui ne dispose pas d' armes nucléaires, mais exécute un programme relativement important d' utilisation de l' énergie nucléaire à des fins pacifiques. Après avoir ratifié le Traité sur la non-prolifération des armes nucléaires, la Tchécoslovaquie a pris toute une série de mesures techniques et administratives en vue d' adapter son système national de comptabilité et de contrôle des matières nucléaires aux prescriptions de l' AIEA. Les auteurs examinent ce système du point de vue de son application aux divers types d' installations nucléaires existant en Tchécoslovaquie. En outre, ils font état de l' expérience acquise lors de l' établissement de l' inventaire initial de toutes les matières nucléaires, y compris la création des diverses zones de bilan matières. Ils décrivent les modes de présentation des rapports et l' activité du service de radioprotection et de contrôle de la Commission tchécoslovaque de l' énergie atomique, qui s' appuie sur des données librement traitées et vérifiées. Un élément important du système national de contrôle est le laboratoire central d' analyses chargé de fournir des renseignements précis sur l' état des matières nucléaires. Les auteurs décrivent les procédés d' analyse et types d' appareils utilisés aux fins des garanties. Enfin, ils esquissent l' évolution probable du système national de contrôle en tenant compte du programme tchécoslovaque de développement de la production d' énergie d' origine nucléaire.

ОПЫТ НАЦИОНАЛЬНОЙ СИСТЕМЫ УЧЕТА И КОНТРОЛЯ ЯДЕРНЫХ МАТЕРИАЛОВ В ЧССР.

Чехословакия является государством, которое не обладает ядерным оружием, но которое имеет относительно развернутую программу использования ядерной энергии в мирных целях. После того как Чехословакия ратифицировала Договор о нераспространении ядерного оружия, осуществлен целый ряд технических и организационных мероприятий с целью приспособления чехословацкой системы контроля ядерных материалов к требованиям МАГАТЭ. В докладе рассмотрена национальная система учета и контроля ядерных материалов с точки зрения их применения в различных типах ядерных установок в Чехословакии. Далее описывается опыт осуществления первоначального инвентарного учета всех ядерных материалов,

включая учреждение отдельных зон материального баланса. Описана система отчетов и деятельность отдела ядерной безопасности и гарантий Чехословацкой комиссии по атомной энергии на основании сводной обработки данных и их проверки. Важным звеном национальной системы гарантий является центральная аналитическая лаборатория для определения точных данных о состоянии ядерного материала. В докладе описаны используемые аналитические методы контроля и виды аппаратуры, применяемой для целей гарантий. В заключительной части доклада намечено ожидаемое развитие национальной системы гарантий с учетом будущей программы развития ядерной энергетики в Чехословакии.

EXPERIENCIA ADQUIRIDA CON EL SISTEMA NACIONAL DE CONTABILIDAD Y CONTROL DE MATERIALES NUCLEARES EN LA REPUBLICA SOCIALISTA CHECOSLOVACA.

Checoslovaquia es un país que, sin poseer armas nucleares, tiene un programa relativamente desarrollado de energía nuclear con fines pacíficos. Desde la ratificación por Checoslovaquia del TNP, se han tomado una serie de medidas técnicas y administrativas con objeto de modificar el sistema de control de materiales nucleares checoslovacos de acuerdo con las estipulaciones del OIEA. Esta memoria describe el sistema nacional de contabilidad y control de materiales nucleares tal como se aplica a los diversos tipos de instalaciones nucleares en Checoslovaquia. Se expone, además, la experiencia adquirida al efectuar el inventario inicial de todos los tipos de materiales nucleares, inclusive la determinación de las distintas zonas de balance de materiales, así como al establecer el sistema de informes y las actividades de recopilación y verificación del Departamento de Seguridad Nuclear y de Salvaguardias de la Comisión de Energía Atómica Checoslovaca. El laboratorio nacional de control constituye un elemento importante dentro del sistema nacional de salvaguardias, para determinar los datos exactos relativos a los materiales nucleares. Por último, se presenta un esquema de la evolución probable del sistema nacional de salvaguardias, teniendo en cuenta el futuro programa de desarrollo de la electricidad de origen nuclear en Checoslovaquia.

ВВЕДЕНИЕ

Чехословацкая Социалистическая Республика (ЧССР) является государством, которое не обладает ядерным оружием, но которое имеет относительно развернутую программу использования атомной энергии в мирных целях. ЧССР располагает достаточно широкой научно-исследовательской базой. В настоящее время готовится к пуску в эксплуатацию первая чехословацкая атомная электростанция (АЭС) с реактором мощностью 110 МВт (на тяжелой воде, с газовым охлаждением). До 1980 года предполагается построить еще две АЭС, каждая мощностью 880 МВт, с реакторами водо-водяного типа (ВВЭР-440). В долгосрочной перспективе предполагается строить АЭС именно этого типа, и общая мощность АЭС должна достичь в 1985 году приблизительно 5000 МВт, а к 1990 году – 10 000-12 000 МВт [1].

ЧССР является с самого начала членом Международного агентства по атомной энергии (МАГАТЭ). Правительство ЧССР активно поддерживает деятельность МАГАТЭ, которая направлена на развитие международного сотрудничества в области использования атомной энергии в мирных целях. Одним из условий такого развития является широкое международное признание Договора о нераспространении ядерного оружия, который ЧССР ратифицировала в числе одного из первых государств.

Учет ядерных материалов осуществлялся в ЧССР и раньше, причем особое внимание уделялось постановлениям по вопросам радиации и гигиены. В связи с Договором о нераспространении ядерного оружия указанную систему контроля и учета стали в 1967 году приспосабливать к требованиям, вытекающим из Договора.

Необходимая юридическая основа в ЧССР была заложена изданием закона № 133 от 1970 года о деятельности федеральных министерств [2]. Из этого закона вытекает, что Чехословацкая комиссия по атомной энергии (ЧСКАЭ) утверждает использование всех видов ядерных материалов,

решает вопросы экспорта и импорта, а также обеспечивает их учет и контроль в общегосударственном масштабе. ЧСКАЭ издает для этой работы на основе данного закона обязательные для исполнения директивы. В 1969 году при ЧСКАЭ был образован самостоятельный отдел, занимающийся вопросами ядерной безопасности и гарантий, которому было поручено исполнение директив и организация национальной системы учета и контроля ядерных материалов.

НАЦИОНАЛЬНАЯ СИСТЕМА УЧЕТА И КОНТРОЛЯ ЯДЕРНЫХ МАТЕРИАЛОВ

Согласно определению, содержащемуся в документе INF/CIRC/153, к материалам, подлежащим контролю, относятся: обедненный уран, природный уран, обогащенный уран, торий и плутоний.

Основу национальной системы учета и контроля составляет разрешение на распространение и применение отдельных видов ядерных материалов, находящихся под контролем. Осуществляется контроль экспорта и импорта, поэтому исключается какая-либо возможность перевоза через границу ЧССР ядерных материалов без уведомления об этом ЧСКАЭ. Ни одна организация в ЧССР не получит ядерных материалов, подлежащих контролю, без собственного запроса или без запроса в ЧСКАЭ вышестоящей инстанции. На все экспортируемые и импортируемые материалы, подлежащие контролю, должно быть получено разрешение от компетентного органа внешней торговли, причем этот орган должен согласовать данный вопрос с ЧСКАЭ.

Каждое разрешение, позволяющее владеть или применять ядерный материал, подлежащий контролю, связано с условиями, вытекающими из директив по ядерной безопасности. Проводятся также инспекции, целью которых является обеспечение строгого соблюдения установленных условий. Кроме того, все подобные разрешения должны быть согласованы с другими обязанностями, касающимися надлежащего учета материалов.

При усовершенствовании национальной системы учета и контроля ядерных материалов в ЧССР был разработан, в первую очередь, перечень основных ядерных установок и мест, где проводятся работы с ядерными материалами [3]. На основе подробного анализа проектной документации этих установок и рабочих мест были разработаны исходные материалы для постепенного осуществления следующих мер:

а) достаточно подробная идентификация оборудования и ядерных материалов;
б) определение зон баланса материалов (ЗБМ), учетных мест и измерительных пунктов для отдельных установок;
в) определение способов и сроков представления докладов и отчетов;
г) определение требований, предъявляемых к содержанию докладов и к методам их оценки;
д) подбор соответствующих методов контроля.

Большинство зон баланса материалов национальной системы учета и контроля представляет собой зоны, где производится регистрация количеств материалов. В этих ЗБМ осуществляется учет поступлений и выхода ядерного материала по данным на входе или по данным экспедитора, а также по расчетным данным, постепенно исправляемым согласно величинам потерь ядерных материалов и продукции. В книжном и физическом

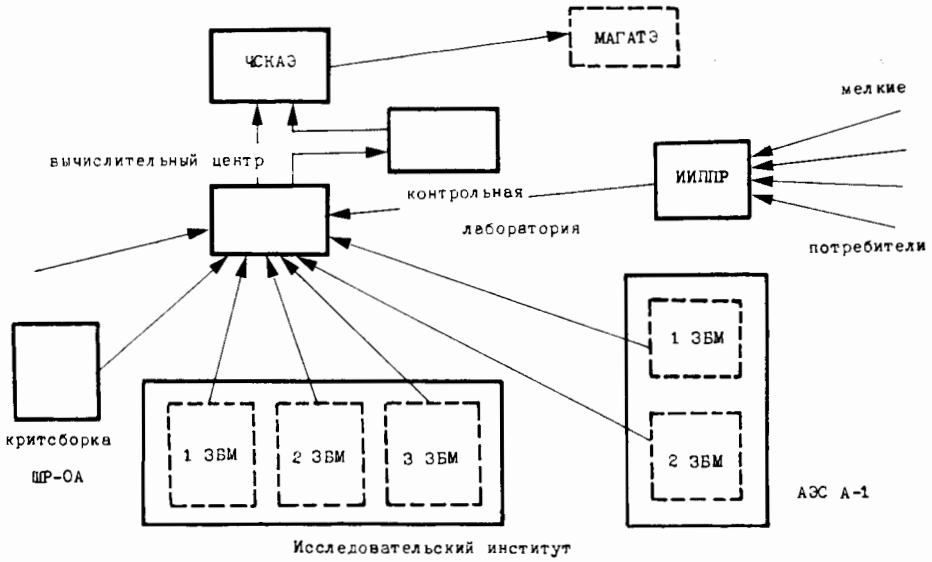

Рис.1. Часть общей схемы потока информации от отдельных ЗБМ.

учете данных в этих ЗБМ не должно быть какой-либо разницы, а неподдающийся учету материал должен быть, в нормальном случае, равен нулю.

Текущие материальные балансовые учетные документы (учетные документы об изменениях инвентарных количеств, итоговые учетные отчеты, отчеты о материальном балансе для отдельных зон баланса материалов) и предварительные отчеты о международных перевозках, направляемые в центральный орган учета, содержат сведения о ядерном и химическом составе, физической форме, общем весе элемента или весе делящегося изотопа для каждого вида ядерного материала, подлежащего учету и контролю. В отчетах приводятся исходная и приемная зона баланса материалов или (в случае экспорта из Чехословакии) первоначальные ЗБМ и получатель. В предварительных сообщениях указывается предварительная дата перевозки ядерного материала. Все упомянутые виды отчетов и предварительных уведомлений составляются по единой форме. Отдел ядерной безопасности и гарантий ЧСКАЭ установил для всех организаций и всех ЗБМ единую систему кодов по всем данным о ядерном материале и его передвижении. Несмотря на то, что в настоящее время для суммирования все еще применяются механические счетные машины, все сообщения, тем не менее, обрабатываются с учетом возможного применения электронно-вычислительной машины. Обработка данных на электронно-вычислительных машинах пока проводится лишь в опытном масштабе, но как только возрастет объем данных у системы контроля, обработка данных на электронно-вычислительных машинах будет внедрена постоянно.

Для наглядности на рис.1 приводится часть общей схемы потока информации от отдельных ЗБМ. ЧСКАЭ с помощью вычислительного центра и контрольной аналитической лаборатории суммирует полученные данные, ведет учет и проводит контроль силами собственных инспекторов. Чтобы не загружать информационные каналы второстепенными данными, потребители небольших количеств ядерных материалов были объединены

в одну самостоятельную ЗБМ, для которой соответствующий учет проводит Институт исследования, производства и применения радиоизотопов (ИИППР).

СИСТЕМА УЧЕТА И КОНТРОЛЯ ЯДЕРНЫХ МАТЕРИАЛОВ В НАУЧНО-ИССЛЕДОВАТЕЛЬСКОМ ИНСТИТУТЕ

В качестве первого примера комплексного внедрения системы учета и контроля можно привести Институт ядерных исследований Академии наук ЧССР, который является основным центром ядерных исследований в ЧССР. Данный центр приводится в качестве примера потому, что во многих странах имеются институты подобного же типа и направления.

После проведения анализа проектных данных [4] оборудования и рабочих мест в указанном выше институте были установлены три самостоятельные ЗБМ:

I ЗБМ – экспериментальный реактор (ВВР-С);
II ЗБМ – тяжеловодная критическая сборка ТР-О;
III ЗБМ – остальные радиохимические, физическо-металлургические и прочие рабочие места, а также те места, где находятся различные количества ядерного материала.

Остановимся более подробно на I-ой ЗБМ. Экспериментальный реактор ВВР-С является реактором бассейного типа, который служит источником излучения при производстве изотопов, при проведении экспериментальных работ в области атомной физики и инженерных исследований в области реакторной технологии. Реактор может работать на тепловой мощности до 4 МВт. Горючее содержит окись урана, обогащенную на 4-10% по изотопу ^{235}U. Замедлителем является дистиллированная легкая вода, служащая одновременно теплоносителем, отражателем и, частично, защитой.

С точки зрения гарантий данный тип реактора классифицируется как основное ядерное оборудование, а с точки зрения контроля и централизованного учета – обозначается как одна самостоятельная зона баланса материалов книжного типа.

I ЗБМ разделена на следующие учетные места:

1) склад топливных элементов;
2) реактор;
3) склад облученных топливных элементов;
4) горячие камеры.

В каждом учетном месте поштучно учитываются кассеты тепловыделяющих элементов (твэлов). Кроме того, рассчитываются ядерные потери ^{235}U и образование плутония в кассетах твэлов на основе мощности реактора. Физический учет ядерного материала в установке проводится раз в год. Каждое учетное место является одновременно контрольным местом.

В нормальных случаях проводится ревизия учетных данных, идентификация кассет твэлов, а в некоторых случаях – определяется содержание урана недеструктивными методами. Данные об общей мощности реактора проверяются на основе рабочих записей, проводимых на установке. Данные о ядерных потерях и содержании плутония в кассетах твэлов, которые должны быть отправлены из ЗБМ экспериментального реактора, сравниваются с данными об общем производстве энергии, ее распределением в активной зоне, а также с данными, занесенными в учетные карты

кассет твэлов. Ядерные потери урана и воспроизводство плутония учитываются только у кассет твэлов, которые должны быть отправлены из ЗБМ. Эти величины рассчитываются на основании данных, занесенных в учетную карту кассеты твэлов.

Система рабочих записей для реакторного оборудования следующая:

а) записи показывают действительную мощность реактора и состав активной зоны в любое время;
б) записи информируют об операциях, связанных с перегрузкой топлива в реакторе, и о каждом значительном ремонте;
в) при изменении состава активной зоны, но как минимум один раз в год, записывается общая выработанная энергия и ее распределение в отдельных кассетах твэлов в активной зоне.

Учетная система отчетов, почти совпадающая с установленными Агентством требованиями, базируется на следующем:

а) все типы учетных сообщений составляются по единой форме;
б) о ядерных потерях и производстве плутония отчитываются только по кассетам твэлов, которые должны быть отправлены из ЗБМ, в отчетах, касающихся данной партии;
в) окончательный отчет, переданный в виде суммарных отчетных сообщений, в материальном балансе разделен на отчетные места, указанные выше;
г) виды сообщений, сроки и частота их представления определены следующим образом:
   - извещения об изменениях учета подаются в течение 14 дней со дня возникновения учетного изменения;
   - суммарные отчетные сообщения представляются один раз в год и охватывают период с 1 января по 30 июня;
   - сообщение о материальном балансе, касающемся периода с 1 января по 31 декабря и включающем также отчетную сумму за период с 1 июля по 31 декабря, представляется один раз в год.

Рабочий отчет содержит короткую запись о целях работы реактора за отчетный период, описание существенных ремонтных работ и общее количество выработанной энергии в течение данного периода. Эти сообщения подаются два раза в год.

Подобный контроль осуществляется и во II-ой ЗБМ, т.е. в тяжеловодной критической сборке.

В III-тью ЗБМ включены все остальные рабочие места, лаборатории, отделения и небольшие склады, расположенные в институте. Это, например, рабочие помещения физической металлургии, лабораторных исследований переработки облученного топлива, переработки радиоактивных отходов и т.п. С целью уменьшения загрузки информационных каналов централизованного учета все рабочие места входят в состав одной ЗБМ с соответствующими отчетными местами. О переводах ядерного материала между этими местами ведутся соответствующие записи, которые, однако, не передаются в центр.

СИСТЕМА УЧЕТА И КОНТРОЛЯ ЯДЕРНЫХ МАТЕРИАЛОВ НА ПЕРВОЙ ЧЕХОСЛОВАЦКОЙ АТОМНОЙ ЭЛЕКТРОСТАНЦИИ

Первая чехословацкая атомная электростанция А-1 расположена в Ясловских Богуницах (на северо-востоке от Братиславы) и в настоящее

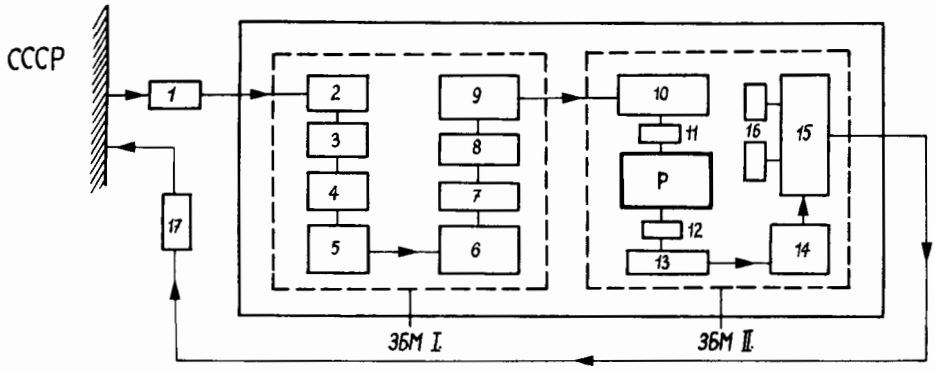

Рис.2. Сокращенная схема передвижения топлива по атомной электростанции А-1:

1 -перевозка;
2 – склад стержней твэлов;
3 – контроль оболочки твэлов;
4 – монтаж стержней твэлов;
5 – промежуточный склад;
6 – сборка кассет;
7 – контроль и обозначение;
8 – промежуточный склад;
9 – сборка комплектов;
10 – склад комплектов;
11,12 – загрузочная машина;
Р – реактор;
13 – промежуточное хранилище;
14 – камера резки;
15 – долговременное хранилище;
16 – горячие камеры;
17 – отправка вагон-контейнером.

время находится в стадии энергетического пуска. Поскольку эта атомная электростанция, работающая на тяжелой воде, имеет с точки зрения учета и контроля топлива некоторые нетипичные и в определенной степени уникальные свойства, будет целесообразно рассмотреть более подробно некоторые основные аспекты [5].

Основной частью атомной электростанции является реактор КС-150 со стальным корпусом давления. Топливом является природный металлический уран, замедлителем – тяжелая вода. Охладителем первого контура является CO_2, циркулирующий под давлением 60 атм. Предполагается, что тепловая мощность реактора составит 560 МВт, а электрическая мощность (брутто) – 143 МВт. Активная зона реактора состоит из двух типов кассет твэлов. В центральной зоне находятся 44 кассеты диаметром 112 мм, а в переферийной зоне – 104 кассеты диаметром 100 мм. Оболочкой кассеты является циркониевая труба. Общая загрузка реактора составляет примерно 25 т металлического природного урана. Высокая удельная мощность топлива – 20 МВт (тепл)/т урана – приводит к относительно быстрому выгоранию топлива и быстрому образованию плутония, что потребует частой замены кассет твэлов. Перегрузка кассет твэлов осуществляется непрерывно во время работы реактора. Предполагаемое время пребывания кассеты твэлов в реакторе – 99-235 дней, в зависимости от размещения в активной зоне. Предполагаемая глубина выгорания топлива составляет около 2800 МВт·сут/т.

Сложное передвижение топлива на атомной электростанции потребовало с точки зрения введения эффективной контрольной и учетной системы установления двух зон баланса материалов:

I ЗБМ включает пространство для монтажа и ремонта тепловыделяющих элементов с соответствующими складами;

II ЗБМ включает собственно реактор с соответствующими кратковременными хранилищами, горячими камерами, загрузочными машинами и долговременное хранилище облученных кассет твэлов.

Упрощенная общая схема потока ядерного топлива на атомной электростанции А-1 приведена на рис.2. Движение ядерного топлива начинается с поставки урановых стержней из СССР в I-ую ЗБМ атомной электростанции. На рис.2 схематично изображен поток ядерного топлива в I-ой ЗБМ при монтаже, подготовке, контроле, обозначении и хранении.

I-ая ЗБМ разделена на следующие отчетные места:

а) входной контроль (склад урановых металлических стержней);
б) промежуточный склад смонтированных стержней;
в) межоперационный склад кассет;
г) монтаж комплектов.

Главные части топливного элемента обозначены на видном месте номером. Каждый собранный топливный элемент далее обозначается на трех местах подвесной головки. О монтаже каждого топливного элемента ведется запись в учетной карте, содержащей основные данные производственного, учетного и контрольного характера. На рис.2 также изображено передвижение топливных элементов во II-ой ЗБМ, где расположен реактор, который обслуживается без остановки двумя загрузочными машинами, соответствующие кратковременные хранилища, горячие камеры и долговременное хранилище для хранения облученных кассет твэлов.

II-ая ЗБМ разделена на следующие отчетные места:

а) вход во II-ую ЗБМ (склад свежих кассет);
б) реактор;
в) кратковременное горячее промежуточное хранилище;
г) камера для резки;
д) долговременное хранилище облученного топлива с горячими камерами.

Для более ясного представления введенного учета и контроля топлива, проходящего через II-ую ЗБМ электростанции, рассмотрим некоторые аспекты, которые следует учитывать при определении контрольных мест и методов контроля.

Непрерывный обмен кассет твэлов при работе реактора, относительно небольшое количество технологических каналов, а также относительно небольшое количество кассет твэлов во всем топливном цикле электростанции позволяют осуществлять индивидуальное наблюдение за передвижением кассет твэлов. Все операции с твэлами – от склада свежих комплектов, далее через активную зону реактора, кратковременное горячее промежуточное хранилище, камеру резки, долговременное хранилище и вплоть до вагона-контейнера – осуществляются одной из двух загрузочных машин в одном закрытом помещении (реакторном зале). Из этого помещения транспортировка облученной кассеты каким-либо другим способом, чем через люк в вагон-контейнер, совершенно исключена.

На основании приведенных фактов были предложены ключевые контрольные места, через которые должен проходить весь ядерный материал (в форме металлических стержней, облученных и необлученных кассет) на своем пути передвижения по атомной электростанции, а именно:

В I ЗБМ

а) входной контроль на входе в склад урановых металлических стержней;
б) выходной контроль на выходе из монтажного отделения кассет при переходе в склад (выход из I ЗБМ и поступление во II ЗБМ).

Во II ЗБМ
а) вход в склад кассет твэлов;
б) реактор;
в) камера резки;
г) выходной люк из реакторного зала в вагон-контейнер.

Система отчетов и отчетных документов для атомной электростанции А-1 будет аналогична той, которая была приведена для экспериментального реактора ВВР-С, а в окончательной фазе – будет приспособлена к требованиям Агентства.

Важной составной частью национальной системы гарантий является Центральная контрольная лаборатория для аналитического контроля ядерного горючего. Лаборатория находится на территории Института ядерных исследований Академии наук ЧССР и непосредственно подчинена Чехословацкой комиссии по атомной энергии. В ее распоряжении имеются обычные радиохимические лаборатории, лаборатории исследований материалов средней активности (примерно до 1 или нескольких кюри) и лаборатории для работы с плутонием. Для радиометрического контроля, кроме обычного оборудования, применяются также тысячиканальные анализаторы с германиевыми и кремниевыми детекторами для гамма- или альфа-спектрометрии. Недавно была создана лаборатория масс-спектрометрии, оснащенная советским масс-спектрометром МИ 1311, предназначенным для изотопного анализа микро-количеств и микро-примесей газов и твердых веществ.

Лаборатории поручено контролировать ядерное топливо в течение всего топливного цикла, начиная с исходных соединений, далее промежуточные продукты, твэлы, продукты переработки облученного топлива, и кончая радиоактивными элементами. Кроме контроля, Лаборатория должна заниматься проверкой и разработкой новых методов [6].

ТАБЛИЦА I. ОПРЕДЕЛЕНИЕ УРАНА

Методика	Принцип метода	Применение	Коэффициент вариации, %
Титриметрические (объемные) методы	Восстановление в редукторе Джонса Титрование Ce (IV)	U (металл), окислы	0,2
	Экстракция купфероном, восстановление в редукторе Джонса, титрование Cr (IV)	окислы, сплавы флуориды	0,3 - 0,4
	Восстановление железом (II) Титрование Cr (IV) или Ce (IV)	U (металл), окислы фториды, U в растворах	0,2 - 0,3
	Восстановительное титрование двухвалентным железом Потенциометрическая индикация	U (металл), окислы сплавы, U в растворах	0,2 - 0,4
Потенциостатическая кулонометрия	Восстановление на Hg-катоде	U в растворах U (металл), окислы	0,2
Спектрофотометрические методы	Комплекс с роданидом Комплекс с Арсеназо III	U в растворах U в растворах	3 для следов 4-5

ТАБЛИЦА II. ОПРЕДЕЛЕНИЕ ПЛУТОНИЯ

Методика	Принцип метода	Применение	Коэффициент вариации, %
Титриметрические (объемные) методы	Отделение Pu на анионите, восстановление амальгамированными Zn или $TiCl_3$ Титрование Ce (IV), потенциометрическая индикация	Pu (металл), окислы	0,3
Потенциостатическая кулонометрия	Платиновый электрод, 1M HC или 1M $HClO_4$	Pu (металл), окислы в растворах	0,5
Спектрофотометрические методы	Комплекс с Арсеназо III	Pu (металл), окислы	5
	Комплекс с торием	Pu в растворах	5
Измерение α-активности или α-спектрометрия	После экстракции TTA	Pu в растворах жидкие отходы	3-5

ТАБЛИЦА III. ОПРЕДЕЛЕНИЕ ИЗОТОПНОГО СОСТАВА И ВЫГОРАНИЯ ЯДЕРНОГО ГОРЮЧЕГО

Методика	Принцип метода	Применение	Коэффициент вариации, %
Изотопический состав	γ-спектрометрия	$^{235}U/^{238}U$ в металлическом U	1,5-3
	Измерение запаздывающих нейтронов	в окислах	2-4
	Активационный анализ	в сплавах	3-5
Определение выгорания	Радиохимические методы (определение радиоактивных или стабильных продуктов деления)	природный и обогащенный U	

Перечень методов, выбранных к настоящему времени и прошедших проверку, приводится в табл.I-III. Из табл.I-III видно, что точность определения для данных целей в большинстве случаев является удовлетворительной.

Эффективность гарантий зависит от разработки надежных контрольных методов – достаточно точных, независимых от условий эксплуатации установок и не ограничивающих собственную работу установок. Поэтому составной частью развития чехословацкой системы учета и контроля являются также исследовательские и рациоанализаторские работы, направленные на разработку методов независимой проверки полученных данных. Некоторые из этих работ проводятся по контрактам с МАГАТЭ (например, анализ возможного определения количества плутония, образующегося при работе тяжеловодной атомной электростанции, на основе количества выработанной электроэнергии [7] и т.д.).

ЗАКЛЮЧЕНИЕ

Приведенная общегосударственная система учета и контроля ядерного материала будет и далее уточняться на основании требований Агентства, предъявленных в ходе переговоров о заключении соглашений между Чехословакией и Агентством, в особенности – при разработке дополнительных положений о контроле отдельных установок, которые войдут в сферу действия гарантий.

Целью интенсивных работ по усовершенствованию национальной системы учета и контроля ядерного материала является разработка технических и юридических аспектов и условий, отвечающих полностью требованиям, вытекающим из Договора о нераспространении ядерного оружия. Удовлетворение этих требований позволило бы Агентству исполнять свои обязанности при минимальных усилиях в деле проведения инспекций, а также при минимальных финансовых расходах.

Мы надеемся, что опыт ЧССР по созданию национальной системы учета и контроля ядерного материала будет полезен и для других государств, которые находятся или будут находиться в подобном положении.

ЛИТЕРАТУРА

[1] НЕУМАНН, Я, БАРАБАС, К., Опыт, накопленный в ЧССР, в области развития ядерной энергетики. P/541, Proc. 4th Intern. Conf., Geneva, 1971, IAEA, Vienna 1 (1972).
[2] Закон № 133 от 1970 года "О деятельности Федеральных министерств".
[3] ЖОХ, О., Перечень основных ядерных установок и мест в ЧССР, содержащих ядерные материалы, которые будут подлежать гарантиям, и проект определения зон баланса материалов. ЧСКАЭ, Прага, 1970.
[4] Проект Института ядерных исследований, Хемопроект, Прага, 1956.
[5] Проект атомной электростанции А-1, Энергопроект, Прага, 1966.
[6] KŘIVÁNEK, M., KRTIL, J, The determination of burn-up of Nuclear Fuels by Radiochemicals Methods, Referát na panelu IAEA "Analytical Chemistry of Nuclear Fuels", Vienna, 1970.
[7] DRAHNÝ, M., KUČERA, J., SVĚTLÍK, J. et al, Analysis of Possibility of Estimating the Quantity of Plutonium Produced in a Nuclear Power Plant on the Basis of Measured Quantity of Generated Electricity, Final Report under IAEA Research Contract No. 735/RB, 1969 (Summary published by IAEA, Research Contracts, Eleventh Annual Report, IAEA, Vienna, 1971).

LE CONTROLE DE SECURITE DE LA COMMISSION DES COMMUNAUTES EUROPEENNES

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Abstract–Résumé–Аннотация–Resumen

THE SAFEGUARDS APPLIED BY THE COMMISSION OF THE EUROPEAN COMMUNITIES.

The Treaty establishing the European Atomic Energy Community set up the first complete safeguards system for application by an independent authority within an organization comprising several States. The first part of the paper contains an analysis of the main Treaty provisions governing the activity of the Commission's Directorate for Safeguards and a detailed description of its structure. This is followed by an outline of the functions of the inspectorate, with emphasis on the various procedures applied according to the particular case in point (routine inspections, special inspections, spot checks, etc.), and a description of the operation of the centralized accounting system, under which all the data communicated by the Parties subject to such controls in the six Member States are brought together at the Headquarters of the Commission. At the end of the first part assessments are made of the overall effort put forth by the Commission's Directorate for Safeguards in order to ensure that nuclear material is used for peaceful purposes only.

The second part deals with research aimed at optimizing the safeguards and controls system from the standpoint of efficiency, economy and minimum intrusion. This work was initiated at the establishments of the Joint Research Centre in 1969, and has been concerned primarily with: (a) systems studies: mathematical models have been created for description of fabrication and reprocessing plants, and practice controls have been carried out on various facilities; (b) correlation studies: these are concerned in particular with the correlations between fission gases and burn-ups; (c) the development of direct and indirect methods: neutron physics and gamma-ray spectrometry techniques have been devised and appliances developed for the non-destructive measurement of fissile materials, and a method of automating isotopic dilution and mass spectrometric analysis is under study; and (d) the elaboration of identification techniques: the bulk of this work is devoted to the use of a technique based on ultrasonic detection of random distributions of inclusions.

LE CONTROLE DE SECURITE DE LA COMMISSION DES COMMUNAUTES EUROPEENNES.

Le Traité instituant la Communauté européenne de l'énergie atomique crée le premier système complet de contrôle par une autorité indépendante, dans le cadre d'une organisation comptant plusieurs Etats. Dans la première partie du mémoire, après une analyse des principales dispositions du Traité qui régissent l'activité du Contrôle de sécurité, les auteurs font un exposé détaillé de la structure du Service mis en place par la Commission; ils décrivent ensuite l'activité d'inspection, en mettant en lumière notamment les différentes modalités de celle-ci, appliquées selon les cas d'espèce (inspections périodiques, renforcées, inopinées, etc.). Le fonctionnement du système de comptabilité centralisée qui réunit au siège de la Commission l'ensemble des données communiquées par tous les assujettis des six pays membres est également décrit. Les auteurs concluent la première partie en faisant l'évaluation de l'effort global fourni par les services de la Commission pour assurer un contrôle efficace de l'utilisation des matières nucléaires à des fins exclusivement pacifiques.

La deuxième partie est consacrée aux études sur les garanties dans le Centre commun de recherche. Dans le souci d'optimiser le système de contrôle du point de vue de l'efficacité et de l'économie et d'assurer un minimum d'intrusions, des recherches ont été entreprises dès 1969 dans les établissements du Centre commun

de recherche, et notamment: a) études de systèmes: des modèles mathématiques ont été établis pour la description des usines de fabrication et de retraitement, des exercices de contrôle ont été effectués sur des installations; b) études de corrélation: des études sont exécutées, en particulier, sur les corrélations entre gaz de fission et taux de combustion; c) mise au point de méthodes directes et indirectes: pour la mesure non destructive des matières fissiles, des techniques neutroniques et de spectrométrie γ ont été étudiées et des appareillages ont été mis au point; une méthode d' automatisation de l' analyse par dilution isotopique et spectrométrie de masse est en cours d' étude; d) mise au point de techniques d' identification: l' effort principal porte sur l' utilisation d' une technique basée sur l' identification par ultra-sons de distributions aléatoires d' inclusions.

ГАРАНТИИ, ПРИМЕНЯЕМЫЕ ЕВРОПЕЙСКИМ СООБЩЕСТВОМ ПО АТОМНОЙ ЭНЕРГИИ. К ВОПРОСУ ИССЛЕДОВАНИЙ В ОБЛАСТИ ГАРАНТИЙ, ПРОВОДИМЫХ В ОБЪЕДИНЕННОМ ЦЕНТРЕ ИССЛЕДОВАНИЙ.

Договор о создании Европейского сообщества по атомной энергии установил первую цельную систему гарантий, применяемую независимым органом в рамках организации, охватывающей несколько государств. В первой части доклада после анализа основных положений Договора, определяющих деятельность дирекции Комиссии по гарантиям, авторы дают подробное описание ее структуры. Затем указываются функции инспекторов с уделением особого внимания различным процедурам, применяемым в зависимости от конкретного случая (обычные инспекции, специальные инспекции, проверки на местах), дается также описание работы централизованной системы учета, предусматривающей сбор в центральных учреждениях Комиссии всех данных, сообщаемых лицами, подлежащими такому контролю в шести государствах-членах. Наконец, дается оценка всей работы, выполняемой дирекцией Комиссии по гарантиям в целях обеспечения эффективного контроля за использованием ядерного материала только в мирных целях.

Во второй части говорится о том, что в 1969 году в Объединенном центре исследований были начаты работы, направленные на оптимизацию системы гарантий и контроля с точки зрения эффективности, экономической выгоды и минимального вмешательства, включая, в частности: а) изучение систем (были разработаны математические модели для описания установок по изготовлению и переработке топлива; на различных установках осуществлялся практический контроль); b) изучение корреляций (эти исследования касались, в частности, корреляций между образованием газообразных продуктов деления и глубиной выгорания); с) прямые и косвенные методы (разрабатывались нейтроннофизические и спектрометрические методы и приборы для измерений расщепляющихся материалов без разрушения образца; разрабатывается метод автоматического изотопного разбавления и масс-спектрометрии); d) идентификация методов (основная часть этой работы посвящена использованию метода, основанного на ультразвуковом детектировании беспорядочного распределения включений).

EL SISTEMA DE SALVAGUARDIAS DE LA COMISION DE LAS COMUNIDADES EUROPEAS.

El Tratado que funda la Comunidad Europea de Energía Atómica estableció el primer sistema completo de salvaguardias para ser aplicadas por una autoridad independiente, dentro del marco de una organización integrada por varios Estados. En la primera parte de esta memoria y después de un análisis de las principales disposiciones del Tratado que rigen las actividades de salvaguardia, los autores exponen en detalle la estructura de los servicios correspondientes; a continuación describen las actividades de inspección, poniendo especialmente de manifiesto los diversos procedimientos aplicados en los distintos casos (inspecciones ordinarias, inspecciones especiales, sin preaviso, etc.). Se describe también el funcionamiento del sistema contable centralizado, en virtud del cual se reúnen en la Sede de la Comisión los datos comunicados por todas las entidades sometidas a estos controles en los seis países miembros. Finalmente, los autores concluyen la primera parte con una evaluación de la labor realizada por los servicios de la Comisión para asegurar un control eficaz de la utilización de materiales nucleares con fines pacíficos exclusivamente.

La segunda parte está dedicada a los estudios sobre salvaguardias realizados en el Centro Común de Investigaciones. Con el fin de conseguir los mejores resultados desde el punto de vista de la eficacia, de la economía y de la mínima intrusión se han iniciado en 1969 estudios en los establecimientos del Centro Común de Investigaciones sobre las siguientes materias principales: a) estudios de sistemas: se han establecido modelos matemáticos para describir las plantas de fabricación y de reelaboración; se han realizado ejercicios de control en varias instalaciones; b) estudios de correlación: estos estudios se refieren en particular a las correlaciones entre gases de fisión y grados de quemado; c) puesta a punto de métodos directos e indirectos: se han estudiado técnicas neutrónicas y de espectrometría y para la medición no destructiva de materiales fisionables, y se han desarrollado los equipos correspondientes; se está investigando un método de automatización del análisis por dilución isotópica y espectrometría de masas; d) puesta a punto de técnicas de identificación: la mayor parte del trabajo se ha dedicado al empleo de una técnica basada en la detección ultrasónica de distribuciones aleatorias de inclusiones.

PREMIERE PARTIE

1. STRUCTURE ET ACTIVITES DU SERVICE CONTROLE DE SECURITE DE LA COMMISSION DES COMMUNAUTES EUROPEENNES

Depuis la fin de la guerre, un effort a été entrepris pour placer sous contrôle international les armes atomiques et les activités qui permettent de les produire. Cet effort, qui a abouti récemment à la conclusion d'un accord international de très large portée, le Traité de non-prolifération (TNP), a été précédé à l'échelle européenne par la mise sur pied d'un système de contrôle multinational entre les six pays membres de la Communauté européenne, système qui jouit d'une expérience opérationnelle qui dépasse dix ans.

Le Contrôle de sécurité, confié par les Traités de Rome à la Commission des Communautés européennes, est basé sur l'acceptation de règles et d'une autorité commune et a des caractéristiques qu'il convient de souligner.

a) Ce système de contrôle a un caractère obligatoire et il est directement appliquable sur le territoire de chacun des Etats membres.

b) L'organisation de contrôle – en l'occurrence la Commission – est mise directement en relation avec les détenteurs de matières assujetties au contrôle, ce qui permet à la Commission, et plus particulièrement aux inspecteurs mandatés à cet effet, de s'adresser directement aux entreprises.

c) Enfin, ce système s'applique sans aucune limitation de durée à toutes les activités en rapport avec les utilisations pacifiques de l'énergie nucléaire dans les pays membres.

Pour souligner l'aspect particulier des caractéristiques que l'on vient de mentionner, il convient de rappeler – entre autres – que, dans le cadre du système de contrôle de l'AIEA, qui a été en vigueur jusqu'à présent, l'Agence ne peut pas s'adresser directement aux détenteurs de matières assujetties au contrôle, mais elle doit passer par l'intermédiaire du pays membre intéressé; en outre, son système de contrôle ne s'applique qu'aux cas d'utilisation pacifique de l'énergie nucléaire que les Etats membres soumettent volontairement au contrôle, et ceci uniquement pendant la période prévue par l'accord conclu en matière de contrôle. Par l'entrée en vigueur du TNP et la conclusion de nouveaux accords de contrôle avec les pays membres, le système de l'Agence devient obligatoire et total – pour les Etats non dotés d'armes nucléaires qui sont parties au Traité – en ce qui concerne l'engagement d'utilisation pacifique des matières nucléaires. Le système de l'Agence vient donc d'acquérir ainsi l'une des caractéristiques essentielles du système du contrôle communautaire européen.

Le Contrôle de sécurité de la Commission des Communautés européennes porte sur l'uranium, le thorium et les matières fissiles spéciales qui en dérivent; il porte, en outre, en raison d'engagements souscrits avec des pays tiers, sur d'autres matières telles que l'eau lourde et le tritium. Le contrôle ne s'applique pas à l'essai, à la mise au point, à la fabrication, au stockage, ni des armes atomiques ni des moyens de les porter jusqu'à leur objectif. Il résulte de l'exécution des dispositions du Chapitre VII du Traité et des Règlements qui ont été pris pour leur application.

La Commission a pris jusqu'à présent deux Règlements: le Règlement n°7 publié au Journal officiel des Communautés européennes du 12 mars 1959 et le Règlement n° 8, publié au Journal officiel du 29 mai 1959.

Comme on vient de le souligner, les textes ci-dessus mentionnés sont directement obligatoires pour les entreprises et les institutions publiques et privées qui y sont assujetties, et cela sans intervention des gouvernements ou des administrations des pays membres.

Les objectifs du Contrôle de sécurité de la Commission sont définis à l'Article 77 du Traité, aux termes duquel la Commission doit s'assurer, sur les territoires des Etats membres, que les minerais, matières brutes et matières fissiles spéciales ne sont pas détournés des usages auxquels leurs utilisateurs ont déclaré les destiner; que sont respectées les dispositions relatives à l'approvisionnement; qu'est respecté tout engagement particulier relatif au contrôle souscrit par la Communauté dans un accord conclu avec un Etat tiers ou une organisation internationale.

Pour permettre à la Commission de répondre à l'ensemble de ses obligations, le Traité impose à quiconque établit ou exploite des installations produisant, séparant ou utilisant des matières brutes ou des matières fissiles spéciales, ou bien traitant des combustibles nucléaires irradiés, de déclarer à la Commission les caractéristiques techniques fondamentales de ces installations (Article 78). Il impose, en outre, la tenue et la présentation de relevés d'opérations permettant la comptabilité des minerais, matières brutes et matières fissiles spéciales utilisés ou produits (Article 79).

Ces dispositions sont renforcées par le droit que le Traité donne à la Commission d'envoyer sur les territoires des Etats membres des inspecteurs qui ont à tout moment accès à tous lieux, à tous éléments d'information et auprès de toutes personnes qui s'occupent des matières, équipements ou installations soumis au contrôle.

On peut constater que les dispositions que l'on vient de décrire visent à doter la Commission des moyens appropriés pour accomplir la mission de contrôle qui lui a été confiée par le Traité, mais qu'elles ne limitent pas, et il convient de le souligner, la liberté dont les entreprises ou les institutions disposent pour déterminer, en fonction de considérations techniques ou économiques, leurs investissements ou l'exploitation de leurs installations.

En effet, les éléments essentiels du système de contrôle sont, d'une part, les déclarations faites par les entreprises à la Commission et, d'autre part, les vérifications sur place réalisées par les inspecteurs.

L'application du Règlement n° 7 pris en exécution de l'Article 78 fournit à la Commission, et lui permet de tenir à jour, les plans et la connaissance des installations en ce qui concerne leur capacité, la nature des matières utilisées et produites, les procédés techniques employés et les méthodes appliquées pour mesurer et vérifier les quantités et la qualité des matières détenues dans les installations assujetties au contrôle.

Par les informations qu'elle reçoit en application du Règlement n° 8, la Commission a connaissance des stocks détenus par les assujettis, de la localisation des matières, des mouvements d'une installation à l'autre, des importations et des exportations avec les pays tiers.

La comptabilité-matières organisée par la Commission comporte des comptes par installation et par matière, ventilés suivant l'origine et le statut de celles-ci.

La Commission des Communautés veille au respect des dispositions du Traité et, le cas échéant, peut constater les infractions et prononcer des sanctions: avertissement, retrait d'avantages particuliers tels qu'assistance financière ou aide technique, mise de l'entreprise, pour une durée maximale de quatre mois, sous administration contrôlée, retrait total ou partiel des matières brutes ou matières fissiles spéciales.

Les Etats membres sont tenus d'assurer l'exécution de ces sanctions.

Par ailleurs, le Contrôle de sécurité étant partie intégrante du système institutionnel de la Communauté européenne, c'est la Commission des Communautés, dans ce domaine comme dans les autres, qui est responsable de son action devant l'Assemblée parlementaire européenne et les intéressés peuvent, en cas de contestation, former contre ses décisions un recours devant la Cour de Justice des Communautés. Ainsi doivent se trouver combinés, dans la réalité, le pouvoir nécessaire de décision de la Commission et la garantie des droits individuels contre l'arbitraire.

1.1. Structure du Contrôle de sécurité

Ce Service comprend actuellement une soixantaine d'agents regroupés en deux divisions: Division 1, «Contrôle et comptabilité sur pièces», et Division 2, «Inspections et engagements extérieurs». Le corps d'inspecteurs de la Commission compte 32 inspecteurs et inspecteurs-adjoints dûment habilités au secret et nommés dans leurs fonctions par la Commission, après consultation des Etats membres. Les inspecteurs ont une formation appropriée aux tâches qu'ils doivent exécuter: un certain nombre d'entre eux accomplissent le contrôle «comptable», d'autres le contrôle «technique». Les premiers ont une formation économique et juridique; les seconds proviennent, pour la plupart, des Etablissements du Centre commun de recherche nucléaire de la Communauté et leur formation relève de domaines tels que le génie nucléaire, la physique nucléaire, l'électronique, la chimie nucléaire, la métallurgie, la radioprotection, etc.

1.1.1. La comptabilité-matières

On a déjà mentionné précédemment que c'est grâce aux données fournies par les assujettis d'après les prescriptions du Règlement n° 8 que la Commission entre en possession des informations qui lui sont nécessaires pour suivre l'évolution des matières nucléaires dans les six pays.

Sur la base des déclarations des assujettis, la Commission des Communautés européennes a mis sur pied et tient à jour une comptabilité de l'ensemble des minerais, matières brutes et matières fissiles spéciales existant dans la Communauté, ainsi que des stocks, mouvements et pertes. Cette comptabilité est assurée par des machines IBM.

L'examen des déclarations fournies par les assujettis fait apparaître parfois la nécessité d'explications ou de corrections. Dans ce cas, la Commission se met directement en rapport avec les installations. Cette procédure a toujours eu l'avantage de créer des contacts personnels entre le Service du Contrôle et les entreprises intéressées.

Les déclarations périodiques fournies par les installations sont appréciées sous deux angles différents:

a) Du point de vue comptable, on vérifie l'exactitude et la concordance des déclarations, en particulier:

- si les augmentations et diminutions enregistrées par rapport au stock initial d'une installation sont égales à la variation de ce stock;
- si les fournitures entre installations de la Communauté s'équilibrent et se compensent.

b) Du point de vue technique, on étudie et examine, entre autres, les points suivants:

- la composition de l'inventaire mensuel,
- la nature des mouvements de matières,
- les pertes de matières déclarées.

A titre d'information, il convient d'indiquer qu'au 31 décembre 1970 250 installations avaient déclaré leurs caractéristiques techniques fondamentales (Règlement n° 7). En outre, à la même date, 22 mines et 67 installations n'appartenant pas au cycle du combustible nucléaire étaient enregistrées auprès du Service Contrôle de sécurité.

1.1.2. L'inspection

La Commission des Communautés fait procéder à des inspections sur place qui, selon le cas, ont un caractère «intermittent», «continu» ou – d'après une instruction prise par la Commission récemment – «renforcé», ce dernier type d'inspections s'appliquant à certaines installations de production d'éléments de combustible.

Le programme d'inspections, fixé sur la base, notamment, de la pondération des installations, regroupe en une «mission» plusieurs installations à inspecter, localisées sur un même site ou dans le voisinage géographique: cela pour des raisons d'économie, afin de limiter au maximum les déplacements et d'utiliser au mieux le temps des inspecteurs. La durée d'une mission d'inspection est normalement comprise dans les jours ouvrables d'une semaine.

Chaque équipe d'inspection est composée, généralement et suivant les dimensions et la complexité de l'installation qui doit être visitée, de deux ou trois inspecteurs. Ceux-ci jouissent, ainsi qu'on l'a indiqué précédemment, des prérogatives les plus larges en matière d'investigation. A la demande de l'Etat intéressé, les inspecteurs peuvent être accompagnés de représentants de cet Etat, agissant en qualité d'observateurs.

Avant de quitter le siège de la Commission pour partir en mission, les inspecteurs reçoivent un programme détaillé, à la préparation duquel ils ont participé. Ce programme délimite le cadre des vérifications à effectuer et précise:

- l'installation qui doit être inspectée,
- la date de l'inspection,
- les matières et/ou matériaux faisant l'objet de l'inspection,
- les contrôles comptables à effectuer,
- les contrôles physiques à réaliser.

a) En ce qui concerne les vérifications comptables, les inspecteurs de la Commission se font présenter la comptabilité-matières et les

documents émanant des fournisseurs et des transporteurs. Ils procèdent à un inventaire comptable des stocks détenus dans l'installation à la date de l'inspection. Ils comparent les comptes de l'entreprise et les documents émanant de tiers aux déclarations faites à la Commission en vertu du Règlement n° 8.

b) Les vérifications techniques comportent notamment l'établissement de l'inventaire physique au jour de l'inspection; les opérations à effectuer sont détaillées dans les manuels particuliers à chaque type d'installation. Ces vérifications ont pour objet, notamment:

- de vérifier, s'il y a lieu, que les caractéristiques fondamentales de l'installation sont effectivement celles qui ont été déclarées à la Commission par application du Règlement n° 7;
- d'établir, par nature de matière (ou par type technologique de matériau, par exemple noyaux U/Al, aiguilles d'UO_2 enrichi, éléments de combustible, plaquettes UO_2/PuO_2) et par localisation dans l'installation, les stocks partiels. Cette détermination est faite soit par mesure physique (pesée, mesure de volume et prise d'échantillons, spectrométrie gamma) soit à l'aide des documents «opérationnels» internes (bons de réception, avec documents de livraison correspondants, fiches de pesées, fiches de fabrication, fiches de contrôle de fabrication, bordereaux d'expédition avec bons de prise en charge, etc.);
- d'établir ensuite, au moyen des résultats obtenus, un inventaire global pour la (ou les) matière(s) soumise(s) à l'inspection;
- de contrôler que les matériaux et produits finis correspondent, quant à leurs caractéristiques, aux usages qui ont été déclarés, ainsi que de vérifier, en cas de besoin, le respect des engagements extérieurs.

c) La suite à l'inspection. Dès leur retour au siège de la Commission, les inspecteurs établissent un rapport d'inspection exposant, point par point, par référence à l'ordre de mission, les résultats acquis au cours des vérifications, contrôles et relevés effectués auprès de l'installation. Selon les cas, ce rapport est soit transmis à l'installation pour observations, soit diffusé à l'intérieur du Service et conservé en archive. Lorsque l'installation ne reçoit pas copie du rapport, elle est en tout état de cause informée des résultats de l'inspection par lettre. Il est, en effet, toujours donné une suite contradictoire aux inspections.

1.2. L'étude et le développement des techniques de contrôle

La complexité croissante des opérations d'inspection a amené le Contrôle de sécurité à mettre à l'étude, en collaboration avec le Centre commun de recherche de la Commission, et à expérimenter sur place différentes techniques de contrôle. Le programme de recherches exécuté par le CCR et auquel le Service Contrôle de sécurité participe activement avec un certain nombre d'agents de sa Division 1 est illustré dans la deuxième partie de ce document. On se bornera donc à souligner que les techniques décrites dans les pages qui suivent et les appareils qui ont été mis au point en collaboration avec le CCR sont déjà en partie utilisés par les inspecteurs de la Commission dans l'exercice normal de l'inspection.

DEUXIEME PARTIE

2. ETUDES SUR LES GARANTIES DANS LE CENTRE COMMUN DE RECHERCHE (CCR)

L'activité de recherche et de développement sur les garanties a été poursuivie depuis 1969 dans les établissements d'Ispra, Karlsruhe et Petten du CCR dans les domaines suivants:
- analyse des systèmes,
- étude des corrélations isotopiques,
- méthodes indirectes,
- méthodes directes,
- techniques d'identification.

Le choix des sujets d'étude a été conditionné d'une part par les besoins du Service Contrôle de sécurité et d'autre part par une coordination des activités dans le cadre de l'association pour la recherche et le développement concernant les garanties, association à laquelle adhèrent, outre l'Euratom, le CEN (Bruxelles), le CNEN (Rome), la GfK (Karlsruhe), le RCN (La Haye).

2.1. Analyse des systèmes

Les études d'analyse des systèmes ont été développées principalement dans deux voies:
- l'examen théorique et critique de méthodes de contrôle au moyen de modèles mathématiques,
- l'exécution d'exercices de contrôle.

2.1.1. Modèles mathématiques et simulation numérique

Une usine de fabrication d'éléments de combustible à plutonium a été étudiée par le moyen d'un modèle stochastique qui a permis d'atteindre des résultats intéressants [1, 2]. Le modèle décrit la partie centrale de l'usine, avec certaines hypothèses assez réalistes quant aux règles de fonctionnement qui lui sont appliquées. On suppose qu'un magasin intermédiaire se trouve à chaque étape du processus de fabrication, et qu'après chacune de ces étapes un contrôle de qualité est effectué, avec une probabilité de rejet des pièces constante dans le temps, mais différente, en principe, d'une étape à l'autre.

On peut alors démontrer qu'un fonctionnement de l'usine en régime stationnaire est possible seulement si les probabilités de rejet satisfont à une relation simple. En outre, on trouve la loi stochastique qui détermine en fonction du temps la quantité contenue dans le magasin du matériel à recycler. On peut envisager un système de contrôle se basant sur la mesure des flux d'entrée et de sortie et, de temps en temps, la mesure de l'inventaire du matériel recyclable. Les résultats de l'étude permettent de déterminer les moments qui maximisent la probabilité que l'inspecteur découvre un détournement éventuel, par le moyen de cette dernière mesure.

En ce qui concerne les usines de retraitement, le problème s'est posé de déterminer dans quelles conditions une certaine méthode de contrôle peut être appliquée. Cette méthode consiste à utiliser les isotopes mineurs des matières fissiles comme traceurs pour la mesure

de l'inventaire en cours de traitement. Pour en étudier les possibilités d'emploi un modèle numérique a été mis au point et exploité sur l'ordinateur IBM 360/65. Ce modèle identifie les deux lignes de purification de l'uranium et du plutonium (pour éléments de combustible faiblement enrichi) de l'installation de retraitement Eurochemic de Mol (Belgique). La simulation numérique a permis d'obtenir une série de résultats importants [3] qui concernent, entre autres, les points suivants:

- étude des rapports entre les quantités disponibles de matériel à retraiter et le pourcentage de l'inventaire qui peut être mesuré;
- biais dans la mesure de l'inventaire résultant de la variabilité des spécifications qui caractérisent le matériel à retraiter;
- estimation de l'erreur statistique de l'inventaire.

Les études de simulation effectuées ont aussi permis d'établir pour l'expérience de contrôle JEX 70 (MOL III) les règles d'opération de l'installation qui minimisent les erreurs dans la mesure de l'inventaire en cours de traitement.

2.1.2. Exercices de contrôle

Les méthodes et techniques de contrôle, suggérées par des études d'analyse de systèmes, doivent être expérimentées dans la pratique avant de pouvoir être appliquées d'une façon routinière. A cette fin l'Euratom a projeté et effectué l'exercice de contrôle KRITO, a participé très activement à l'exercice JEX 70 (MOL III) et a entamé l'exercice ECE.

KRITO [4]. Cet exercice de contrôle a été effectué dans le cadre d'une collaboration entre la Direction Contrôle de sécurité et le CCR. Il a eu lieu à l'usine NUKEM, lors d'une fabrication spéciale d'éléments combustibles mettant en œuvre approximativement 190 kg d'uranium hautement enrichi. Le but de cet exercice était essentiellement la détermination du bilan indépendant des matières fissiles pour la campagne de fabrication. Pendant cet exercice, des techniques non destructives de mesure ont été largement utilisées pour le contrôle des produits finis.

JEX 70 (Joint Experiment 1970) [3]. Cet exercice, promu par la GfK en coopération avec l'Euratom, a été effectué avec la collaboration de plusieurs équipes de différents pays et de l'AIEA. Le but en était principalement de vérifier la possibilité d'application à l'usine de retraitement d'Eurochemic (Mol, Belgique) d'une méthode pour la détermination de l'inventaire en cours [5]. L'Euratom a développé un effort considérable, qui a été dirigé principalement sur les points suivants:

- études d'analyse des systèmes, par la méthode de simulation numérique (voir paragraphe 2.1.1);
- traitement des données relatives aux mesures d'inventaire; évaluation des résultats obtenus et conséquences sur les procédures d'application des garanties;
- exécution d'analyses chimiques pour la détermination de la concentration et de la composition isotopique des matériaux fissiles;
- évaluation des résultats de l'expérience de comparaison entre huit différents laboratoires d'analyses (dont trois appartenant au CCR de l'Euratom).

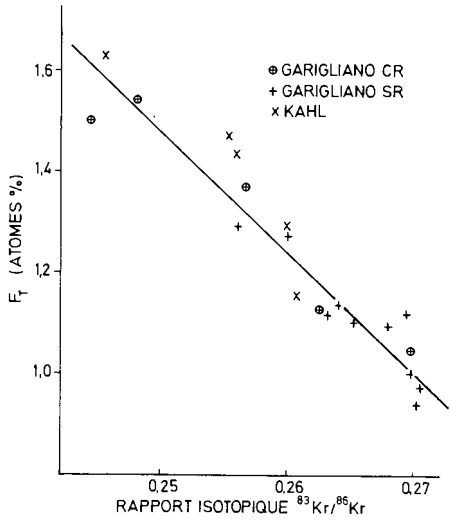

FIG. 1. Rapport isotopique $^{83}Kr/^{86}Kr$ en fonction du taux de combustion (F_T).

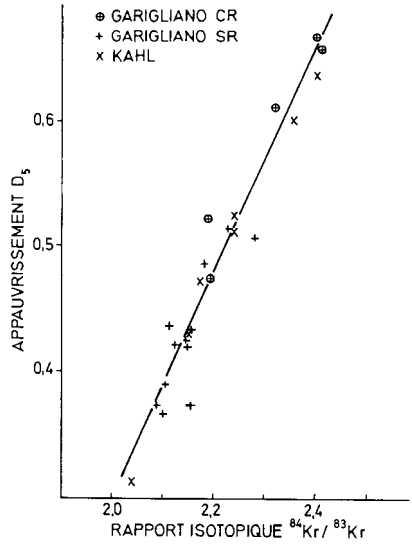

FIG. 2. Rapport isotopique $^{84}Kr/^{83}Kr$ en fonction de l'appauvrissement (D_5).

ECE (Eurex Control Experiment). Cette expérience, qui vient de démarrer (printemps 1971) sur l'usine de retraitement Eurex (Saluggia, Italie), a été projetée principalement pour mettre au point dans la pratique une méthode originale pour la vérification statistique du bilan des matières déclaré par l'opérateur. La méthode prend en considération les données provenant d'autres installations du cycle du combustible nucléaire. L'expérience ECE sera aussi utilisée pour vérifier encore une fois la possibilité d'application de la méthode décrite sous [5].

2.2. Etude des corrélations isotopiques

Plusieurs échantillons de combustible irradiés dans différents réacteurs (BWR, PWR, HWR, réacteurs rapides) ont été analysés [6]. Le but consistait à évaluer la quantité de matières fissiles à l'entrée de l'usine de retraitement et à vérifier la précision des méthodes théoriques [7]. Des corrélations entre les rapports des isotopes lourds et le taux de combustion ou la consommation des matériaux fissiles ont été observées, mais ne seront pas traitées ici. Par contre cette présentation est limitée aux corrélations entre les isotopes des gaz de fission. Pour deux BWR (Kahl et Garigliano) avec trois enrichissements initiaux différents, on a tracé les graphiques des rapports isotopiques de $^{83}Kr/^{86}Kr$ et $^{84}Kr/^{83}Kr$, mesurés expérimentalement, en fonction du taux de combustion F_T et de l'appauvrissement $D_5 = (^{235}U^0 - {}^{235}U)/^{235}U^0$ respectivement (fig.1 et fig.2). Dans les figures 1 et 2, Garigliano CR et Garigliano SR se rapportent respectivement à des barres d'angle avec un enrichissement initial de 1,6% et à des barres standards ayant un enrichissement initial de 2,1%.

L'analyse par les moindres carrés a montré qu'en première approximation les deux équations suivantes étaient valables pour une corrélation linéaire:

$$-23,2\ {}^{83}Kr/{}^{86}Kr + 7,3 = F_T \pm 3,3\% \qquad (1\,\sigma) \qquad (1)$$

$$0,9\ {}^{84}Kr/{}^{83}Kr - 1,5 = D_5 \pm 1,1\% \qquad (1\,\sigma) \qquad (2)$$

Comme l'inspecteur doit connaître l'enrichissement initial et la quantité de combustible retraité, l'uranium-235 restant peut être calculé à partir de l'équation (2). La précision de ces méthodes sera améliorée en utilisant des échantillons provenant de la dissolution de grappes de combustible complètes, au lieu d'échantillons sous forme de pastilles, qui furent utilisés au cours de cette étude. La technique des corrélations des gaz de fission présente deux avantages:

a) Elles ne sont pas sensibles à l'enrichissement initial du combustible.
b) Les gaz de fission provenant de la cheminée d'une usine de retraitement peuvent être analysés «en ligne» par un spectromètre de masse quadrupole. Ce développement a été conseillé lors d'une réunion de l'AIEA [8].

2.3. Méthodes indirectes

2.3.1. Techniques neutroniques

Mesure des neutrons de fission spontanée. Le combustible plutonifère peut être analysé par la détection des neutrons de fission spontanée émis par les isotopes 240 et 242 du plutonium. La mesure des neutrons de fission spontanée dans un fond de neutrons provenant de réactions (α, n) est effectuée en analysant les corrélations entre les impulsions fournies par le détecteur. Ces corrélations reflètent la multiplicité de l'émission des neutrons de fission. L'application de cette méthode est simple dans le cas d'échantillons de petites dimensions. L'extension à des échantillons de plus grande taille devient par contre très complexe par suite de la multiplication des neutrons. Dans ce cas, il est assez difficile de mettre au point une procédure de mesures de routine en vue d'applications dans des conditions réelles d'inspection garantissant que les mesures effectuées auront une précision optimale. En conséquence, un programme, basé sur la méthode de Monte Carlo et simulant complètement la technique des corrélations, a été écrit. Ce simulateur permet de mettre au point les procédures optimales de mesure pour n'importe quel type d'échantillon avant de commencer une inspection. De plus, le programme est capable d'évaluer les corrections à appliquer aux données expérimentales brutes, telle, par exemple, la correction des facteurs de calibration lorsque l'échantillon mesuré s'écarte en géométrie ou en composition isotopique de l'échantillon standard utilisé pour la calibration. Les problèmes liés à l'inviolabilité d'une mesure peuvent être traités quantitativement par ce programme. Un instrument basé sur les autocorrélations intégrales a été construit et testé (fig.3). Ses performances ont été décrites dans la référence [9]. Un deuxième instrument est en cours de développement et permettra de mesurer, pour chaque neutron détecté, l'instant et la région de détection. Ces données détaillées contiennent des informations relatives à l'ordre de l'émission, à la multiplication et au spectre moyen des neutrons détectés. Il est prévu d'analyser cette information à l'aide d'un petit ordinateur en ligne, en vue d'obtenir une détermination plus précise et inviolable de l'émetteur de neutrons de fission spontanée.

Mesure du ^{235}U et du ^{239}Pu dans les combustibles mixtes. Les teneurs en ^{235}U et ^{239}Pu dans des combustibles mixtes peuvent être déterminées en mesurant les rendements en neutrons prompts et retardés émis lors de l'irradiation d'un échantillon par un flux de neutrons, d'énergie inférieure au MeV. La practicabilité de cette méthode a été étudiée théoriquement [10]. En liaison avec ces études, un instrument a été construit qui permettra de mesurer les neutrons prompts et retardés émis par une aiguille combustible du type RAPSODIE, irradiée par un flux de neutrons ($E < 1$ MeV) provenant d'une source Sb/Be de 300 curies.

Application de la technique des neutrons retardés. La technique des neutrons retardés, basée sur l'irradiation de matières fissiles à l'aide d'une source de neutrons et sur la mesure de l'émission de neutrons retardés, a été expérimentalement évaluée en vue de la détermination du contenu en matières fissiles d'échantillons de types variés. Un appareil

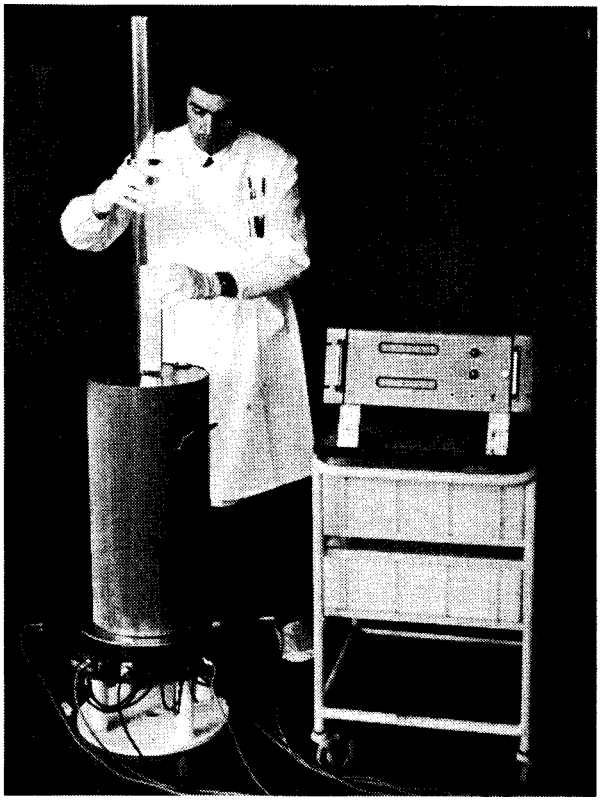

FIG. 3. Compteur de neutrons de fission spontanée.

a été développé permettant d'utiliser cette technique pour la détermination de l'uranium contenu dans des déchets provenant des usines de fabrication d'éléments du type MTR ainsi que dans des solutions purifiées provenant des usines de retraitement. Des sources de neutrons du type Am/Be et Sb/Be sont utilisées pour l'irradiation. La mesure des neutrons retardés s'effectue, après enlèvement de la source de neutrons, à l'aide de détecteurs du type BF_3 ou ^3He noyés dans du polyéthylène.

2.3.2. Techniques de spectrométrie gamma

Des techniques de spectrométrie gamma, basées sur les mesures d'émission et d'absorption du rayonnement gamma, ont été évaluées expérimentalement pour le contrôle d'éléments combustibles MTR irradiés et non irradiés [11, 4].

Pour la détermination du contenu en uranium et de l'enrichissement dans des éléments de combustible non irradiés, du type MTR, on a développé des instruments utilisant des cristaux NaI comme détecteur et

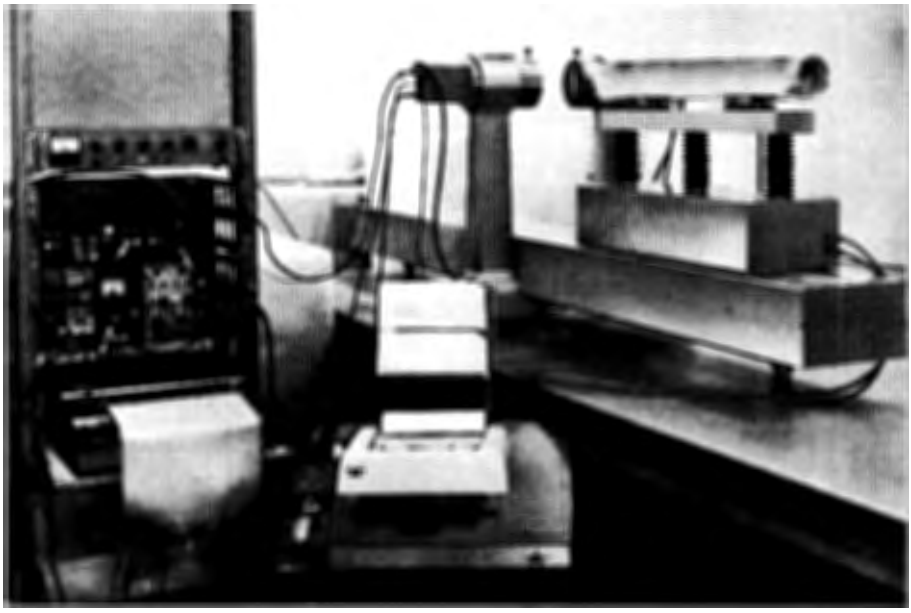

FIG. 4. Appareil de balayage gamma.

fonctionnant suivant le principe du balayage gamma (fig.4). Ces instruments sont utilisés en routine par le personnel de la Direction du Contrôle de sécurité lors des opérations de contrôle dans des usines de fabrication.

En vue de la détermination du contenu en uranium dans des éléments combustibles MTR irradiés, une technique d'absorption gamma a été testée expérimentalement. Un détecteur au Ge(Li) est utilisé pour la mesure des photons gamma de 145 keV, émis par le ^{141}Ce, qui sont atténués dans l'élément combustible.

Des études sont également en cours pour la détermination des nucléides du plutonium par spectrométrie gamma, à l'aide de détecteurs au Ge(Li).

2.4. Méthodes directes

Le plus gros effort dans ce domaine a été consacré, en collaboration avec la GfK à Karlsruhe, à l'automatisation de l'analyse par dilution isotopique et spectrométrie de masse. Les travaux d'automatisation de l'analyse complète, c'est-à-dire du traitement chimique et des mesures par spectrométrie, sont en cours [12, 13].

Dans le domaine de la spectrométrie d'émission, des expériences sont en cours visant à mettre au point une méthode de détermination absolue du rapport $^{235}U/^{238}U$ et pour augmenter d'une part la précision et d'autre part la vitesse de l'analyse.

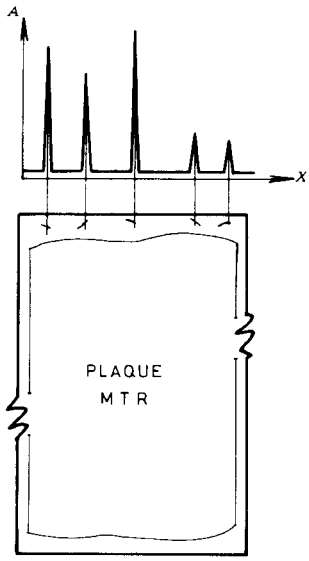

FIG. 5. Exemple d'une plaque MTR marquée avec des inclusions de tungstène et de l'enregistrement du signal ultrasonore correspondant.

2.5. Techniques d'identification

Un effort important a été fourni en vue du développement de techniques d'identification à preuve de fraude pour différents types d'éléments combustibles et pour les scellements [14]. L'effort principal a porté sur l'utilisation des ultra-sons pour l'identification de défauts naturels ou d'inclusions distribuées d'une manière aléatoire. Des études ont été faites également en vue de l'utilisation de la fluorescence X et des courants de Foucault. Les procédés d'enregistrement automatique et de vérification ultérieure de ces mesures ultrasonores sont en cours de développement: enregistrement analogique ou digital sur bande magnétique ou liaison directe avec un ordinateur, confrontation automatique d'identités et constitution d'archives. L'application de ces techniques a été recherchée pour différents types d'éléments combustibles et pour les scellements.

Eléments combustibles MTR. En vue de l'identification des éléments combustibles MTR, le marquage des plaques seules a été considéré en premier lieu. Les inclusions (morceaux de fils de tungstène) sont introduites facilement à l'intérieur d'une partie inactive de la plaque pendant le procédé normal de sa fabrication. La figure 5 montre un exemple d'une plaque marquée et de l'enregistrement du signal ultrasonore correspondant. Comme la majorité des plaques combustibles MTR sont assemblées en éléments combustibles qui ne peuvent pas être désassemblés sans destruction du boîtier, un procédé est actuellement à l'étude en vue du marquage et de l'identification de la plaque de rive du boîtier des éléments combustibles MTR.

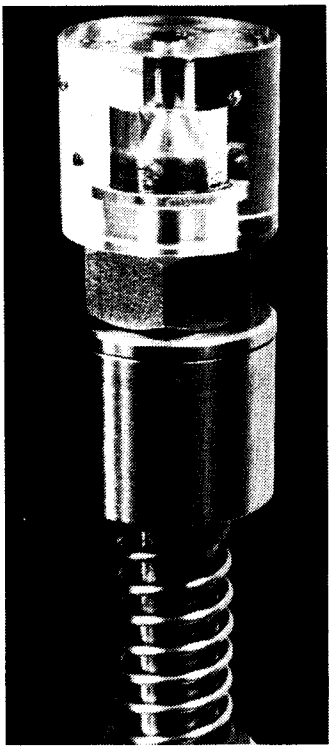

FIG. 6. Modèle d'un sceau de bloquage sur l'écrou de fixation d'une barre combustible.

Eléments combustibles LWR. En vue de l'identification des barres seules, l'enregistrement ultrasonore de la soudure d'un bouchon d'extrémité peut être utilisé. Cependant, il semble plus intéressant d'envisager un système d'identification pour l'assemblage entier par marquage d'une ou plusieurs parties spéciales du boîtier. Lorsque l'élément combustible peut être désassemblé, le scellement de l'assemblage peut être assuré par bloquage d'un ou plusieurs écrous de fixation au moyen de chapeaux avec inclusions qui peuvent être identifiées au moyen des ultra-sons. La figure 6 montre une application pratique de cette procédure.

Scellements. Différents types de sceaux à preuve de fraude ont été conçus. Le même principe de marquage par inclusions et de détection par ultra-sons ou par fluorescence X a été appliqué. Au moyen de la métallurgie des poudres, il est possible de fabriquer différents types de sceaux avec inclusions internes. La matrice des sceaux peut être un plastique ou un métal (aluminium, acier inoxydable, zirconium) selon l'utilisation.

REFERENCES

[1] LARISSE, J. et al., in Safeguards Techniques (C. r. Coll. Karlsruhe, 1970) II, AIEA, Vienne (1970) 333.
[2] LARISSE, J., Revue française de l'informatique et de recherche opérationnelle, R3, Série rouge (1970) 91.
[3] Rapport sur l'exercice de contrôle JEX 70, Rapport EUR et KFK sous presse (1971).
[4] MIRANDA, U. et al., in Safeguards Techniques (C. r. Coll. Karlsruhe, 1970) I, AIEA, Vienne (1970) 105.
[5] WINTER, H. et al., Rapport KFK-904 (1969).
[6] KOCH, L., BRAUN, H., CRICCHIO, A., in Safeguards Techniques (C. r. Coll. Karlsruhe, 1970) I, AIEA, Vienne (1970) 539.
[7] ARIEMMA, A. et al., Rapport EUR-4638 (1971).
[8] IAEA Safeguards Research Coordination Meeting on Instrumental Techniques for Determining Nuclear Material Flow in Reprocessing Plants (Vienne, déc. 1970), rapport interne non publié.
[9] BIRKHOFF, G. et al., in Safeguards Techniques (C. r. Coll. Karlsruhe, 1970) II, AIEA, Vienne (1970) 477 (résumé).
[10] BIRKHOFF, G., BONDAR, L., LEY, J., Rapport EUR sous presse (1971).
[11] KREYGER, P.J., HARRY, R.J.S., KRÖCKEL, H., in Safeguards Techniques (C. r. Coll. Karlsruhe, 1970) I, AIEA, Vienne (1970) 545.
[12] WILHELMI, M. et al., Ibid., II, p. 165.
[13] BAECKMANN, A., von et al., Ces Actes, vol. 9, mémoire 809.
[14] JEHENSON, P.S. et al., in Safeguards Techniques (C. r. Coll. Karlsruhe, 1970) II, AIEA, Vienne (1970) 215.

IAEA
INVITED REVIEW PAPER
A/CONF.49/P/770

DEVELOPMENT OF THE IAEA SAFEGUARDS SYSTEM FOR NPT

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Abstract–Résumé–Аннотация–Resumen

DEVELOPMENT OF THE IAEA SAFEGUARDS SYSTEM FOR NPT.

Against the background of today's nuclear technology and of the fundamental ideas governing international safeguards work in connection with the Non-Proliferation Treaty (NPT), the Agency's Safeguards System is basically an information system relying on the technical safeguards measures: accountancy, containment and surveillance, and making use of procedural steps such as design examination, record keeping, reporting and inspections. The paper specifies the objectives for and the technical and organizational aspects of the developmental work being carried out to adapt the Agency's Safeguards System to the NPT situation. The paper reviews the working methods and results of the Safeguards Committee, which worked out recommendations for a complete agreement text, and it is pointed out how the further quantification steps are relegated to the "Subsidiary Arrangements". The paper finally evaluates the need for further development of objectives, such as the replacement of human observation by instrumental techniques, simplified information requirements in distinct cases, the utilization of correlation techniques, etc., and the need for continuous developmental effort to iterate the safeguards system to an optimum.

ELABORATION D' UN SYSTEME DE GARANTIES DE L' AIEA DANS LE CADRE DU TNP.

Dans le cadre de la technologie nucléaire d' aujourd' hui et des grands principes régissant les activités internationales de garanties au titre du Traité sur la non-prolifération (TNP), le système de garanties de l' Agence est essentiellement un système d' information fondé sur l' application de mesures techniques: comptabilité matières, confinement et surveillance, complétées par des modalités comme l' étude des renseignements descriptifs, la tenue d' une comptabilité, la présentation de rapports et les inspections. Le mémoire définit les objectifs et les aspects techniques et organiques des travaux de mise au point actuellement en cours pour adapter le système de garanties de l' Agence à la situation créée par l' entrée en vigueur du TNP. Il passe en revue les méthodes de travail utilisées et les résultats obtenus par le Comité des garanties qui a élaboré et recommandé un texte complet d' accord, en soulignant que les détails pratiques d' application feront l' objet d' «arrangements subsidiaires». Enfin, le mémoire évoque le besoin de se rapprocher davantage d' objectifs tels que le remplacement de l' observation humaine par l' emploi d' appareils automatiques, la simplification des renseignements à communiquer (dans certains cas particuliers), l' utilisation de la méthode des corrélations, etc. et souligne la nécessité d' un effort continuel de mise au point pour perfectionner sans cesse le système de garanties.

РАЗВИТИЕ СИСТЕМЫ ГАРАНТИЙ МАГАТЭ В СВЯЗИ С ДОГОВОРОМ О НЕРАСПРОСТРАНЕНИИ ЯДЕРНОГО ОРУЖИЯ.

На фоне современной ядерной технологии и основных идей, определяющих деятельность по международным гарантиям в связи с Договором о нераспространении ядерного оружия, система гарантий Агентства является, в основном, информационной системой, зиждущейся на отчетности при использовании технических методов осуществления гарантий, на опечатывании и наблюдении. При этом используются такие процедурные меры, как рассмотрение конструкции, составление учетных документов, представление отчетов и инспекции. В докладе излагаются цели, а также технические и организационные аспекты соответствующей работы по приспособлению системы гарантий Агентства к ситуации, созданной Договором о нераспространении ядерного оружия. В докладе рассматриваются оперативные методы и результаты деятельности Комитета по гарантиям, подготовившего рекомендации по всему тексту соглашения о гарантиях, а также указывается на то, какие дальнейшие меры по количественному определению относятся к "Дополнительным положениям". Наконец, в докладе дается оценка необходимости дальнейшей работы над такими вопросами, как замена наблюдения, проводимого человеком, инструментальными методами, в конкретных случаях – упрощение требований необходимой информации, применение корреляционных методов и т.д., указывается необходимость постоянных усилий по поддержанию системы гарантий на оптимальном уровне.

DESARROLLO DEL SISTEMA DE SALVAGUARDIAS DEL OIEA PARA LA APLICACION DEL TNP.

Considerado en el contexto de la tecnología nuclear actual y de los conceptos fundamentales acerca de las actividades internacionales de salvaguardia relacionadas con el Tratado sobre la no proliferación de las armas nucleares (TNP), el sistema de salvaguardias del Organismo constituye esencialmente un sistema de información basado en las medidas técnicas, contabilidad, contención y vigilancia, y que aplica procedimientos tales como la información sobre el diseño, mantenimiento de registros, presentación de informes e inspecciones. La memoria expone las objetivos y los aspectos técnicos y orgánicos de las actividades desarrolladas para adaptar el Sistema de salvaguardias del Organismo a las circunstancias del TNP. Se pasa revista a los métodos de trabajo y resultados del Comité de Salvaguardias, que elaboró recomendaciones para la formulación íntegra de un acuerdo modelo de salvaguardias, destacándose el hecho de que los detalles de índole cuantitativa se relegan a los «Arreglos Subsidiarios». La memoria examina finalmente la necesidad de conseguir nuevos objetivos, tales como el reemplazamiento de las observaciones humanas por técnicas instrumentales, la simplificación de la información requerida en determinados casos, la utilización de técnicas de correlación, etc., así como la necesidad de un esfuerzo continuo de desarrollo para perfeccionar al máximo el sistema de salvaguardias.

1. RECOGNITION OF THE NEED FOR FURTHER DEVELOPMENT OF THE AGENCY'S SAFEGUARDS SYSTEM

1.1. The successful conclusion of the Treaty of Non-Proliferation of Nuclear Weapons was the culmination of ten years of effort in the United Nations and in the Conference of the Eighteen-Nation Committee on Disarmament. Well before the General Assembly of the United Nations adopted a Resolution commending the Treaty and expressing the hope for the widest possible adherence to it in June 1968, the Agency already had a quite well-developed Safeguards System in use for similar aims. Naturally, therefore, the Treaty explicitly designates the Agency for implementing safeguards in connection with the Treaty and in accordance with its Statute and existing system.

1.2. Within the Agency's Secretariat it was recognized at an early stage that considerable preparation was needed to enable the Safeguards Department to cope with the new task, not only with a view to adapting the organization to a rapidly expanding workload, but mainly to developing further and systematically detailed safeguards techniques.

This was, for instance, stated at the opening of a Panel of Experts on Safeguards Techniques invited to Vienna in August 1967 [1]. To formulate technical approaches to various aspects of safeguards, five more such Panels were held during the years 1968 and 1969 [2]. In addition, the Director General appointed in 1968 a group of eight experts as consultants to study and report on the impact of NPT on the Agency's Safeguards work [3]. In fact, these consultants re-formulated some basic concepts. At the same time, a new Division, devoted to Safeguards development work, was established within the Safeguards Department. In 1970 an IAEA Symposium on Safeguards Techniques was held in Karlsruhe [4] and experts from several Member States were at the IAEA Headquarters to participate in four different working groups.

1.3. Three weeks after the coming into force of the Non-Proliferation Treaty on 5 March 1970, the Board of Governors of the Agency decided to establish a Committee on which any Member State could be represented and which was given the task, among others, to: "advise the Board as a matter

of urgency on the Agency's responsibilities in relation to safeguards in connection with the Treaty, and in particular on the content of the agreements which will be required in connection with the Treaty".

Within nine months (June 1970 to March 1971) the Committee, with the participation of Delegations from some 50 Member States, completed the required work, including the additional task of developing and recommending a financing scheme for the Agency's safeguards activities.

2. THE BASIS FOR THE SAFEGUARDS COMMITTEE'S WORK

2.1. In general terms the Non-Proliferation Treaty [5] itself determines the point of departure for the Committee's work, and this in three ways.

Firstly, the safeguards agreements to be negotiated and concluded between the States party to the Treaty and the International Atomic Energy Agency should be based on the Statute of the latter.

Secondly, the safeguards functions should be defined according to the Agency's Safeguards System for the exclusive purpose of verification of the fulfilment of a State's obligation assumed under the Treaty "with a view to preventing diversion of nuclear energy from peaceful uses to nuclear weapons or other nuclear explosive devices".

Thirdly, the principle would be applied of safeguarding effectively the flow of source and special fissionable material by the use of instruments and other techniques at certain strategic points.

In addition, one of the very basic principles already set forth in the Agency's existing Safeguards System is explicitly repeated in the Non-Proliferation Treaty. That is, that safeguards should be designed to avoid hampering the economic or technological development of the Parties, or international cooperation in the field of peaceful nuclear activities, including the international exchange of nuclear material and equipment for the processing, use or production of nuclear material for peaceful purposes. This is not only meant as an instruction to minimize intrusion – there are many technical articles of the proposed agreement to take care of that – but also reflects the fundamental idea of the Treaty that safeguards should be able to follow any peaceful nuclear activity without limiting or directing it. In that sense it belongs to the most important criteria for further system development.

2.2. The Agency's Safeguards System, which now has been in use for over 10 years and is still being used, also provided a major input for the Committee's work. The Safeguards System had been revised and improved several times, the last time in 1968 by a Committee of the Whole of the Board of Governors and is generally known under the reference INFCIRC/66/Rev.2 [6]. It contains four well-known procedural elements, namely,

(a) The provision of design information, to supply the inspectorate with a defined and limited knowledge of the facilities in which nuclear material is produced, used, processed or stored.
(b) The requirement for the operators of such facilities to keep records on material production, change and movement.

(c) The provision of reports on material production, change and movement to the safeguarding authority.
(d) The independent verification, by inspections of recorded and reported data as well as of material flow and inventory.

These four procedural elements are common to all safeguards systems presently in use; they appear in domestic and bilateral safeguards as well as in the regional systems of the European Nuclear Energy Agency, or the European Community (Euratom). Naturally, they were taken over as the main procedural part for non-proliferation safeguards.

2.3. However, conceptual studies by the above-mentioned consultants, systems analysis work done within the Secretariat of the Agency and in Member States, and results from integral safeguards experiments and experience gained in years of safeguards practice have led to new formulations and important expansions.

Systems analysis work in particular showed the need to determine to which technical conclusions the application of safeguards should lead. None of the existing safeguards systems had provided for this. The most suitable basis for such a provision was found in the notion "material unaccounted for (MUF)". The amount of this MUF in respect of a given area and over a specific period should be stated as a result of the Agency's verification activities, giving also the limits of accuracy.

Systematic analysis, by the Secretariat and an expert working group which met in 1970, of the optimal means by which this objective can be reached shows that:

(i) Material accountancy should be adopted as the safeguards measure of fundamental importance, with
(ii) Containment and
(iii) Surveillance as important complementary measures.

This formulation has been incorporated as one of the principal articles in the material which is to be used as the basis for the negotiation of the agreements that should be concluded between the Agency and States Party to the Non-Proliferation Treaty.

In practice, bookkeeping can actually be the backbone of a safeguards system because today's nuclear technology centres on the fission of nuclei contained in only two chemical elements used almost exclusively for nuclear purposes: uranium and plutonium.

2.4. It might be appropriate at this stage to attempt an intermediate summary of the fundamental ideas governing the international safeguards work in connection with the Non-Proliferation Treaty:

The Treaty determines the political aim as well as the political constraint: prevention of diversion of nuclear material from peaceful activities – which as such are not limited – to nuclear explosives and weapons.

This political aim must be realized within the framework of today's nuclear technology. From this the technical objective of safeguards can be derived as: the detection of amounts of nuclear material that might be missing from peaceful nuclear activities, defined by the precise notion "material unaccounted for".

This makes the Safeguards System basically an information system relying on the technical safeguards measures: accountancy, containment and surveillance, and making use of the procedural steps: design review, records, reports and inspections.

2.5. Against this background safeguards systems development is mainly concerned with technical and organizational aspects. Although the Safeguards Committee naturally also had some political aspects to consider, this paper will mainly deal with the technical and organizational matters.

Returning to the history of preparations for the Committee's work, the important concept of a "national (or regional) system of accountancy and control of nuclear material" now has to be introduced:

The sequence of early agreement drafts reveals a gradual development of that concept. The basic idea is in fact already implied in the original Agency system by the fact that records and reports requirements are formulated as an obligation on the Government Party to a safeguards agreement with the Agency. Many discussions on the interface between the three levels at which safeguards activities take place – those of the facility operator, those of governmental organizations of the State concerned, and those of an international body – ended in suggesting that the establishment of a "State's system", with well-defined technical functions, would be an essential obligation to be incorporated in the safeguards agreements.

2.6. Optimization studies on the information system covering the complete fuel cycle (as distinct from the previous system related to individual facilities) showed that more emphasis should be placed on timely reporting to the inspectorate. Therefore a concept, put forward by the consultants, of reporting monthly on every transaction of significant amounts of nuclear material was introduced in the form of a requirement for "Inventory Change Reports".

So as to enable the inspectorate to form a clear picture of the nuclear material flow a scheme was developed to subdivide the nuclear fuel cycle into distinct "Material Balance Areas". Only material transactions over the boundaries of such areas would be the object of inventory change reports.

Finally, the question had to be considered how records would have to be kept, particularly in respect of nuclear material flowing through a process and losing its identity from one end to the other, and how reports would be made. The most practical approach seemed to be the use of what facility operators usually call and handle as a "batch of material". It means a portion of material handled as a unit and for which the composition and quantity are defined by a single set of specifications or measurements. Within that definition a batch can be composed of many similar items or of a volume of solution divided over several vessels. Thus it allows very concise reporting.

2.7. Whereas these technical concepts resulted from a cooperative effort between the Secretariat and a considerable number of experts, considerable input to the development work was also derived from early consultations with a Member State on the possible conclusion of an agreement in the spirit of NPT. These brought about many valuable clarifications on legal and organizational matters; the most striking one was the structuring of the agreement in two main parts. The first part should set forth the fundamental

rights and obligations of the parties and contain such other provisions that are unlikely to require modification. The second part should set forth the technical principles and safeguards procedures to be employed and, wherever possible, a simpler amendment procedure should be envisaged for it.

2.8. The list of sources for the ideas and concepts underlying the NPT Safeguards System is not yet complete. At the time when the Board of Governors of the Agency decided to establish the Safeguards Committee it also invited all Member States to communicate to the Director General of the Agency their views on these matters. Thirty-one Member States responded to this invitation by transmitting comprehensive memoranda on the subject to the Secretariat just before and during the first months of the Committee's work.

2.9. In view of the wealth of material it is not possible to give more than a brief summary of it here.

A major part of the demands for an improved system for NPT safeguards was related to the wish to determine in advance as precisely and as completely as possible what safeguards would consist of in each case. This included the wish to define precisely such basic technical notions as Material Balance Area, Key Measurement Point, Design Information, etc. to quantify the entire verification effort, to fix the mode, intensity and duration of inspections, and to determine the point in the fuel cycle where safeguards should start, the latter being coupled to the idea of moving this starting point rather far up in the line, away from the uranium and thorium mines and ore concentration mills.

Somewhat in contradiction to, but often parallel with, the attempt to achieve a higher degree of precision and predetermination, ideas were formulated on how to keep the system open to technical adaptation necessitated by scientific and technological progress.

An important number of suggestions dealt with the protection of commercial and industrial knowledge, mostly to the effect that such knowledge should remain a secret from the inspectorate as long as it was not absolutely necessary for effective safeguards. Accordingly, design information, inspection access and inspection effort should be limited.

Proposals for simplifying the safeguards system and for improving cost effectiveness were numerous. They dealt mainly with two important aspects: the unified nuclear material inventory per State, and the gradation of verification effort according to the potential risk of diversion.

Finally, it should be mentioned that many communications contained in one form or another the suggestion to elaborate a typical safeguards agreement in great detail, so that it could serve as a model and would represent the improved system of the Agency for Safeguards under the Non-Proliferation Treaty.

2.10. The first report of the Director General to the Safeguards Committee incorporated all the material from the practical experience with the existing system, the draft agreements worked out by several joint staff working groups in the Secretariat, the panel recommendations, the conclusions of the consultants, the systems studies of the newly formed Division of Development and the results of the discussion with the delegation of one Member State, and finally from the communications of 31 Member States. It also contained the

introductory remarks, a proposal for the first fundamental part of an agreement, and explanatory notes for a second part on technical implementation procedures. Only after the first round of Committee meetings was it possible, during the summer of 1970, to draft the second part in a form as near as possible to the structure and content of an agreement. After two weeks of extensive discussions of the explanatory notes in the Committee, numerous comments from many delegations could be worked into that draft.

3. THE COMMITTEE'S WORKING METHOD AND RESULTS

3.1. After a long general debate the Committee decided to use the above-mentioned two drafts – two parts of a typical NPT safeguards agreement – suggested by the Director General as a basis and agenda for further work. Article by article was discussed and the relation of each to other parts of the text analysed. This latter was the most difficult part of the step-wise development process. The technical part of a safeguards agreement in its entirety must give a solid framework for an effective information system. Adjustments to any one part of the framework call for compensatory adjustments to other parts.

Without departing from the principle that all delegations should be able to participate in all deliberations of the Committee, consultations took place in smaller groups. Informally and voluntarily, working parties gathered together to prepare the more difficult issues for the plenum discussion. For instance, in order to evaluate the maximum routine inspection workload to be fixed in the agreement, case studies carefully prepared by different delegations and by the Secretariat were compared. This work led, e.g. to the proposal of a completely new article on inspection workload which the Committee could endorse without much discussion.

In the Committee itself decisions were approached by an iteration process: after the first analysis of an article, either one, or several delegations jointly, would submit amendments in writing. The examination of these often led to further amendments and discussions. Only the last touch was applied by amendments suggested orally.

3.2. In this way the Committee worked out recommendations for a complete agreement text consisting of 116 articles and including definitions of the most important technical notions. This material represents the Safeguards System of the Agency which must be used as the basis for negotiating and concluding agreements between States party to the Non-Proliferation Treaty and the Agency. It was published in May 1971 as document INFCIRC/153 [7].

On the following pages an attempt is made to describe the technical parts as a coherent information system. How it will be used in practice is shown by way of an example in the paper by Shmelev [8].

3.3. As mentioned before when discussing the technical objective of safeguards, the system must provide the inspectorate at any time within a defined period with a complete picture of the whereabouts of all nuclear material in a State and it must fulfil a great number of boundary conditions, such as being unintrusive, guaranteeing protection of information received, making use wherever possible of the fact that nuclear material is safely contained for long periods, providing for the use of statistical measuring

techniques and random sampling, providing for the utilization of the accountancy system of the States by verifying its findings, e.g. by independent measurements and observations, etc. Thus, the real problem is not to lay down the need for information – that would be simple – but to define the minimum of readily available information and the minimum amount of additional measurements that will fulfil the purpose of the Agency's system. Therefore, the description of that system looks rather like an enumeration of its limitations.

3.4. These limitations vary with the type of information. That information which is called for in the first procedural step, i.e. design information, and which should provide the Agency with the more or less static picture of a chain of facilities through which nuclear material may flow, is limited in three ways. The first limitation is the general condition that it should be the minimum necessary for safeguards; the second one is contained in a positive enumeration of the specifications which must be given: geographic location, type, nominal capacity, etc.; and the third and sharpest limitation is given in the definition of the purpose of examining this design information.

The most important result to be achieved by this examination is to determine Material Balance Areas to be used for Agency accounting purposes and to select those Strategic Points which are Key Measurement Points for determining nuclear material flow and inventories. The Committee even specified detailed criteria for material balance area selection. For instance, on the request of a State a particularly small material balance area may be established around a process step involving commercially sensitive information. The idea being that if there is only a small hold-up of nuclear material within such an area, and continuous input and output flow measurements can be made, this would make it possible to avoid inspection access to the area itself.

Thus, in connection with the provision of design information two principles are established for the protection of commercial and industrial secrets. In fact, there are two classes of such secrets. Firstly, those which must in any case be disclosed to the inspectorate which, in turn, must guarantee their protection; these consist of information related to nuclear material flow, such as the capacity of a fuel fabrication plant. Secondly, there is the group of industrial secrets, mainly process know-how, of which the disclosure to the Agency's inspectorate can be avoided. The criterion for selection of a particularly small material balance area around a sensitive process is an example of this second principle.

Other results expected from the examination of design information are the establishment of:

(a) Records and reports requirements and records evaluation procedures,
(b) Requirements and procedures for verification of the quantity and
(c) Location of nuclear material,
(d) Nominal timing and procedures for physical inventory taking.

In addition, the selection of appropriate combinations of containment and surveillance methods and techniques and the strategic points for their application should result from the examination of design information.

This enumeration shows how far the Committee went in formulating precise technical procedures and the reasons for them.

However, there is one more step to be taken: quantification. For practical reasons the agreement itself, which has to be concluded independently of the quantity of nuclear material required to be safeguarded and the number and type of facilities through which this might flow, cannot give all the quantitative values involved. Therefore in the agreement the quantification step is relegated to "Subsidiary Arrangements"; these will be described later. At this stage it has to be mentioned that the results of the examination of the design information, such as the selection of material balance areas and key measurement points, etc. have to be included in such Subsidiary Arrangements.

3.5. With regard to the information on nuclear material flow and inventories, the limitations could be handled in a much simpler way. Both the new record system, dealing with that information which has to be continuously accumulated at the facilities, and the new reports system, covering the information to be provided regularly to the Agency, are straightforward developments from the Agency's existing Safeguards System. What in the latter was a skeleton has now become much more fully formalized by stating all types of records and reports and prescribing their content. Thus, as mentioned in the introduction, the concept of transaction or inventory change reports has also been formally incorporated into the system. Again, quantification takes place in the Subsidiary Arrangements; these should for instance specify the amounts of nuclear material which, if inadvertently lost, would trigger the submission of a special report to the Agency.

3.6. A great step forward in formalization was also made concerning inspection procedures. Clear limitation of inspection rights without curtailing verification efficiency was one of the major concerns of the Committee. A complete definition of the purpose and scope of inspection was used to establish indirectly the limitations.

There are three types of inspections connected with different purposes: ad hoc inspections which include verification of the initial report on nuclear material, verification and identification of changes before the routine scheme of inspection is established and verification of nuclear material before and after international transfers; routine inspections with the purposes of verifying consistency between records and reports, of verifying location, identity, quantity and composition of nuclear material and of possible causes for MUF (material unaccounted for); special inspections which are obviously concerned with the verification of special reports but also with such additional investigations as are needed when the information obtained through normal means is not adequate for achieving the technical objective.

The scope of inspections is determined by giving a long list of actions permitted to the inspector. Typical examples are independent measurements of nuclear material, verification of the functioning and calibration of instruments, observation of sampling and analysis, arrangement for the receipt of duplicate samples, etc., but described with a higher degree of precision than in this incomplete enumeration.

The first direct limitation concerns inspection access. The decisive element is that routine inspections are limited to strategic points specified in the Subsidiary Arrangements and to the records. There is a safety valve for additional access under special circumstances, but this must be preceded by consultations between the State and the Agency.

The second direct limitation on inspections is given by stating the maximum yearly workload (in man-years) to be deployed per group of facilities. For reactors and sealed stores this is one-sixth of a man-year of inspection for each such facility in the State. For all other types of facilities the inspection workload is made dependent on throughput or inventory of nuclear material, expressed in effective kilograms to take care of the gradation in the diversion risk. For facilities involving plutonium or more than 5% enriched uranium, the annual workload may be as high as 30 times the square root of the number of effective kilograms in inventory or throughput, whichever is the greater, and not lower than 1.5 man-years. This is valid for e.g. reprocessing plants, whereas fabrication plants for power reactor fuel are covered by a simpler formula: one-third of a man-year plus 0.4 times the amount of effective kilograms.

Such is the framework of inspection rights; again quantification is left to the Subsidiary Arrangements, in which, for example, the number and location of strategic points will be fixed. On the other hand, it is not possible to determine the actual inspection workload in advance. This must be left to future experience, but criteria to be taken into account are already enumerated in the NPT agreements. An example of their application is given in Ref. [8].

3.7. The Committee also succeeded in determining precisely the fields of activity within which nuclear material has to be followed by safeguards. There were a number of functional boundaries to be defined, such as exemptions for small quantities or non-nuclear use, or termination of safeguards upon consumption or dilution of nuclear material, etc. The most important boundary is given by the definition of that point in the nuclear fuel cycle at which nuclear material has to come under safeguards for the first time. After careful consultations it was set as the point where any nuclear material reaches a purity and composition suitable for fuel fabrication or enrichment.

3.8. An essential feature of the proposed agreement is the complete description of a versatile feedback system from the Agency to the State. Three types of statements are required from the Agency. Firstly, a semi-annual statement of the nuclear material inventory on the books of the Agency, which permits a check on the completeness of the information flow. Secondly, information on the result of inspections in the State, at intervals to be specified in the Subsidiary Arrangements. And finally, a statement on the technical conclusions drawn from verification activities, i.e. the difference between book inventory and physical inventory, or, expressed in another way, the material unaccounted for (MUF), as well as the accuracy with which this quantity can be known. Through this feedback system the safeguards information flow gains an additional justification.

3.9. Finally a word of explanation has to be said on the Subsidiary Arrangements. In the Agency's Safeguards practice up to now this kind of document has figured as an additional protocol concluded between the Secretariat of the Agency and a State party to a safeguards agreement, in order to fix all the detailed procedures and technicalities left open in the agreement itself. In the new and more complete type of agreement many of these details are incorporated. But Subsidiary Arrangements are still necessary to lay down

those quantities, dates and numbers which, within the validity of the agreement, undergo a steady development parallel with the growth of nuclear activities. In more than 20 agreement articles the phrase appears: "as specified in the Subsidiary Arrangements". Contrary to previous practice these Subsidiary Arrangements do not contain any rights or obligations additional to those in the NPT agreements, but consist of a series of tables, information sheets, report forms, etc., and of course indicate the mechanism for keeping them up to date. As mentioned before, Subsidiary Arrangements are the instrument for quantification of safeguards, as far as this is at all possible at this stage.

4. NEED FOR FUTURE DEVELOPMENT

4.1. The foregoing presentation of the Agency's Safeguards, to be applied in the frame of the Non-Proliferation Treaty, brings out the best sides of the improved system. Naturally, any presentation concerned with the results of development work tends to emphasize the progress made. However, this should not obscure the fact that there is much left to be done.

4.2. The replacement of human observation and surveillance by instrumental techniques, for instance, is far from being as advanced as one might think. Very interesting measuring devices have been invented in recent years but the development of what is often referred to as "black boxes", i.e. automatic accounting and surveillance instruments, which have been imagined as the solution to many problems, has not yet progressed sufficiently to make them immediately applicable. The draft agreement foresees that the Agency shall take full account of technological developments and that, if so agreed and specified in the Subsidiary Arrangements, installation of such equipment may be initiated. So the way is paved for step-wise, practical improvements.

4.3. On the other hand, it has not been possible so far to use all the valuable theoretical conclusions of systems analysis work for practical adaptations of the Safeguards System. Particularly, there might still be some room for simplifying information requirements on nuclear material flow in States where a more or less complete nuclear fuel cycle is being safeguarded. In those cases correlations between the material flow through different stages of the cycle tend to reduce the total number of independent measurements necessary for verifying the entire flow picture.

The Agency is preparing to make use of such correlation techniques in its computerized program for the evaluation of accounting and inspection reports. These techniques are also taken into account in estimates of future safeguards workloads based on simulated nuclear fuel cycles [9]. But, of course, these computations have to be repeated each year with input data corrected to the actual situation. Thus, a step-wise approach of the theoretical picture to reality is achieved. In the same way, by checking step by step with the results of practical experience, further optimization of the information requirements will be reached.

4.4. To the mathematician and systems analyst, approximation by steps is a well-known process; he even has a precise term for it, iteration. In practice it means that the way to reach an optimum is understood; however, a continuous effort is required.

REFERENCES

[1] INTERNATIONAL ATOMIC ENERGY AGENCY, Panel on Safeguards Techniques, Vienna, 1967, IAEA PL-256 (1967).

[2] INTERNATIONAL ATOMIC ENERGY AGENCY, Panel on Safeguards Technical Practices for Irradiated Fuel Plants, Vienna, 1968, IAEA PL-279 (1968).
INTERNATIONAL ATOMIC ENERGY AGENCY, Panel on Safeguards Methods for Reactors, Vienna, 1968, IAEA PL-315 (1968).
INTERNATIONAL ATOMIC ENERGY AGENCY, Panel on Safeguards Methods for Conversion and Fuel Fabrication Plants, Vienna, 1969, IAEA PL-332 (1969).
INTERNATIONAL ATOMIC ENERGY AGENCY, Panel on Safeguards Systems Analysis of Nuclear Fuel Cycles, Vienna, 1969, IAEA PL-352 (1969).
INTERNATIONAL ATOMIC ENERGY AGENCY, Panel on Safeguards Methods and Techniques with Special Reference to Inspections, Tokyo, 1969, IAEA PL-368 (1969).

[3] INTERNATIONAL ATOMIC ENERGY AGENCY, Vienna, 1969, GOV/INF/212.

[4] INTERNATIONAL ATOMIC ENERGY AGENCY, Safeguards Techniques (Proc. Symp. Karlsruhe, 1970), Vols 1 and 2, IAEA, Vienna (1970).

[5] See e.g. INTERNATIONAL ATOMIC ENERGY AGENCY, INFCIRC/140, 1970.

[6] INTERNATIONAL ATOMIC ENERGY AGENCY, INFCIRC/66/Rev.2, 1968.

[7] INTERNATIONAL ATOMIC ENERGY AGENCY, INFCIRC/153, 1971.

[8] GMELIN, W., HOUGH, G., SHMELEV, V., SKJÖLDEBRAND, R., "A technical basis for international safeguards", these Proceedings, Paper 773, Vol.9.

[9] SHMELEV, V., "Progress in safeguards systems analysis", Atom. Energy Rev. 8 2 (1970) 417.

CONTROL OF RADIOACTIVE SOURCES AND SPECIAL NUCLEAR MATERIAL IN BRAZIL

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Abstract–Résumé–Аннотация–Resumen

CONTROL OF RADIOACTIVE SOURCES AND SPECIAL NUCLEAR MATERIAL IN BRAZIL.

The object of this paper is to describe the steps currently being taken for control of special nuclear materials in Brazil. It describes the control exercised by the International Atomic Energy Agency (IAEA) through the trilateral agreement between Brazil, the United States of America and the IAEA. It also describes the relationship between the National Nuclear Energy Commission and the Institutes and private organizations. The paper deals as well with the criteria which will be applied to the development of this control system as a method of safeguards, with special attention being paid to its effectiveness, since it is also needed as a means of correct administration of these materials by the National Nuclear Energy Commission.

CONTROLE DES SOURCES RADIOACTIVES ET DES PRODUITS FISSILES SPECIAUX AU BRESIL.

Le mémoire a pour objet de décrire les mesures actuellement prises au Brésil pour contrôler les produits fissiles spéciaux. Il décrit les mesures prises par l' Agence internationale de l' énergie atomique à cet effet en application de l' accord tripartite entre l' Agence, le Brésil et les Etats-Unis. Il décrit aussi les rapports existant entre la Commission nationale de l' énergie nucléaire et les instituts et organisations privées. Le mémoire analyse également les critères qui seront appliqués pour mettre au point ce système de contrôle en tant que méthode de garanties, en tenant compte particulièrement de son efficacité puisqu' il est également nécessaire pour assurer l' administration rationnelle des produits fissiles spéciaux par la Commission nationale de l' énergie nucléaire.

КОНТРОЛЬ ЗА РАДИОАКТИВНЫМИ ИСТОЧНИКАМИ И СПЕЦИАЛЬНЫМ ЯДЕРНЫМ МАТЕРИАЛОМ В БРАЗИЛИИ.

Целью доклада является описание принимаемых в настоящее время мер по контролю за специальными ядерными материалами в Бразилии. В нем рассматриваются вопросы контроля, осуществляемого Международным агентством по атомной энергии (МАГАТЭ) по трехстороннему соглашению между Бразилией, Соединенными Штатами Америки и МАГАТЭ. Описываются также взаимоотношения между Национальной комиссией по ядерной энергии, институтами и частными организациями. Рассматриваются критерии, которые будут применяться при разработке этой системы контроля как метода гаранитий, с уделением особого внимания ее эффективности, так как она необходима для правильного распределения указанных материалов со стороны Национальной комиссии по ядерной энергии.

EL CONTROL DE LAS FUENTES RADIACTIVAS Y DE LOS MATERIALES NUCLEARES ESPECIALES EN EL BRASIL

El objeto de esta memoria es describir el estado actual del control de materiales nucleares escpeciales en el Brasil, comprendido el control que ejerce el Organismo Internacional de Energía Atómica (OIEA), en virtud del Acuerdo trilateral entre el Brasil, los Estados Unidos de América y el OIEA. Se describen las relaciones entre la Comisión Nacional de Energía Nuclear y otras instituciones y entidades privadas. Se exponen también los criterios que se aplicarán al desarrollarse el sistema de control como método de salvaguardias, prestándose especial atención a la eficacia, indispensable para una correcta gestión de los materiales por parte de la Comisión Nacional de Energía Nuclear.

1. INTRODUCTION

The installation of the first nuclear power plant in Brazil was decided by the Brazilian Government in late 1967. The following year an agreement was signed for mutual collaboration between the National Nuclear Energy Commission (CNEN) and the Centrais Brasileiras S/A (ELETROBRAS) for the establishment of rules to be used for the planning, construction and operation of a nuclear power plant [1]. In the same year, at the request of the Brazilian Government, the IAEA commissioned a study team of international experts to assist the Brazilian authorities in preparing plans for development of nuclear power in Brazil. The primary conclusion of the study team was that the introduction of nuclear power in South Central Brazil could be justified for the early 1980s under the assumptions which were presented [2]. The Study Team advised that the procurement of a proven type of nuclear power plant, to go into operation in 1976/7, was desirable as a means of initiating an effective nuclear power program.

Following this recommendation the Brazilian Government scheduled installation of the Angra Nuclear Power Plant in South Central Brasil, State of Rio de Janeiro, scheduled to be connected to the network in 1976. It will be a 600-MW(e) PWR [3].

The nuclear program that the CNEN intends to pursue in the next few years underlines the importance of nuclear material management in the country. In connection with this, one of the first steps made by CNEN was to send some of its professionals abroad to be trained.

This paper discusses a proposal for the utilization of safeguards methods as a requirement for better management of nuclear materials by CNEN.

2. CURRENT LEGAL ASPECTS OF NUCLEAR MATERIAL CONTROL BY THE NATIONAL NUCLEAR ENERGY COMMISSION

Pursuant to "Lei e Regulamento da Comissão Nacional de Energia Nuclear" [4], Chapter II, Section 1, Art. 4, §I, II, IV, VI and Art. 8, Chapter III, Art. 31, and Chapter IV, Art. 34, 36 and 39, the CNEN has the task of promoting the utilization of nuclear energy, ensuring national security with regard to importation, production, utilization or export of nuclear materials, and maintaining a register of nuclear material resources, reserves or accounts in Brazil. Research, mining, industrialization and trade in radioactive sources and special nuclear material are under federal monopoly. The Presidency of the Republic is the competent authority to orient the National Nuclear Energy Policy.

3. PRESENT RADIOACTIVE MATERIAL CONTROL BY CNEN

After criticality of the first Brazilian nuclear research reactor was reached late in September 1957, the production of small quantities of radioisotopes was initiated, the first shipments being made internally in the country in 1959 [5]. With the growing demand for radioisotopes,

mainly for medical uses, the IEA (S. Paulo) had to face the problem of distribution to qualified users. The first regulation for the distribution of radioisotopes came into force, the initial step being the requirement for users to attend special courses in the methodology and application of radioisotopes in medicine, agriculture and industry.

Then, following the creation of the other Institutes and the growth of radioisotope demand in the country, additional courses in the same subjects were given in Belo Horizonte (IPR), Rio de Janeiro (IEN and CNEN) and Piracicaba (CENA).

The Department of Radioactive Material Control (DFMR) is in charge of the registration of all radioactive material imported into or produced in Brazil. All importation of any type of radioactive material, whether by official institutions or private organizations, can be done only if authorized by the CNEN through the DFMR. This therefore is at present the organ which exercises control over all types of radioactive materials in the country.

At present the uses of radioisotopes in Government institutions, other than those having liaison with CNEN, and in private organizations, are under the control of CNEN through its DFMR.

Existing regulations by which CNEN gives permits for the installation and use of teletherapy sources demand a preliminary study for the design and installation, and also a survey before operation. CNEN is now developing a system for the concession of licences for radioisotope uses in the country.

The administration of nuclear materials in the various institutions collaborating with the CNEN in the nuclear program is carried out by those responsible for the programs in which the nuclear material is used.

Concerning the uses of nuclear materials by official research institutions, control is exercised in such a way that special nuclear materials are acquired with the knowledge of DFMR, which is responsible for the import documentation, accounts and documentation pertinent to the agreements by which the materials were obtained.

All the fertile materials produced in the country have their own account at the site of preparation and in a special register at the DFMR, as is the case with thorium and uranium.

4. TRILATERAL AGREEMENT

The first Brazilian research reactors were installed as a result of a Cooperative Agreement signed between Brazil and the United States of America. This Agreement established that the installations and materials put at the disposal of the Brazilian authorities should be used exclusively for peaceful purposes and should be under USA Safeguards [6]. Furthermore, the USA initiated the process of transferring its International Safeguards to the International Atomic Energy Agency [7]. As a consequence, the Trilateral Agreement between the United States of America, the Federal Republic of Brazil, and the International Atomic Energy Agency was signed on 10 March 1967 in Vienna, thus placing safeguards under the responsibility of the IAEA [8]. The first inspection by the Agency occurred on 6 October 1969.

TABLE I. SUMMARY OF THE PRINCIPAL BRAZILIAN FACILITIES UNDER AGENCY SAFEGUARDS

Facility type	Name	Classification and location	Designer and builder	Owner and operator
Swimming-pool Reactor	IEA-RI	Principal Nuclear Facility (PNF): São Paulo (S.P.)	Babcock and Wilcox Co., Instituto de Energia Atômica - Martins Engel Co.	Instituto de Energia Atômica (IEA)
Triga Reactor	Triga-MK-1	PNF: Belo Horizonte	General Atomic Div. of General Dynamics Corp.	Instituto de Pesquisas Radioativas (IPR)
Argonauta Reactor	IEN-R-1	PNF: Rio de Janeiro	Argonne National Laboratory, Mecânica CBV [b]	Comissão Nacional de Energia Nuclear, Instituto de Engenharia Nuclear (IEN)
Subcritical assembly [a]		Other location: São José dos Campos, S.P.	Nuclear Chicago	Instituto Tecnológico de Aeronáutica (ITA)
Subcritical assembly D_2O	Capitú	Other location: Belo Horizonte	Instituto de Pesquisas Radioativas and a complex of Brazilian Industries	Instituto de Pesquisas Radioativas (IPR)
Fuel fabrication plant [c]	Divisão de Metalurgia Nuclear (DMN)	Other location: São Paulo	Instituto de Energia Atômica	Instituto de Energia Atômica (IEA)

[a] Donated by USAEC.
[b] Brazilian Company.
[c] Safeguards applied when safeguarded material is present.

4.1. Safeguards inspection system by IAEA in Brazil

The Trilateral Agreement is based on the Agency's Safeguards System which sets out in detail the procedure for applying safeguards to a facility. Specific references are also made in the Agreement to the Agency's Inspectors Document, which designates inspectors and defines their powers and duties in general terms. Subsidiary arrangements [9] were entered into to define the appointments of and facilities for Agency inspections. Table I gives a summary of the principal Brazilian facilities under Agency Safeguards.

4.2. Some aspects of the facilities under safeguards in Brazil

(a) The IEA-R 1 is a 5-MW research swimming-pool reactor, and is the most important facility from the point of view of safeguards. This reactor [10] is installed at the Instituto de Energia Atômica (IEA), at the University Campus, São Paulo. It is a swimming-pool reactor designed and built by Babcock & Wilcox Co., according to specifications furnished by the Brazilian Atomic Energy Commission. The reactor went critical for the first time on 16 September 1957, and has operated until now without major problems [11].

The reactor operations schedule has been programmed for radioisotope production, training, neutron beam experiments, activation analyses and industrial applications. The reactor operating schedule is 8 hours per operation, four times a week; this has permitted the production of the necessary amounts of the main radioisotopes [5, 12] required for medical and agricultural applications in the country. Radioisotopes with long-lived fission products are now under production [5].

An up-grading of the reactor power to 10 MW is currently under way and is scheduled for the future.

(b) The Triga-MK-1 (originally 30 kW, now 140 kW) is a research reactor used for the production of radioisotopes, educational purposes, training, industrial applications, activation analyses, and radiochemistry. The reactor is located at the Radioactive Research Institute (IPR) at the University Campus, Belo Horizonte, Minas Gerais. Criticality took place in December, 1960. An up-grading of the reactor power to 250 kW is currently under way and is scheduled for 1972.

(c) The "Argonauta" is a research reactor of near zero power designed by the Argonne National Laboratory and constructed entirely in Brazil [13]. It is used in reactor physics, neutron physics, engineering tests, laboratory-scale radioisotope production, and for educational purposes. It is located at the Instituto de Energia Nuclear (IEN), at the University Campus, Rio de Janeiro. Criticality occurred in February, 1965.

(d) Nuclear Metallurgy Division, Instituto de Energia Atômica, São Paulo. This research and development facility has manufactured the fuel elements for the subcritical assembly "Resuco", the core of which has 2378 kg of UO_2 pellets [14]. The first core for the Argonauta Reactor at Rio de Janeiro was also manufactured by the MND, which will also manufacture the enlarged core for this reactor. Its facilities include small-scale production devices for the preparation of fissile material (UO_2 pellets) and the production of nuclear-grade metallic

uranium. Bulk material is stored in sealed containers in the locked vault in the IEA Reactor Building. In addition, the facility makes many types of plate fuel elements, including extra-thin-type element plates.

5. FLOW OF NUCLEAR MATERIAL IN SÃO PAULO

Because of its importance the particular case of the flow of nuclear material in São Paulo is described.

The Administração da Produção da Monazita (APM) is the CNEN organ responsible for the industrial processing of monazite sand, from which comes crude thorium oxide, a chemically pure thorium nitrate, rare-earth chlorides, sodium phosphate and sodium diuranate (SDU, yellow cake) as a by-product. Also handled at the APM installation is the production of lithium compounds such as carbonate and hydroxide. The thorium and uranium compounds produced by APM belong to the CNEN and are under the control of the DFMR. Various other products are sold to national industry or are exported (rare-earth chlorides). The APM also undertakes research and development work, including the preparation and purification of uranium, thorium and zirconium compounds.

The sodium diuranate (SDU) produced by APM is sent to IEA at the request of its Chemical Engineering Division (DEQ). This Division is in charge of the development of chemical processes for purification and transformation of nuclear materials. It has in operation two pilot plants for the preparation of nuclear-grade ammonium diuranate (ADU), one based on ion-exchange techniques [15] and the second on solvent extraction. Its facilities include chemical and instrumental analyses such as support and quality control of purified materials, and atomic absorption and spectrofluorimetric analyses by emission spectrography. Nuclear-grade ammonium diuranate and thorium oxalate are the main products. At this Division the SDU is transformed into nuclear grade ammonium diuranate (ADU), either by the ion-exchange process [15] or by solvent extraction purification. The ADU is sent to the Nuclear Metallurgy Division where it is transformed into UO_2 pellets [16].

6. SYSTEM FOR THE CONTROL OF NUCLEAR MATERIALS IN BRAZIL

The establishment of a control system for nuclear material by the CNEN is mainly to ensure economy, safety and health. Although there is a fundamental difference between this aim and that of safeguards, nevertheless the techniques applied can be the same.

Thus the steps for the improvement of a system for the control of nuclear materials in Brazil will be: (1) the issuing by CNEN of control regulations to apply to nuclear materials and equipment subject to its jurisdiction; and (2) the training of people to fulfil this task.

The system to be applied must be such that the CNEN can obtain at any time information on the quantity and location of nuclear material.

This can be achieved through the establishment of a data reporting and inspection system.

Finally, the control regulations to be established shall be subject to periodic reviews with regard to the experience acquired.

7. NECESSARY TRAINING

The activities of persons responsible for safeguards are complex and the team which carries out this function should be very well trained. While safeguards embraces all the activities required for the management of nuclear material, it also includes new equipment and techniques, the study of several subjects and the complete study of the nuclear fuel cycle [17].

It is advisable to expand the CNEN training program to provide a group able to improve nuclear materials management. This group could be trained with the help of foreign experts, which could instruct those people managing nuclear materials, as well as those in correlated research.

ACKNOWLEDGEMENTS

The authors would like to thank the Scientific Representative of the United States Atomic Energy Commission in Brazil, Mr. R. H. Wilcox, for his valuable contribution in the preparation of this paper. We are grateful to the Head of the office of the President of CNEN, Miss N. G. M. de Faria, for the information given on the administrative aspects of the CNEN, and to Mr. H. R. Franzen, Chief of the Program Sector of ASPED for the explanation about the PNEM.

REFERENCES

[1] "Implantação da Primeira Central Nuclear", Relatório Anual da Comissão Nacional de Energia Nuclear (1969) 42.

[2] LANE, J. et al., "Study of nuclear power for South Central Brazil", International Atomic Energy Agency, Vienna, TA Rep. 412, (15 November 1968) 1A.

[3] LYRA, F., SIMON, D., FERREIRA, Jr., H., "The Angra Nuclear power plant (Brazil)", these Proceedings, Paper 801, Vol. 6.

[4] "Lei e Regulamento da Comissão Nacional de Energia Nuclear", Brazilian Law No. 4118, 27 August 1962.

[5] LIMA, W.F., "Isotope production and activation analysis using research reactors", Paper presented at a Study Group Meeting on Research Reactor Utilization, Bogotá, Colombia, December, 1967.

[6] SMITH, H.D., "Background of International Safeguards" Proc. Symp. on Safeguards R & D, Argonne (26 June 1967) 14.

[7] SEABORG, G.T., "New Problems in Safeguarding the Atom" Proc. Symp. on Safeguards R & D, Argonne (26 June 1967).

[8] INTERNATIONAL ATOMIC ENERGY AGENCY, Vienna, INFCIRC/110 (9 April 1968).

[9] "Safeguards Transfer Agreement Between the International Atomic Energy Agency, the Government of the United States of America and the Government of the United States of Brazil – Subsidiary Arrangements" (1969).

[10] SANTOS, M.D S., SARAIVA de TOLEDO, P., "The Brazilian Research Reactor", Publicação IEA No. 11 (1958).

[11] PENTEADO FILHO, A.C., SANTOS, M.D.S., "Operational experience and utilization of the Brazilian 5 MWth swimming pool reactor", Publicação IEA No. 44, September 1961.

[12] da SILVA, CONSTANCIA, P.G., "Produção de Radioisótopos na Divisão de Radioquímica do IEA", Publicação IEA No. 159 (1968).

[13] SANTOS, J., COUTINHO, D., "The Brazilian Argonauta reactor construction", Utilization of Research Reactors (Proc. Study Group Meeting, São Paulo, 1963), IAEA, Vienna STI-DOC-57, 1 (1965) 179.

[14] SANTOS, T.D.S , HAYDT, H.M., FREITAS, C.T., "Fabricaçaõ de Elementos Combustíveis de UO_2 para o Conjunto Subcrítico Re-Suco", Publicação IEA No. 92, March 1965.

[15] ABRÃO, A., FRANÇA Jr., J.M., "Usina Pilôto de Purificação de Urânio por Troca Iônica em Funcionamento no Instituto de Energia Atômica", Publicação IEA No. 219, August, 1970.

[16] SANTOS, T.D.S., BIDWELL, R.M., FREITAS, C.T., HAYDT, H.M., da SILVA, P.S.C.P., "Estudo Experimental das Principais Variáveis para a Produção de Pastilhas de UO_2 para o Reactor Sub-Crítico Resuco", Publicação IEA No. 82, December 1964.

[17] USAEC Rep. WASH-1122, "Research and Development for Safeguards" (1 December 1968) 1.

DISCUSSION ON AGENDA ITEM 5.1

Safeguards systems analysis and safeguards objectives

DISCUSSION ON THE FOLLOWING GROUP OF PAPERS:

P/771	*FRG*	*Presented by W. Häfele*
P/098	*USA*	*Presented by H.J.C. Kouts*
P/828	*Japan*	*Presented by R. Imai*
P/558	*CSSR*	*Presented by O.Žoch*

M. SANDOVAL VALLARTA: It would seem appropriate to inform this meeting that the Treaty for the Prohibition of Nuclear Weapons in Latin America has already been signed by most of the Latin American countries and is now in effect. It has now been recognized by the United States of America and the United Kingdom. This Treaty includes the obligation on the part of the signatories to sign a safeguards agreement with the Agency within a period specified. Thus, everything that has been put forward here today is equally applicable to safeguards under the Latin American Treaty.

F. MORGAN: I would like to ask the following question in connection with paper P/098 by Kouts, Bennett and Lawroski.

The performance of nuclear plant may introduce swings in material balance which will worry any inspector, due to plant factors which may be incompletely understood, or to the necessity of making estimates of in-process inventory. With very large plants, such swings may easily exceed the small limits implied by the definition of "strategic quantity" employed in the paper. While I can readily agree to the value of a "strategic quantity" in the analysis of a system, from the point of view of planning, I think it may be academic in defining formal procedures. One has to live with the realities of factory life. If the limits are exceeded regularly, the system will fall into disrepute. Equally, if a "normal operating loss" were accepted in advance, this would facilitate diversion of material. Rather than to prejudge the issue, which is what use of the terms "strategic quantity" and "normal operating loss" implies, it seems more appropriate to define an "action level" or levels from the observed performance of the plant, which would automatically trigger enquiry and remedial action. Would the authors care to comment?

H.J.C. KOUTS: The analysis that I presented showed that currently used measurement methods do not in all cases permit us to meet the criteria shown. In some cases they do. Measurement methods that could be used would permit meeting criteria in all cases. However, there is no real plan at this time to apply these numerical criteria. If the criteria do come to be required in this form or a similar one, they will be used for safeguards administered by facilities rather than by national or international safeguards groups. It is true that operational losses, when they occur, will cause difficulties in that control limits will be exceeded. However, operational losses are indistinguishable from diversions except possibly after an

investigation. If accounting is to have value for safeguards, it must be capable of revealing the existence of losses of an important size to permit investigating to see if they were operational losses or diversions.

W. HÄFELE: I observed in Dr. Kouts' paper a greater emphasis on "containment" than was earlier the case. Does he feel that accounting procedures within the containment can be eased if the containment is enhanced?

H.J.C. KOUTS: Yes, I do, especially as regards critical time. We have taken the view that where containment is very good, the time between closing material balances can be extended. Our usual assumption is an extension of the balance period by a factor of three when containment is especially good. One example is a chemical reprocessing plant, where containment is believed useful to trade off against critical time.

A. LARSSON: How is the critical-time figure (Table I) of one month for low-enriched uranium arrived at?

H.J.C. KOUTS: It was simply an assumption made for purposes of the analysis that was described. No particular importance should be attached to any of the critical-time or strategic quantities listed in the paper.

M.N. RYZHOV: Is it possible, within the framework of a mathematical model, to express the level of uncertainty in determining quantities of nuclear materials? This uncertainty depends on a large number of factors, e.g. type of process, level of technology, system of allowing for staff qualification and so forth. Does not an error in determining the level of uncertainty lead to a situation where, in some cases, this value will be an excessive burden for some installations, while in other cases there will be a possibility of switching materials?

W. HÄFELE: Whenever we are dealing with digital accountability, which is the case in all heterogeneous reactors, no such uncertainty exists. As far as open accountability is concerned, one has to consider the specific situation. Generally speaking, the uncertainty is of the order of 1%. A number of criteria governing the frequency of inspections are to be found in the IAEA document INFCIRC/153 ("The Structure and Content of Agreements between the Agency and States required in Connection with the Treaty on the Non-Proliferation of Nuclear Weapons"), which presents the findings of the Agency's Safeguards Committee. Another important source of information is integral experiments, and in this connection I would draw particular attention to the recently published report on the MOL-III experiment, carried out by the Karlsruhe group with extensive international support. The principal finding is that, contrary to earlier assumptions, it is more or less only the systematic error – not the random error – that gives rise to a range of uncertainty. The Safeguards Committee and the working groups of the IAEA have proposed the establishment of a permanent group to make a continuous review of the latest experience in connection with material unaccounted for (MUF), and to assess the acceptable level of uncertainty due to the systematic error, with a view to incorporating such data in, say, the Subsidiary Arrangements wherever appropriate.

K. OSHIMA: In Fig. 2 of paper P/771, it is indicated that the introduction of the "game theory" concept into the system would reduce the safeguards efforts or costs to a large extent. Could Mr. Häfele explain in what way this approach can be implemented in practical procedures?

W. HÄFELE: What is still lacking is a more general understanding and, even more so, practical experience of applying game theory. The models

developed thus far merely reveal the weak points of the envisaged system and identify sensitive input parameters. Moreover, numbers must come into the picture when the pay-off function has to be defined. There are no distinct differences between applying a game theory model, a statistical model or a deterministic model. Generally speaking, there is a need for more scientific groups to explore this game theory aspect.

M. DAYAL: I would like a clarification from Mr. Häfele about the determination of a figure for the "confidence level". Actual confidence could vary from inspector to inspector. Secondly, in practice it is often said that it has to be assumed that, without inspection, the operator will try to divert, and this probably leads to the imposition of more stringent safeguards than would otherwise be necessary. Is this assumption used in the models for arriving at the confidence level figure and related parameters, or have you tried to factor in any assumptions regarding variations in the level of effort that could be put in to divert?

W. HÄFELE: The term "confidence level" has been used in the statistical sense, as a mathematical term. It does not refer to the subjective feeling of an individual inspector as a human being. In order to have statistics, numbers have to be produced and this is the reason for stressing accountancy. Formalization of procedures is another means of helping to bridge the gap between subjective action and objective reality. So defined, the confidence level of 95% used in Fig. 2 is common today.

F. MORGAN: In the opening paragraph of his paper, Mr. Häfele writes: "The application of decision theory allows .... to some extent the non-deterministic description of conflicts between human partners". But this calls for many types of value judgement - for example, the proper equation of gains and losses. How does one equate a kilogram of diverted plutonium with a fine of x pounds sterling or with y years in jail? Equally important is the question of what probability of detection is required over what period of time. Such value judgements, arrived at by a political process, suggest that "objectivity" must in fact depend on agreed and negotiated conventions.

D. GUPTA: I would like to make a few comments on the remarks of Mr. Morgan. It is true that in game theory an effort has to be made to compare seemingly non-comparable quantities. But human beings are compelled to make such comparisons in many phases of their decision-making activities. For example, in the field of justice, a judge has to decide on the number of days of jail to be imposed on a person who has stolen some apples. He is comparing the value of the stolen apples with the freedom lost in jail. Similarly, commercial aviation organizations have to weigh and compare the value of human life with the loss in their profit if the scheduled departure of a plane has to be postponed because of bad weather. In all these cases formalized comparison appears to be feasible. Therefore, it is justifiable to investigate possibilities of such comparison in the matter of safeguards also.

W. HÄFELE: Indeed, value judgement is required. This is different from the case of the traditional sciences, where we can only compare lengths with lengths, masses with masses etc. We are referring to the interface between man and nature that is included in systems analysis but is excluded from traditional science.

R. IMAI: If I may, I would also like to say a word about game theory. It is useful for the formulation of the problem but probably not for producing

procedural solutions. The phase in which game theory can be applied will be fairly limited.

D. GUPTA: In connection with paper P/828, I would like to ask Mr. Imai how many times a year material balances are carried out under the Japanese national safeguards system.

R. IMAI: Under existing Japanese law, this is done twice a year. We are also reviewing this part of the law, in an effort to bring inventory-taking requirements into line with the current system.

DISCUSSION ON THE FOLLOWING GROUP OF PAPERS:

P/725	*Euratom*	*Presented by E. Jacchia and S. Finzi*
P/770	*IAEA*	*Presented by R. Rometsch*
P/204	*Brazil*	

K. OSHIMA: Could Mr. Rometsch tell us what type of information the Agency wants to be included in the Subsidiary Arrangements?

R. ROMETSCH: In the Agency's safeguards practice so far, the Subsidiary Arrangements have to some extent been in the nature of a supplement to the Agreement, specifying those parts of the procedures which were defined only in general terms by the Agreement and the references to the safeguards systems. The new type of NPT Agreement worked out by the Safeguards Committee and laid down in Agency document INFCIRC/153 is much more complete and contains many of the details which previously were set out in Subsidiary Arrangements (e.g. operational procedures). However, there are still a great number of data needed for safeguards implementation, e.g. quantitative data on facilities, which of course cannot be included in the Agreement. The purpose of the new form of Subsidiary Arrangement is merely to ensure this type of quantification. There are some 20 articles in the Agreement which end with the sentence "as specified in the Subsidiary Arrangement". All these technical data will be laid down in the form of tables, models and forms which, together, represent the Subsidiary Arrangements; no extended or supplementary obligations and rights can be derived from them.

A. PETIT: I should like to ask Mr. Jacchia, on the subject of his reference to the challenge of a "nuclear black market" in connection with the measures to be taken by States in the full exercise of their sovereignty, whether he does not think that care should be taken not to confuse these policing problems – which consist in preventing and suppressing criminal activities of private individuals and subversive organizations – with the problems of international controls which are designed to verify whether States are respecting obligations which they themselves have assumed.

E. JACCHIA: I fully agree with Mr. Petit concerning the need to distinguish clearly between activities in these two spheres. What I would like to emphasize is the desirability of cooperation, which would be achieved essentially through an exchange of information between State bodies responsible for national security and the State department responsible for

administering the national safeguards system. It goes without saying, as I pointed out in my oral presentation, that the international authority would have no right of supervision in this purely internal cooperation of the State.

M.N. RYZHOV: What types of portable apparatus are used by Euratom inspectors in carrying out inspections?

S. FINZI: As regards the instrumentation used by the Safeguards Department for qualitative and quantitative non-destructive assay, we have to distinguish between portable and transportable equipment. The former category includes apparatus which can be transported by hand, as normal luggage. At present the Department is using portable gamma-spectrometers with NaI detectors and several gamma-monitors.

The latter category includes equipment which can be transported by car or small truck. Transportable instruments utilized by the Department include the neutron coincidence counter and the gamma-spectrometer, consisting of Ge(Li) detector connected with a multichannel analyser. All these instruments are used during routine inspection operations.

M.N. RYZHOV: What percentage of the overall inspection effort consists in non-destructive testing and what percentage in destructive testing?

S. FINZI: I would say that about 50% of the effort of the Research and Development program mentioned in our paper is devoted towards non-destructive techniques and 10% to the direct methods. As far as the inspection effort is concerned, the distribution is about half and half.

M.N. RYZHOV: Do the duties of Euratom inspectors consist only in carrying out qualitative measurements, or do they perform quantitative measurements connected, for example, with the drawing up of material balances of nuclear materials?

S. FINZI: Both qualitative and quantitative measurements are performed.

C. HOSMER: Assume that the safeguards system fails to deter a diversion but succeeds in making a timely discovery of its occurrence, then the problem of getting the material back from illicit hands arises. What is being done by the IAEA or by other authorities to prepare a national police system to act effectively to recover fissionable materials?

R. ROMETSCH: The setting-up of a police system, which is of course a necessity for such operations as getting back diverted material, is not an obligation of the IAEA.

However, the Agency does have to establish the necessary links and also to be prepared to transmit expert advice on these matters, if so requested by States which have no experience with special-task police systems.

Regarding the information connection between the IAEA and a State, the contact point is foreseen in the Safeguards Agreement: it is the "national system of accountancy and control of nuclear material". This term by itself shows the double function of such a system. Normally only the accountancy part is used in connection with the Agency's work. From it information is fed to the Agency. However, there is also a feedback. This mechanism is absolutely necessary, for instance, in reaching agreement on those special circumstances which require special inspections. This close contact between the Agency's inspectorate and the national system would be used to trigger the other function of the national system – which is none of the Agency's business – namely the control function.

The latter, I imagine, would in one way or the other, depending on national practices, be combined with or related to a police system. In this way, I believe, the arrangements laid down in the Safeguards Agreement establish the contacts necessary for the regular provision of information to the national police system, which alone has the power to take action within a State. The triggering mechanism lies within the "national system of accountancy and control of nuclear material", which is the point of contact between a State and the IAEA.

R. IMAI: Mr. Hosmer's question is a part of the much wider issue of what do you do when a possible diversion has been reported by the safeguarding authority. A statement to this effect would be primarily of a probabilistic nature, rather than being based on someone having been caught red-handed. One would have to discover accidentally an act of diversion or the making of a bomb. What action would be taken on the basis of such a statement is not, in my view, a technical problem.

Japan has not really given any thought to means of returning diverted material. The safeguards system is a method of providing assurance to the international community and it is not conceived of as a police action.

There is always a danger in trying to write detailed specifications in advance for all conceivable cases. While this may be an interesting exercise in logic, it merely makes safeguards complicated, burdensome and unworkable. What is required at this stage is more serious efforts to reconcile the theoretical model of safeguards we have constructed with the reality of fuel-cycle operations.

E. JACCHIA: In reply to the question as to what the international authority would do in the specific case of a violation of the rules, I would point out that Article 83 of the Euratom Treaty lays down sanctions to cover this case. In increasing order of severity, the sanctions provide for the issuance of warnings, withdrawal of special advantages, placing the enterprise under controlled administration, and partial or total withdrawal of source material or special fissionable material.

AGENDA ITEM 5.2

Safeguards techniques and instrumentation

Techniques et appareils pour les garanties

Методы применения гарантий и контрольно-измерительные приборы

Técnicas e instrumentos de salvaguardia

Chairman

K. OSHIMA, Japan

Vice-Chairman

Z. GYIMESI, Hungary

Scientific Secretaries

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DEVELOPMENT OF TECHNIQUES AND INSTRUMENTATION FOR NON-DESTRUCTIVE ASSAY OF FISSIONABLE MATERIALS *

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Abstract–Résumé–Аннотация–Resumen

DEVELOPMENT OF TECHNIQUES AND INSTRUMENTATION FOR NON-DESTRUCTIVE ASSAY OF FISSIONABLE MATERIALS.

A full range of non-destructive assay techniques and instrumentation is being developed in the US safeguards research and development program for application to practical assay problems in the areas of safeguards, nuclear-materials management, criticality safety, and quality and process control. Non-destructive assay methods can be categorized as (1) active and (2) passive. Active assay involves irradiation with neutrons or photons to induce fissions in the sample and interpretation of the resulting neutron or gamma 'signatures' to determine quantitatively the amount of fissionable material present. Passive assay uses naturally occurring neutron and gamma radiations as direct 'signatures' of fissionable material. Innovations in instrumentation and analysis techniques have extended passive methods to categories of materials not previously amenable to quantitative assay by passive methods. However, for other important assay problems, the paucity of suitable passive signatures, together with their inherent physical limitations (e.g. low penetrability of gammas in dense media and low signal-to-noise levels in highly radioactive samples), can severely limit the effectiveness of passive methods. For some problems active assay methods provide the only means for practical non-destructive assay of fissionable material. For assay problems of the future (e.g. plutonium recycle fuel), a combination of active and passive methods will be required for optimum assay capability. Radioactive and isotopic sources, e.g. ^{252}Cf, provide an attractive alternative to the more conventional accelerator sources for neutron interrogation and assay. In-plant and field applications of two different types of radioactive-source assay systems are reviewed.

Non-destructive assay results are reported, and in several cases compared with chemical assay data, in the categories of reactor fuel materials - feed and product, inventory verification samples, and fissionable material scrap and waste. Assay accuracies for reactor fuels and inventory samples range from less than 1% to a few per cent. For fissionable scrap and waste, where sampling and chemical analysis may be difficult or essentially impossible, non-destructive assay accuracies can range from a few per cent up to approximately 25%, depending on the composition and geometric complexity of the materials. During the past year non-destructive assay methods have been applied to a variety of materials in commercial nuclear plants under a Plant Instrumentation Program jointly sponsored by the United States Atomic Energy Commission and several US commercial nuclear companies. The salient results of this program are reviewed.

The USAEC mobile non-destructive assay laboratories, MONAL and GAMAS, respectively, use neutron and photon interrogation sources as well as complementary passive assay equipment. These mobile laboratories provide a unique field capability for evaluating the relative effectiveness of different types of non-destructive assay instrumentation in the actual industrial environment. Field-assay experience is reviewed, and practical assay results are summarized.

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* Work performed under the auspices of the U.S. Atomic Energy Commission. The paper includes contributions from R.H. Augustson, W.C. Bartels, N.S. Beyer, L.E. Bruns, D.R. Cartwright, J.E. Cline, D.G. Costello, B.R. Dennis, L.V. East, A.E. Evans, J.E. Foley, R.A. Forster, W.J. Gallagher, R.O. Ginaven, T. Gozani, E.F. Kurtz, J.E. Lovett, J.I. McMillan, J.J. Malanify, H.O. Menlove, J.H. Menzel, Y.D. Naliboff, N.D. Nutter, R.E. Olson, J.L. Parker, T.D. Reilly, D.E. Rundquist, R.D. Schamberger, D.B. Smith, M.M. Thorpe, G.D. Trimble, R.B. Walton, J.D. Watkins, and C.R. Weisbin.

МИСЕ AU POINT DE METHODES ET D' APPAREILS POUR L' ESSAI NON DESTRUCTIF DE MATIERES FISSILES.

Dans le cadre du programme d' études pour les garanties, les Etats-Unis mettent au point toute une série de méthodes et d' appareils d' essais non destructifs pour résoudre des problèmes pratiques de dosage en matière de garanties, de gestion de matières nucléaires, de protection contre la criticité, et de contrôle de la qualité et des opérations. On peut distinguer les méthodes non destructives selon qu' elles sont actives ou passives. Les méthodes actives consistent à irradier l' échantillon avec des neutrons ou des photons pour y provoquer des fissions et à interpréter ensuite les «signatures» neutroniques ou gamma pour en déduire les quantités de matières fissiles présentes. Le dosage passif met à profit les neutrons et rayons gamma émis naturellement par la matière fissile considérée. Grâce au perfectionnement des appareils et des méthodes d' analyse, on a pu étendre la méthode passive à des catégories de matières qui ne s' y prêtaient pas jusqu' ici. Mais, pour d' autres problèmes importants de dosage, le manque de moyens d' identification convenables pour les méthodes passives ainsi que leurs limitations physiques inhérentes (par exemple le faible pouvoir de pénétration des rayons gamma dans des milieux denses et le faible rapport signal/bruit si les échantillons sont fortement radioactifs) peuvent nuire considérablement à l' efficacité des méthodes passives. Dans certains cas, les méthodes de dosage actives fournissent le seul moyen pratique de procéder à des analyses non destructives. Pour les problèmes de dosage de l' avenir (par exemple pour le combustible au plutonium recyclé), on devra combiner la méthode active et la méthode passive pour atteindre les possibilités maximales de dosage. Les sources radioactives et isotopiques, par exemple ^{252}Cf, peuvent remplacer avantageusement les sources plus classiques que sont les accélérateurs pour l' identification et le dosage par les neutrons. Les auteurs passent en revue les applications de deux méthodes différentes de dosage avec sources radioactives, en laboratoire et en campage.

Ils donnent les résultats de dosages non destructifs et les comparent dans certains cas avec les données obtenues par des moyens purement chimiques, sous les trois rubriques suivantes: matières contenues dans le combustible de réacteur à l' entrée et à la sortie, échantillons prélevés pour vérifier les stocks, et déchets de matières fissiles. L' exactitude des dosages pour le combustible de réacteur et les échantillons servant au calcul du stock varient de 1 à plusieurs points pour cent. Pour les déchets de matières fissiles, lorsque l' échantillonnage et l' analyse chimique sont parfois difficiles ou totalement impossibles, le degré d' exactitude des essais non destructifs peut varier de quelques points pour cent à environ 25%, selon la composition et la complexité géométrique des matières. La méthode des essais non destructifs a déjà été appliquée à toute une gamme de matières dans des centrales nucléaires industrielles dans le cadre d' un programme exécuté conjointement par la Commission de l' énergie atomique des Etats-Unis et plusieurs sociétés nucléaires industrielles. Les auteurs analysent les principaux résultats.

Les laboratoires mobiles de dosage non destructif de l' USAEC (MONAL et GAMAS) utilisent des sources de neutrons et de photons pour la méthode active et du matériel de dosage complémentaire pour la méthode passive. Ces laboratoires mobiles fournissent un moyen unique d' évaluation de l' efficacité relative des différents types d' appareils d' analyse non destructive dans le milieu industriel réel. Les auteurs passent en revue l' expérience acquise dans ce domaine et résument les principaux résultats obtenus.

РАЗРАБОТКА МЕТОДОВ И ПРИБОРОВ ДЛЯ АНАЛИЗА РАСЩЕПЛЯЮЩИХСЯ МАТЕРИАЛОВ БЕЗ РАЗРУШЕНИЯ ОБРАЗЦА.

Американская программа научных исследований и разработок в области гарантий предусматривает разработку целого ряда методов и контрольно-измерительных приборов для решения практических проблем испытания без разрушения образца при осуществлении гарантий, а также для обращения с ядерными материалами, обеспечения безопасности при достижении стадии критичности, контроля за качеством и технологическим процессом. Методы испытания без разрушения образца могут быть разделены на активные (1) и пассивные (2). Активный анализ включает облучение делящихся материалов нейтронами или фотонами, чтобы вызвать деление в образце и интерпретацию результатов такого облучения нейтронами или гамма-излучением для определения количества имеющегося делящегося материала. При пассивном анализе обычно анализируются нейтронные и гамма-излучения, генерируемые делящимся материалом без постороннего воздействия. Нововведения в контрольно-измерительных приборах и методах анализа позволили распространить применение пассивных методов на материалы, которые ранее не поддавались количественному анализу при помощи пассивных методов. Однако в некоторых других важных случаях эффективность пассивных методов может быть значительно ограничена слабостью исследуемых пассивными методами излучений, а также рядом соответствующих физических свойств (например, слабой проницаемостью гамма-излучения в плотных средах и низким уровнем измеряемого сигнала по сравнению с фоном высокорадиоактивных образцов. В некоторых случаях активные методы являются единственным методом практического проведения анализа без разрушения образца делящихся материалов. Для решения проблем анализа в будущем (например, повторное использование плутония) потребуется сочетание активных и пассивных методов в целях оптимального использования возможностей анализа. Радиоактивные и изо-

топные источники, например ^{252}Cf, имеют преимущества по сравнению с обычными источниками излучения для нейтронного анализа. Рассматривается применение в заводских и полевых условиях двух различных типов систем анализа с радиоактивными источниками.

Излагаются результаты анализа без разрушения образца, и в некоторых случаях результаты сравниваются с данными химического анализа. Это сравнение дается по категориям реакторных топливных материалов – загрузка и продукт, образцы для контроля за загрузкой топлива, скрап делящегося материала и отходы. Погрешности анализа реакторного топлива и образцов свежего топлива составляют от менее одного до нескольких процентов. Что касается скрапа делящегося материала и отходов, в которых взятие проб и химический анализ, возможно, затруднительны или,по существу,невозможны, точность анализа без разрушения образца колеблется в диапазоне от нескольких процентов до примерно 25%, в зависимости от структуры и геометрической формы материала. За последний год методы анализа без разрушения образца применялись к разнообразным материалам на коммерческих ядерных установках по программе разработки контрольно-измерительных приборов для атомных установок, осуществляемой Комиссией по атомной энергии США совместно с рядом коммерческих ядерных предприятий США. Рассматриваются положительные результаты выполнения этой программы.

Передвижные лаборатории КАЭ США по проведению анализа без разрушения образца, MONAL и GAMAS, соответственно, используют нейтронные и фотонные источники, а также дополнительное оборудование для проведения пассивного анализа. Такие передвижные лаборатории предоставляют уникальную возможность для оценки в полевых условиях относительной эффективности различных типов контрольно-измерительных приборов, используемых для проведения испытаний без разрушения образца и в реальных промышленных условиях. Рассматривается опыт проведения анализа на местах и кратко суммируются практические результаты анализа.

DESARROLLO DE TECNICAS E INSTRUMENTACION PARA EL ANALISIS NO DESTRUCTIVO DE MATERIALES FISIONABLES.

Dentro del programa de investigación y desarrollo de salvaguardias que ejecutan los Estados Unidos, se está perfeccionando toda una gama de técnicas de análisis no destructivo y de instrumentación con el mismo fin, para su aplicación a la resolución de diversos problemas de índole práctica que respecto de ese tipo de análisis se plantean en los sectores de las salvaguardias, gestión de los materiales nucleares, seguridad frente a la criticidad, y control de calidad y de los procesos industriales. Los métodos de análisis no destructivo se pueden clasificar en dos grupos: 1) de carácter activo y 2) de carácter pasivo. Los análisis de carácter activo entrañan el empleo de la irradiación con neutrones o fotones para provocar fisiones en la muestra, así como la interpretación de las «señales» neutrónicas o gamma resultantes, para lograr una determinación cuantitativa del material fisionable presente. En los análisis de carácter pasivo se emplean como «señales» directas del material fisionable los neutrones y la radiación gamma naturalmente presentes. Las innovaciones introducidas en la instrumentación y en las técnicas de análisis han extendido el empleo de los métodos de carácter pasivo a tipos de materiales que en el pasado no se prestaban al análisis cuantitativo por dichos métodos. No obstante, en el caso de otros importantes problemas de análisis, la escasez de «señales» adecuadas obtenidas por métodos de carácter pasivo, junto con las limitaciones físicas inherentes a esos problemas (por ejemplo, la penetrabilidad baja de las radiaciones gamma en medios densos, y los niveles bajos de la razón señal-ruido en muestras de elevada actividad), pueden limitar considerablemente la eficacia de esos métodos de carácter pasivo. Frente a determinados problemas, los métodos de análisis de carácter activo constituyen el único medio práctico de análisis no destructivo de los materiales fisionables. En cuanto a los futuros problemas en materia de análisis (por ejemplo, los planteados por el reciclado del plutonio), se precisará una combinación de ambos tipos de métodos, activos y pasivos, para lograr una capacidad de análisis óptima. Las fuentes radiactivas e isotópicas, por ejemplo, el ^{252}Cf, constituyen una nueva y sugestiva posibilidad frente a las fuentes más corrientemente empleadas (aceleradores), para la indagación y el análisis mediante neutrones. Los autores examinan la aplicación, en instalaciones y sobre el terreno, de dos tipos distintos de sistemas de análisis mediante fuentes radiactivas.

En la memoria se indican los resultados del análisis no destructivo, y se les compara en varios casos con los datos obtenidos en el análisis químico, en las diversas categorías de materiales empleados en la alimentación de reactores: combustible que entra en el reactor y productos que salen de éste, muestras para la comprobación de las existencias en inventario, y residuos y desechos que contienen materiales fisionables. La precisión de los análisis en el caso de los combustibles de los reactores y en el de las muestras de las existencias inventariadas oscila entre menos del 1% a unas pocas unidades por ciento. En el caso de los residuos y desechos que contienen materiales fisionables, en los que resulte difícil o prácticamente imposible recurrir al muestreo y el análisis químico, la precisión de los ensayos no destructivos pueden oscilar entre unas pocas unidades por ciento hasta un 25% aproximadamente, según la composición y la complejidad estructural de los materiales. En el curso del

pasado año se han aplicado métodos de análisis no destructivo a una amplia diversidad de materiales en centrales nucleares comerciales en virtud de un Programa de Instrumentación de Centrales que copatrocinan la Comisión de Energía Atómica de los Estados Unidos (USAEC) y varias empresas nucleares comerciales del país. Se examinan los resultados más destacados de este programa.

Dos laboratorios móviles de la USAEC para el ensayo no destructivo de materiales nucleares, el MONAL y el GAMAS, emplean respectivamente fuentes neutrónicas y fotónicas para el análisis de carácter activo, así como equipo complementario de análisis pasivo. Estos laboratorios móviles ofrecen posibilidades únicas de trabajo sobre el terreno para evaluar y comparar la eficacia de los distintos tipos de instrumentos de análisis no destructivo que se emplean realmente en la esfera industrial. Se expone la experiencia adquirida con estos análisis efectuados sobre el terreno y se resumen los resultados prácticos de esos análisis.

INTRODUCTION

The nuclear safeguards research and development program in the United States is developing and testing a full range of nondestructive assay techniques and instrumentation for application to the broad spectrum of fissionable materials found in the nuclear fuel cycle. Nondestructive assay instrumentation is being deployed at key commercial facilities through plant instrumentation programs and mobile assay laboratories in order to demonstrate and test the new techniques on practical assay problems in the nuclear industry. These field activities also effect vital liaison between industry and scientists developing the new technology and instrumentation of nondestructive assay. Such close coupling is of great value in keeping technique and instrumentation development "problem oriented" and highly responsive to the real assay needs of the nuclear industry.

PASSIVE ASSAY TECHNIQUES AND APPLICATIONS

Gamma Ray Assay

Innovations and improvements in passive gamma ray and neutron assay techniques have greatly extended the range of applicability of these techniques for quantitative assay. The major limitation on passive gamma techniques is the attenuation of the gamma "signatures" and consequent variation in response depending upon fissile material distribution within a large, attenuating matrix. To alleviate this problem the rotation-collimation method [1] has been employed to effectively "flatten" the response to gammas from various interior positions of cylindrical containers. In this method the container is rotated past a horizontal collimator which selectively reduces the response of sources located near the periphery, thereby compensating for the more-highly-attenuated response from central positions of the cylinder. By varying the collimation angle it is possible to achieve flat response over a large range of attenuations, e.g. up to a maximum diametric attenuation factor of ~ 200. This method is generally applicable over a wide range of sample sizes and detector systems. It is most useful when the sample matrix is reasonably uniform (the spatial distribution of fissile material itself can be quite non-uniform), although rotation-collimation can bring significant improvements in assay accuracy even in the case of non-uniform matrices.

The rotation-collimation technique has been employed in versatile gamma ray assay systems designed for sample sizes ranging from quart bottles to 55-gallon barrels. For example one eight-channel gamma scanning system for

55-gallon barrels [2] consists of eight individual detector modules, each containing a shielded 2 in. x 2 in. NaI crystal with fixed vertical collimation and adjustable horizontal collimation. Eight small transmission sources placed opposite the gamma detectors are utilized for an external transmission scan. For maximum detail of spatial distribution, the individual detector outputs can be multiscaled separately (and simultaneously) using a small computer for data acquisition and reduction. The sensitivity of this system for Pu is ~ 1 g in a 55-gallon drum.

In general, the key to gamma ray assay is an accurate transmission measurement; given this, typical accuracies of gamma scanning systems can range from the percent level for small, well-characterized samples to 10-15% for 55-gallon barrels. Typical assay times are 10 minutes.

For quality control of "thin" finished product, e.g. fuel plates and rods, gamma-ray scanning is an established technique, particularly as applied to highly enriched U components such as MTR fuel plates [3]. With large-volume high-resolution Ge(Li) detectors this technique can be extended to fuels with more complex characteristic gamma-ray spectra. An example of this application for Pu fuel is the stringent quality control of the 12,000 ZPPR (Zero Power Plutonium Reactor) fuel plates at Argonne National Laboratory [4].

The severe attenuation of ^{235}U and ^{239}Pu gamma rays in highly attenuating samples has been used to advantage in the development of the so-called "enrichment meter" [5]. The method is based on the fact that with proper detector collimation the 185 keV gamma signal from a dense, uniform object containing uranium is proportional to the enrichment of ^{235}U. The enrichment meter can be used to measure a wide range of uranium materials including fuel pins, plates or any reasonably homogeneous sample that is fairly rich in uranium. Results obtained with hand-carried instrumentation on an assortment of fuel rods, drums of different uranium compounds, ash residues, and UF_6 cylinders indicate relative accuracies of 5% or better in counting times of a few minutes. Measurements with Ge(Li) detectors yield accuracies approaching 1%. The enrichment-meter concept may also be used for quality control of different reactor fuel blends, e.g. PuO_2, UO_2, U-C, and Th-C. Enrichment-meter measurements may be combined with a ^{238}U assay using the more penetrating 1001 keV gamma ray or the spontaneous fission neutron activity, thus providing a total uranium assay.

TABLE I. COMPARISON OF "ENRICHMENT METER" AND MASS SPECTROMETER DATA ON K-25 ENRICHED-U RESIDUE

Labeled Mass Spec. Enrichment (Can #11547)	Enrichment Meter Measurement (top/bottom)	Core Samples Mass Spec. Enrichment (top/bottom)
2.9308%	2.7%/15.5%	2.683%/15.23%

A noteworthy application of this method is the assay of enriched-U residues (mostly incinerator oxides) from the K-25 enrichment plant at Oak Ridge, Tenn. Enrichment levels of 1-5% were indicated on 216 sealed cans (6.0 in. O.D. x 15 in. high) containing these residues. The enrichment meter measurement revealed a vertical stratification of enrichments in several K-25 cans with small quantities of material enriched as much as 30% in ^{235}U. Of 216 cans, 30 showed significant (up to order of magnitude) discrepancies from the enrichment value labeled on the cans. This stratification in isotopic enrichment was verified quantitatively by mass spectrometry on a series of core samples; the results (top and bottom) for one of the cans are presented in Table I.

Passive Neutron Counting

Recent research in passive neutron counting has been directed toward the development of high-efficiency neutron coincidence detectors for assaying plutonium (via ^{240}Pu spontaneous fission counting) [6,7], and more recently, uranium, (via ^{238}U spontaneous fission counting) [8]. In both cases the isotopic composition must be known, or separately determined, to take account of the fissile component. Coincidence counting of ^{240}Pu spontaneous fission neutrons employs 4π-polyethylene-moderated assemblies with ^{3}He or BF_3 detectors. For assaying Pu-bearing scrap and waste containing light elements (i.e. high (α,n) activity), short die-away time detectors are required to maximize the contribution of real coincidence events relative to accidental events; this led originally to the concept of the variable die-away time counter [9].

A complete assay system [6] employing high efficiency, variable-die-away-time detectors has been developed for assay of one gallon (or smaller) containers. This system (soon to be available commercially) has been used to assay Pu samples varying in mass from a few milligrams to several hundred grams. Accuracies of ± 3% and ± 8% have been obtained for Pu in homogeneous and heterogeneous Pu matrices, respectively. Typical assay time is ~1 min for 5% counting statistics on 1 g of ^{240}Pu.

The success of the one-gallon assay system prompted the development of a similar counter for 55-gallon barrels [10]. This counter has a gross counting efficiency of 14% and a sensitivity of 10-50 mg ^{240}Pu in 55-gallon barrels [10].

In practical assay applications matrix materials can significantly alter the observed coincidence count. Studies on a wide variety of matrix materials have shown the new ^{252}Cf (or ^{240}Pu) source-addition technique [11] to be a very effective means of correcting for matrix perturbations. Another current R&D goal is optimization of detector-moderator configurations to achieve flat energy response and thus minimize matrix perturbations (i.e. keep corrections at the few percent level.)

Although the spontaneous fission rate of ^{238}U is low, practical coincidence counting has been demonstrated on ^{238}U materials containing as little as 50 g of ^{238}U (or still less if necessary). Several hundred cans of residues containing low-enriched uranium (~ 100 g to ~ 10 kg U in each can) have been assayed and the results found to be in excellent agreement with chemical sampling. Assay times of ~ 3 min gave < 5% precision for the 10 kg samples [8].

ACTIVE ASSAY METHODS AND APPLICATIONS

New and improved methods of passive assay have proved adequate for many if not most Pu assay problems. However, some classes of Pu samples are not amenable to passive assay, while for ^{235}U the only passive signature (^{185}keV gamma) is of limited usefulness in highly attenuating matrices. Also passive gamma assay is essentially inapplicable in the case of highly radioactive materials such as spent fuel elements and "hot" scrap. In these cases, active interrogation techniques provide very effective means of quantitative assay with the requisite high penetrability and high signal-to-background signatures for the major fissile species of greatest economic and strategic importance (notably ^{235}U and ^{239}Pu).

Active interrogation employs an external source of highly penetrating neutrons or photons to induce fissions in the material under investigation. Interrogation sources include 14-MeV neutron generators (pumped-tube or sealed-tube type), Van de Graaff accelerators, electron linear accelerators and radioactive sources such as ^{252}Cf, SbBe, and ^{238}PuLi. Quantitative assay is based on detailed observations of one or more types of emissions following fission, notably delayed and prompt neutrons and gamma rays.

NEUTRON INTERROGATION

In assay methods based on neutron interrogation, the energy of the incident neutrons can be varied to take full advantage of the isotopic discrimination afforded by the subthreshold and superthreshold fission characteristics of the various fissionable materials of practical interest. In particular, subthreshold and superthreshold neutron interrogation [12] provides a straightforward, direct separation of the response of the fissile isotopes (e.g. ^{233}U, ^{235}U, ^{239}Pu) from the fertile isotopes (e.g. ^{238}U, ^{232}Th) [12, 13].

The delayed neutron "yield" method of assay [12, 14] has been very effectively combined with the super- and subthreshold interrogation technique to carry out precise isotopic assay on various feed and product materials, including reactor fuel rods and fuel elements, with an assay accuracy of $\sim$ 1% or better over a wide range of fuel loadings and isotopic enrichments. Some recent applications of delayed neutron assay techniques are discussed below.

Delayed Neutron Assay of Highly Enriched UAl_x

Receiver inventory measurements were made on 320 kg of highly-enriched uranium (93% ^{235}U) contained in 90 cans of uranium aluminide powder --a homogeneous powdered alloy of 69 ± 3 wt% uranium, the balance being aluminum. Since the UAl_x powder is both pyrophoric and hygroscopic, all containers were doubly sealed to contain an inert atmosphere; thus conventional sampling/chemical assay procedures on the entire inventory would have been very costly and time consuming. Using a 14 MeV neutron source each individual can (containing $\sim$ 2 kg ^{235}U) was assayed nondestructively in three minutes to an accuracy of $\sim$ 1%. Assays were made relative to a standard which was subsequently multiple-sampled and analyzed for U content by chemical means. As an independent check on the consistency between nondestructive assay and sampling/chemical analysis, a total of thirteen cans were examined with the results

TABLE II. COMPARISON OF CHEMICAL ANALYSIS AND NONDESTRUCTIVE ASSAY RESULTS ON THIRTEEN CANS OF HIGHLY-ENRICHED URANIUM ALUMINIDE

UAl Lot and Container No.	NLCo. Inventory g ^{235}U	ICPP Chem. g ^{235}U	LASL NDA, g ^{235}U [a]
9836-2 4	3236.42	3231	3231
9837-5 6	4540.63	4534	4572
9836-2 3	3217.57	3208	3228
9837-5 5-3	3391.54	3380	3425
Composite #1	4286.00	4287	4252
Composite #2	4540.00	4541	4468
9835-60-3 7	2226.26	2219	2204
9834-60-6 3	2503.06	2500	2498
9834-60-5 6	3437.65	3445	3489
9835-60-4 5	3705.51	3724	3712
9834-60-1 5	3799.56	3791	3806
9835-60-1 2	801.02	801	750
9835-60-1 6	3206.39	3216	3192
TOTALS	42,893.43	42,877	42,827

[a] ICPP's chemical assayed content of Lot No. 9836-2, Container 4, used as standard.

shown in Table II. The agreement between nondestructive and chemical assay is seen to be excellent. It is important to note that the favorable situation for sampling/chemical analysis encountered in this example of feed material often does not hold in practice, e.g. see following discussion of studies on high enrichment U-graphite scrap.

Nondestructive assay results on uranium-graphite "Rover" scrap [1] suggested that the sampling/chemical assay procedure normally used to obtain SS values may introduce significant bias in the chemical assay results. Consequently a systematic comparison is being made of the effectiveness and accuracy of both nondestructive assay and sampling/chemical assay as applied to high-enrichment-U scrap materials. The inventory used in this study at Los Alamos includes approximately 560 recovery cans ($\sim$ 3,000 kg bulk weight, $\simeq$ 300 kg enriched uranium) in seven categories, i.e. plant solids, leached fusion solids, sludge, and miscellaneous residues.

At this writing studies on the organic ash category, consisting of 146 cans, have been completed; Table III lists those cans of organic ash for which the sampling/chemical assay differed appreciably from the nondestructive assay. Causes for the errors in the original sampling/chemical values are

[1] Rover scrap consists of 0-70 wt% uranium (nominally 93% ^{235}U) in a variety of matrix materials.

TABLE III. SELECTED ASSAY RESULTS ON ORGANIC ASH "ROVER" SCRAP (93% ^{235}U ENRICHMENT)

Container Can Number	Original Sampling/ Chemical Assay g U	Nondestructive Delayed Neutron Assay g U	New Sampling/ Chemical Assay g U
ASH 430406	34	1625	1576
ASH 430402	186	1533	1480
IAZH 00001	275	446	432
ASH 463703	834	172	150
ASH 282801	104	508	Not available to date
ASH 350901	114	364	311
ASH 301802	758	1319	1193
ASH 340001	317	424	413
ASH 351005	628	760	660
ASH 351101	418	309	282
ASH 430502	812	977	877

not yet understood. The "new sampling/chemical assay" values in the last column of Table III are systematically ~ 10% lower than the nondestructive assay values; the reason for this apparent bias is under investigation. It is abundantly clear that rapid nondestructive assay of whole cans provides an incisive means of checking sampling/chemical assay results, as well as an independent, reliable method of assaying complete material inventories.

Delayed Neutron Assay of ^{235}U-Thorium Carbide Samples

Since certain refractory compounds of U, Pu and Th are difficult to dissolve completely, and have for one reason or another proved especially difficult to assay by chemical analysis, it is clearly desirable to assay these materials by nondestructive methods. Six 5-gram samples of $(^{235}U\text{-}Th)C_2$ carbon-coated microspheres and $^{235}UO_2ThO_2$ powders were obtained from the U.S. AEC's New Brunswick Laboratory and assayed by the delayed neutron yield method. The 3-MeV Van de Graaff accelerator at Los Alamos, provided subthreshold interrogating neutrons in the energy range 355 to 625 keV. The results obtained by delayed neutron assay, together with results obtained with a new ^{252}Cf radioactive source system, and with passive gamma assay, are shown in Table IV. The thorium content of these ^{235}U-Th-C samples has also been determined using "superthreshold" neutron interrogation.

Assay of Power Reactor Fuel Elements

Techniques to measure the total ^{235}U content of whole power reactor fuel elements are under development at Los Alamos. The delayed neutron yield technique [12] was used with 100 to 400 keV interrogating neutrons produced by

TABLE IV. NONDESTRUCTIVE ASSAY OF ^{235}U-THORIUM CARBIDE SAMPLES

Comparison of results obtained by three assay methods: (1) delayed-neutron assay using subthreshold neutron interrogation, (2) fission prompt-neutron assay following interrogation by thermalized-^{252}Cf-neutrons, and (3) 185-keV ^{235}U gamma ray assay.

Sample No.	Delayed Neutron Assay (g^{235}U)	^{252}Cf Assay (g ^{235}U)	Gamma Ray Assay (g ^{235}U)
3-14-17	.148 ± .003	.147 ± .006	.131 ± .009
-38	.441 ± .005	.438 ± .018	.415 ± .026
-50	.212 ± .003	.202 ± .008	.188 ± .011
-57	.383 ± .004	.406 ± .016	.368 ± .028
-129	.905 ± .010	---	.887 ± .098
-197	.328 ± .004	.333 ± .013	.331 ± .025

a Van de Graaff accelerator. A source-sample-detector arrangement was determined which had essentially uniform response across the width of the element [15]. Various individual rods were then removed successively to generate a calibration curve of assayed total mass vs known (weighed) total mass of U in a fuel element. The resulting calibration was linear to within 1% over a 20% change in fuel element loading. Thus practical measurement of total fissile content in a power reactor fuel element to better than 2% accuracy is feasible by means of the delayed neutron method. Further R&D on delayed and prompt [16] neutron assay methods applied to actual PWR and BWR power reactor fuel elements is presently underway.

Assay of Spent Power Reactor Fuel Elements

The determination of burnup of spent reactor fuels, e.g. as inferred from fission product gamma-ray analysis [17], does not yield directly the specific information (fissile content) needed for safeguards or accountability. The delayed neutron yield technique, on the other hand, does provide a practical method for quantitative assay of the total fissile content of irradiated fuel [12]. A combined experimental-analytical approach to nondestructive isotopic assay of spent reactor fuel has been under investigation at LASL for some time. It appears that a delayed neutron yield measurement of total fissile content, combined with appropriate isotope correlation calculations [18], can provide an accurate means of determining the inventory of the major fissile species contained in an individual spent fuel element. Currently, the U and Pu content of spent fuels must be inferred from measurements after batch dissolution of several hot elements.

BREMSSTRAHLUNG INTERROGATION

Photoinduced reactions in the energy interval near the photoneutron thresholds and photofission barriers (5-10 MeV) provide an alternative method for active assay of nuclear materials contained in samples of arbitrary

geometry [19, 20]. Assay approaches which may be utilized with variable energy electron accelerators include detection of prompt neutrons, delayed neutrons and fission product gamma rays from photofission and the use of bremsstrahlung-produced photoneutrons (sub MeV or higher energy) to produce characteristic fission reactions.

The bremsstrahlung beam is quite penetrating and has the desirable characteristic that a single gamma scattering by the matrix material nearly always lowers the photon energy below the threshold for essentially all of the relevant nuclear reactions--notable exceptions being photoneutron production in D, ^{13}C and ^{9}Be. Thus a shaped beam profile can often be used to reduce beam attenuation effects to an unimportant factor even in 55-gallon sized samples weighing hundreds of pounds. When neutrons are the observed reaction product, uncertainties introduced by their scattering can limit measurement accuracy. This scattering can be compensated to some extent by adjustment of the profile of the bremsstrahlung beam.

The electron accelerator has a demonstrated isotopic assay capability for small samples using several different techniques [21]. One technique uses the fact that the ratio of prompt-to-delayed neutrons is significantly different for different isotopes. Another physical basis for isotopic analysis is the different electron energy dependence of the prompt and of the delayed neutrons. Isotopic assays of binary mixtures of nuclear materials have been done using fission product gamma rays produced in small samples (fuel sticks). The use of fission product gamma rays has some advantages, i.e. there are no neutron scattering effects and the gammas result only from fission. Significant progress has been made on minimizing geometrical effects in the assay of large samples. Future developments will include studies on the feasibility of isotopic analysis as applied to the practical assay of 55-gallon barrels.

The bulk sample assay technique being developed at Gulf Radiation Technology involves detection of prompt and delayed neutrons produced by irradiating the sample with a bremsstrahlung beam produced by electrons from the LINAC operating at energies between 6 and 10 MeV. In the lower portion of this energy range, prompt neutrons are produced in only a few materials including isotopes of plutonium, uranium and thorium, while over the whole energy range delayed neutrons provide a unique indication of the presence of fissionable material.

RADIOACTIVE SOURCES FOR NEUTRON INTERROGATION

Because of their inherent reliability and simplicity, radioactive neutron sources provide an attractive means for the nondestructive assay of fissionable materials. Such sources are particularly promising for in-plant measurements and quality control of reactor fuel components as well as some categories of scrap materials and process-stream measurements.

Using subthreshold neutron sources (e.g. $^{238}PuLi$, SbBe) for neutron interrogation the prompt neutrons from fissions induced in the sample can be counted with detectors which are biased above the energy of the interrogating neutrons [22]. Several biased neutron detectors have been evaluated for this purpose including fission counters, liquid scintillators, ZnS detectors, and ^{4}He-gas proportional detectors.

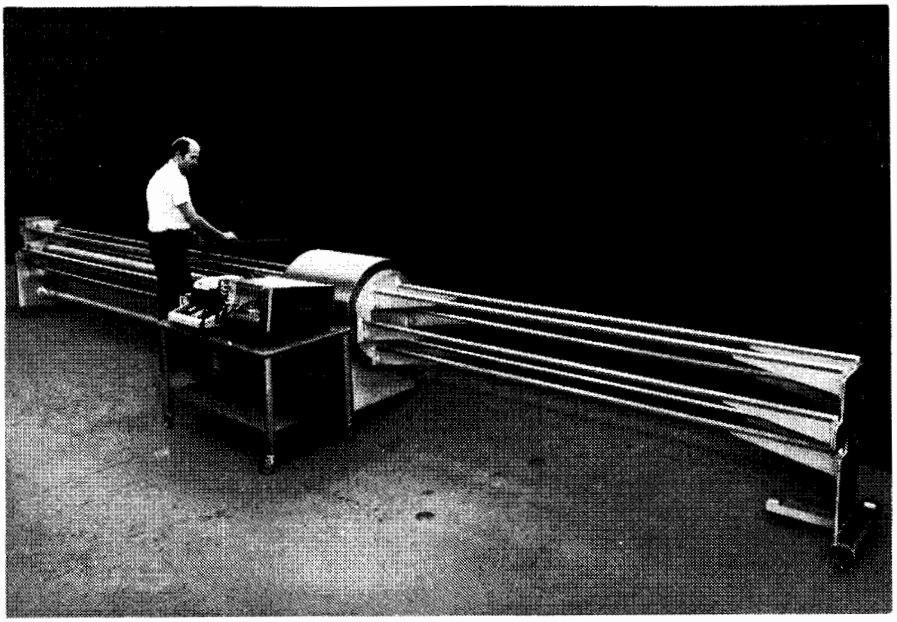

FIG. 1. Complete ^{252}Cf fuel rod assay system with 6-channel rod handling mechanisms, and automated data acquisition, reduction and print-out. Assay accuracy < 1% for fissile content of power reactor fuel rods; assay time ~30 sec for 6 simultaneous channels.

^{252}Cf neutron sources have received considerable attention for assay applications because of their high specific neutron yield (~ 2.4×10^{12} n/g-sec) and low gamma and heat outputs relative to other sources. Transport calculations [23] have been performed to optimize the combination of moderating materials, detectors, and geometric configurations for particular assay applications. Such calculations have been used to design a ^{252}Cf moderator system for assaying the fissile content of low enrichment fuel rods and small inventory verification samples. Moderated neutrons from the ^{252}Cf source interrogate the fuel material and prompt neutrons from the induced fission reactions are counted using energy-biased ^{4}He-gas recoil detectors.

A complete ^{252}Cf assay system (cf. Fig. 1) optimized for power reactor fuel rods [24], includes a fully-engineered 6-channel rod handling mechanism, data acquisition, reduction and printout equipment. Since the high-energy neutrons from the ^{252}Cf source which reach the detector have essentially the same spectrum as the signal neutrons from fission in the sample, this constant background rate has been utilized in a feedback loop to automatically stabilize the response of the system. Performance of the complete fuel rod assay system is currently being evaluated in the plant production line of a major manufacturer of power reactor fuel elements.

Isotopic Source Assay System (ISAS)

A unique fission detector [25], shown in Fig. 2, has been used by Gulf Radiation Technology in a system for assay of nuclear materials contained in heterogeneous samples up to one gallon in size. Either spontaneous or neutron

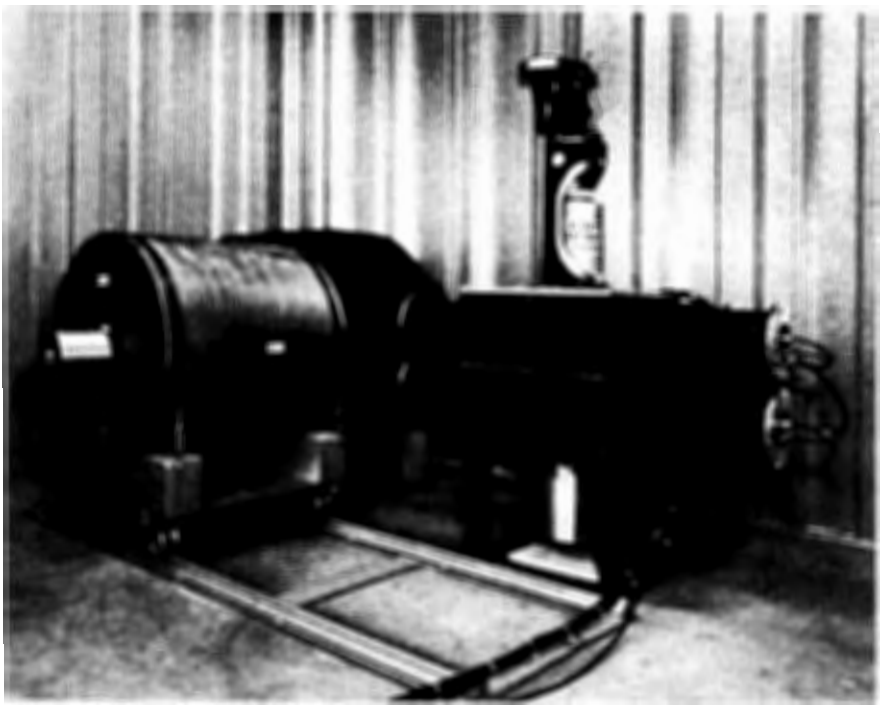

FIG. 2. Isotopic Source Assay System for bulk samples containing uranium or plutonium. The cylindrical neutron collimator of ISAS is moved to one side to show the fission multiplicity detector. This is the configuration used for passive measurements.

induced fission events in the nuclear material are identified by coincidences produced by fission neutrons and gamma rays in four detectors that are adjacent to the sample. Simple modular electronics is used, and the number of fissions observed is read out directly on a teletype.

Plutonium is assayed by measuring the ^{240}Pu spontaneous fission rate and, separately, the ^{239}Pu fissions induced by a neutron source (typically about 20 μg of ^{252}Cf). Assay of ^{235}U is done using neutron irradiation, and ^{238}U content may also be determined separately using the two-spectrum irradiation technique.

The irradiation and detection geometry has been designed to minimize the effects of sample size, neutron scattering, and gamma ray attenuation. The neutrons are collimated to a 4 in. x 8 in. beam at the sample position. For gallon-sized containers an unmoderated neutron spectrum is used, and the sample is rotated and translated through the beam.

Using unmoderated (LiH shielded) neutrons, the sensitivity of the system is less than one gram of ^{239}Pu or ^{235}U, and using spontaneous fission, the sensitivity is about 0.03 grams of ^{240}Pu. Typically, an assay takes about 100 sec. With appropriate standards, the accuracy of measurement ranges from 1 to 20%, depending principally on the extent of neutron scattering effects in the sample to be measured. Typical results using the ISAS on plutonium-bearing samples are shown in Table V.

TABLE V. ISAS ASSAY RESULTS ON ONE-GALLON Pu STANDARDS

Category	Standard	PASSIVE (^{240}Pu)		ACTIVE (^{239}Pu)	
	Pu Content (g)	Calibration (cts/g)	Measured ÷ Standard	Calibration (cts/g)	Measured ÷ Standard
Slag	15	190	0.87	577	1.04
	160		0.997		1.02
	45		0.994		1.07
Graphite Scarfing	25	161	1.015	523	1.03
	175		0.98		0.92
	140		1.01		0.99
Insulation	15	149	0.895	689	0.915
	130		0.935		0.81
	60		1.005		1.025
Glass	1	108	1.06	320	0.99
	38		1.01		0.997
	20		0.93		0.985
Ash Heel	10	152	0.99	746	1.06
	10		1.00		1.04
	55		1.00		0.99
Standard Deviation of mean rates			± 0.045		± 0.05

IN-PLANT AND FIELD ASSAY EXPERIENCE

Since mid-1970 the United States has conducted extensive programs of performance evaluation and testing of nondestructive assay instrumentation in the actual industrial environment. The salient results of in-plant and field assay experience in these field programs are summarized briefly.

Plant Instrumentation Program

During the past year nondestructive assay methods have been applied to a wide variety of materials in commercial nuclear plants under a Plant Instrumentation Program jointly sponsored by the U.S. Atomic Energy Commission and several U.S. Commercial nuclear companies. The purpose of the program was to evaluate nondestructive measurement techniques for quality control and safeguards purposes. The following nondestructive methods were applied: (1) calorimetry of plutonium oxide, plutonium nitrate and mixed plutonium-uranium oxide-feed materials, (2) calorimetry and passive neutron measurement of mixed oxide pellets, (3) passive neutron and gamma ray assay of wastes and filters containing small amounts of mixed oxides, (4) active neutron assay of recoverable scrap containing substantial amounts of mixed oxides, as well as scrap and waste containers of low to high enriched uranium, and (5) gamma-ray assay of mixed-oxide rods.

Experience obtained with the several methods is summarized briefly below. Further details are given in the project reports and a number of papers describing specific instruments or applications.

Mixed Oxide Fuel Rods

Two classes of mixed plutonium-uranium oxide rods are presently being fabricated in small quantities in the United States: experimental plutonium recycle rods for use in light water reactors and rods for breeder reactor experiments. The former have a PuO_2/UO_2 ratio of 2 to 5%, the latter a ratio of 20-30%. Plutonium which has been recovered from operating power reactors has a high concentration of Pu-240, appreciable amounts of Pu-238, 241 and americium and possibly traces of fission products and U-232. Consequently the gamma spectra of recycle fuel are both complex and variable from batch to batch. Even with the high resolution of a germanium detector, care must be exercised to avoid mistakes. Since pellets range in diameter from ½ to 1½ cm, gamma ray absorption is significant. Various combinations of NaI(Tl) and Ge(Li) detector methods were investigated at GE (Pleasanton, Calif.) and Westinghouse (Cheswick, Penna.). For quality control purposes, a pellet-by-pellet scan of each rod is desired, whereas for safeguards purposes one desires a reliable integral measurement of the plutonium in each rod. Using reasonable values for collimator and detector dimensions, a NaI(Tl) detector gives sufficient counting rate to identify "bad" pellets at a scan rate of ~½ cm per sec. (Rods containing ≥ 20% PuO_2 give sufficient counting rates for pellet-by-pellet assay using Ge(Li) detectors.) As shown by work at GE-Pleasanton and LRL-Livermore, a multichannel analyzer is required to accurately strip the Pu activity from the complex spectrum actually observed. While accuracies obtained in these experiments were in the range of 1%, it should be possible to obtain accuracies of better than 0.25% [26].

A number of plutonium-recycle rods were also scanned using an active interrogation device which employs an isotopic neutron source to induce fissions in the fuel, the resulting fission neutron signature then being observed with a fast neutron detector [27]. Measurement precision was comparable to that obtained using gamma-ray assay.

PuO_2 Feed Materials, Product and Scrap

Mound Laboratory (Monsanto Co., Miamisburg, Ohio) supplied two calorimeters for use at the GE Plutonium Fuels Development Center, Sunnyvale, Calif. The larger calorimeter was used to measure feed materials and scrap and the smaller unit was used for pellets and samples.

Significant differences occur between shipper and receiver measurements of PuO_2 due to sampling errors, loss or gain of moisture and possibly other effects. Provided isotopic ratios can be precisely determined, calorimetric measurements offer a promising alternative to or verification of chemical analysis. For a number of cans, each containing about ½ kg of PuO_2, the precision of a given reading was .025% or less while the calorimetric assay differed from chemical assay by ± 0.3% due to inter-laboratory differences in isotopic analyses. Similar results were obtained for plutonium-nitrate solution in 2-litre bottles (chemical heating may affect results for freshly filled bottles). Poorer results were obtained for pellets and scrap samples because the amount of Pu in the samples was below that required for optimum use of the calorimeters available.

Recoverable PuO_2/UO_2 Scrap

Recoverable scrap may consist of cracked pellets, dust from glove boxes, grinder sludge, ashed combustibles, etc., in a variety of packages; quantities vary from a fraction of a gram to a kilogram or more of mixed oxide.

An effort was made at Nuclear Materials and Equipment Corporation (Apollo, Penna.) to measure the content of such packages using a Ge(Li) detector and making corrections for self-absorption of gamma-rays. Since the self-absorption is very high and the density variable, the preliminary results were not encouraging. Most of the measurements of this class of material (made by GE, NUMEC and Westinghouse) were made using a neutron coincidence well counter [7] which measures Pu-238, 240, 242 spontaneous fission neutrons with a (coincidence) detection efficiency of about 5%. When the scrap is segregated by campaign, so that the relative abundance of these isotopes is known, relatively accurate (5-10%) measurements can be made. For example, some 50 scrap and 80 waste containers from one campaign were measured for 10 minutes each, placing the total plutonium inventory at 650 ± 50 g (95% confidence level).

Uranium Scrap

Passive gamma ray methods have been widely used for measurement of uranium although self-absorption of the 185 keV gamma rays from U-235 is a serious problem in many cases. Also, since passive neutron and calorimetric methods are not applicable, active assay methods are of considerable importance in uranium scrap assay. Three active assay stations were installed in commercial plants to measure uranium scrap. One station employs a neutron generator (sealed tube) for neutron interrogation and delayed neutron assay. The other two make use of isotopic neutron sources (^{252}Cf) and a high-efficiency coincidence detector. These systems were described in some detail in earlier sections of this paper.

The neutron generator and one of the isotopic source units are installed at the United Nuclear Corp. (UNC) plant in Wood River Junction, R. I. The second isotopic source unit is at the UNC plant in Hematite, Mo. The isotopic source units have been used to scan a large variety of scrap in small containers (15 cm diam. or less) on a production basis. The neutron generator system, which also can assay ^{235}U in large containers, e.g. 55-gallon barrels, had been in operation only a short time when this report was written. The experience at both plants has been encouraging.

Low Level PuO_2/UO_2 Wastes

The wide variety of low level wastes, small amounts of oxide mixed with paper, rubber, bricks, etc. is usually packed in small containers (1 to 10 litres) which are then packed in 55-gallon drums. NUMEC has applied Ge(Li) assay systems to waste packages as well as 55-gallon drums [28], and similar methods have been applied at GE, United Nuclear Corp. (Pawling, N.Y.) and Westinghouse. In brief, this work showed that more accurate measurements can be made on small packages than on large drums and, as expected, drum measurements are more accurate if the contained packages are similar in composition. Corrections can be made for self-attenuation either by measuring transmission through the container using an external source or in some cases by comparing the self-attenuation for gamma-ray lines of different energy from the same isotope (e.g. Pu-239). The latter method may be more sensitive to the effect of local inhomogeneities than the former, but it requires the use of a high resolution detector and more complicated data analysis.

In summary, the plant instrumentation program has been a cooperative effort involving many individuals. In addition to the institutions mentioned, scientists at Argonne National Laboratory, Gulf Energy and Environmental Systems, Idaho Nuclear Corp., and the Livermore and Los Alamos Scientific

Laboratories assisted in designing instruments and solving problems. It has been demonstrated that a wide range of materials infuel fabrication and scrap recovery plants can be measured rapidly and economically using nondestructive assay instrumentation.

Gulf Atomic Materials Assay System (GAMAS)

The Gulf Radiation Technology transportable assay system (GAMAS) is housed in a 39 ft. x 8 ft. trailer. The major components of the system are an electron linear accelerator, a barrel scanner, active and passive radiation detectors and electronics instrumentation. GAMAS was designed to provide a general nondestructive assay capability, particularly for large containers, and at the same time be a tool for research and development on assay techniques. Construction of GAMAS was completed in July 1970, and field tests have been performed at a site with a scrap inventory of enriched uranium and at a plutonium facility.

A radiation-shielded electron linear accelerator was designed and constructed which provides high energy electrons (4.5 - 9 MeV) with better than 10% energy resolution. After energy analysis by a bending magnet, a portion of the primary electrons strikes a bremsstrahlung converter to produce the active barrel scanning beam. The barrel scanner rotates the barrel and moves it through the bremsstrahlung beam, and the resulting neutrons are detected on two sides of the barrel by BF_3 tubes embedded in polyethylene moderator. The detected neutron pulses are initially stored in a scaler system which is interfaced to a Hewlett-Packard 2116B computer.

The active assay technique used in the first two field tests of GAMAS is based on the photoinduced prompt neutron yield. During irradiation the barrels were moved through the 12-inch-wide fan-shaped bremsstrahlung beam in such a way that uniform sensitivity to nuclear material at any position in the barrel was obtained [19]. For assay of 55-gallon barrels of typical scrap using prompt neutrons, a non-negligible yield is obtained from the matrix materials themselves (e.g. D, Be and C). The contributions from D and Be to total prompt neutron yield are measured through their energy dependence and subtracted to determine the quantity of nuclear material.

The results obtained with GAMAS on low density enriched uranium waste from a fuel fabrication facility showed that the sensitivity to uranium in the active bremsstrahlung assay of a 55-gallon drum was less than 5 g. Passive results obtained using an optimized collimation and scan procedure were very useful, but not very accurate ($\sim$ 30% at best) because of the large attenuation effects present in these inhomogeneous drums. Since the uranium content of the 34 barrels assayed was indeed unknown there was no independent check of the accuracy of the assay results. For one-gallon samples containing uranium, standards were available to determine the accuracy of active and passive assays. The results, in Table VI, show that 7% accuracy was obtained for passive and 1-2% for active measurements.

At the plutonium facility, standard 55-gallon drums were available and a detailed check of the assay accuracy on large drums containing randomly distributed plutonium was possible. In standard drums containing dry combustibles, Raschig rings, wet combustibles, resin, washables, graphite and Benelex-Lucite, the standard deviation was ± 7% if the waste is segregated. In the more typical situation of incomplete segregation of waste in an unknown drum, present assay accuracy would in most cases be about 15%. Both active and passive

TABLE VI. ACTIVE AND PASSIVE RESULTS FOR ONE-GALLON URANIUM RESIDUE STANDARDS

Nominal Uranium (g)	PASSIVE		ACTIVE
	Uncorrected (g)	Corrected [a] (g)	(g)
(5.38)	(5.38)	(5.38)	(5.38)
10.8	10.44	10.74	10.8 ± 0.2
21.5	18.02	19.06	21.4 ± 0.2
53.8	41.61	47.3	53.5 ± 1.3
109.0	78.06	103.0	107.3 ± 1.9
218.0	132.26	224.3	219.6 ± 4.4
Standard Deviation from known content		7%	1%

[a] The passive results were corrected by assuming a homogeneous distribution of uranium to compute an attenuation correction.

(NaI) measurements were performed on 20 production waste drums containing Pu. In contrast to the case for uranium, the passive results on Pu are reliable (assuming no Pu lumps) and have an average deviation of 20% from the active results. The accuracy for active uranium assay in drums is the same as that determined for the Pu drums. Whereas active techniques are usually essential for accurate assay of uranium in 55-gallon drums, passive assay may be sufficient for Pu.

Mobile Nondestructive Assay Laboratory (MONAL)

Another important tool in the U.S. Atomic Energy Commission's field assay program is the Los Alamos Scientific Laboratory's Mobile Nondestructive Assay Laboratory, MONAL [29], which contains a full range of passive and active assay instrumentation. Following initial systems tests at the Los Alamos plutonium processing facility, the MONAL was deployed (during May and June 1970) at Dow Chemical Company's Rocky Flats Division (RFD).

The accuracy of the MONAL neutron coincidence can counter was demonstrated on 1-gallon bottles of Pu scrap and waste in the RFD scrap recovery plant by treating sets of RFD production standards as unknowns. For very heterogeneously distributed plutonium (3 to 155 g Pu) in five different matrix categories the assay accuracy was ± 8%. In the case of the homogeneous standards containing 10 to 500 g Pu in graphite or ash the assay accuracy was ± 3%. The average counting time per sample was approximately 15 minutes including the "add-a-source" matrix correction [11].

A set of RFD 55-gal production standards (scrap and waste) containing 10 to 195 g Pu in a range of low Z and hydrogenous matrices were used for parametric studies of neutron interrogation as well as a test of the accuracy of the MONAL 8-channel NaI barrel scanner. In addition, two groups of barrels containing process line waste were selected at random, namely seven metal scrap barrels for neutron interrogation and seven low-Z production scrap barrels. The results can be summarized as follows: (1) The two complementary techniques of active neutron interrogation and passive gamma assay enabled MONAL to cover the full range of matrix materials with the assay accuracies (for 55-gallon barrels) indicated below. (2) Active neutron interrogation for high-Z

matrix materials: demonstrated assay accuracy 15-20%. (3) Passive gamma assay for low-Z matrix materials: demonstrated assay accuracy 11% for well characterized mixtures. (4) The high fissile material sensitivity of active neutron interrogation has been demonstrated at the ~ 5 g level in non-moderating matrices and the < 0.1 g level in moderating matrices.

From November 1970 to March 1971 MONAL was deployed at the Feed Materials Production Center, National Lead Company of Ohio (NLO), Cincinnati, with the assignment of assaying UC, UO_2, and U_3O_8 scrap (≤ 10% ^{235}U enrichment) in various fuel rods, pellets, and incinerator ash. The NLO inventory included 196 cans of U_3O_8 residues (specified enrichment 1-5% ^{235}U) from the Oak Ridge gaseous diffusion plant. Neutron coincidence counting of ^{238}U and Ge(Li) "enrichment meter" measurements were successfully combined to determine the SS-value and enrichment passively for all 196 cans; assay accuracies were 5% for the enrichment (5 minutes counting time), 5% and 10-15% for the SS value of heavy and light cans, respectively (10 minutes total assay time). As noted earlier the "enrichment meter" revealed unexpected large variations in enrichment (up to 30%), in contrast to the specified mass-spectrometric values of 1-5% (based on a single-sample mass-spectrometric determination for each can). Passive assay results, including isotopic composition, were verified by active assay (8 min. per sample) on ~ 120 of the cans.

The MONAL "enrichment meter" was also used for assay of UO_2 fuel pellets and scrap fuel rods of varying diameter, cladding, and uranium composition with enrichment ranging from depleted U to 10% ^{235}U.

In March 1971 MONAL was deployed at the Nuclear Materials and Equipment Corporation (NUMEC), Leechburg, Penna., to measure a substantial amount of enriched (≥ 20%) uranium scrap.

Field assay experience has repeatedly demonstrated the value of a fully-instrumented Mobile Nondestructive Assay Laboratory in effecting vital technical interaction between industry and the scientists developing assay techniques. The bulk of fissionable material-unaccounted-for (MUF), in U.S. facilities at least, is in the area of scrap and waste which is characteristically difficult to assay chemically because of sampling problems. In the future, nondestructive assay will be applied increasingly to samples, or whole containers, of materials in these "difficult" categories. Neither sampling nor shipping of bulk quantities of scrap and waste are really satisfactory from the accountability, economic and contamination-hazard standpoint. Thus if these materials are to be measured economically and expeditiously (as will likely be required in future), this task must be carried out at the plant site using nondestructive assay techniques. Such future in-plant inventory measurements, and perhaps ultimately the overall material balance of a plant, could then be verified (on a random sampling basis) by an effective field assay and inspection capability, e.g. as implemented by a mobile assay facility.

CONCLUSIONS

For the decade of the 70's all indicators point toward increasingly stringent safeguards and accountability requirements as well as tighter environmental, quality control and waste management constraints. All these factors, coupled

with increased competitive pressures and just plain economic self-interest will require all nuclear facilities (government and commercial) to take full advantage of nondestructive assay instrumentation. The in-plant use of nondestructive assay, especially as applied to whole containers of materials which are difficult to sample or otherwise not amenable to wet chemical analysis, can effect very significant savings in time and money while maintaining, and oftentimes even increasing, overall assay accuracy of a complete inventory.

The future role of instrumented material measurement has long been heralded by safeguards experts from many countries [30-32]. It is therefore most gratifying to be able to report at this Geneva Conference that new nondestructive assay instrumentation is now beginning to contribute substantially to safeguards and materials management as well as to increased quality control and operational safety throughout the nuclear fuel cycle.

REFERENCES

[1] REILLY, T. D., LA-4457-MS (1970) 21.

[2] PARKER, J. L., REILLY, T. D., and WALTON, R. B., LA-4523-MS (1970) 29.

[3] TINGEY, F. H., Nucleonics 20 (1962) 76.

[4] BEYER, N. S., HURST, L. K., PERRY, R. B., and BRANDENBURG, R. W., Proc. Tenth Annual Meeting of the Institute of Nuclear Materials Management, Las Vegas, Nevada (1969) 143.

[5] REILLY, T. D., WALTON, R. B., and PARKER, J. L., LA-4605-MS (1970) 19.

[6] WALTON, R. B., and MENLOVE, H. O., LA-4457-MS (1970) 26.

[7] STRAIN, C. V., and OMOHUNDRO, R. J., U. S. Naval Research Laboratory Report No. 2107 (1970); STRAIN, C. V., ibid. No. 2127 (1970).

[8] FOLEY, J. E., LA-4605-MS (1970) 18, 34.

[9] WALTON, R. B., MENLOVE, H. O., AUGUSTSON, R. H., and CALDWELL, J. T., LA-4315-MS (1969) 20.

[10] FOLEY, J. E. and WALTON, R. B., LA-4523-MS (1970) 18.

[11] MENLOVE, H. O., AUGUSTSON, R. H., EAST, L. V., EVANS, A. E., and KEEPIN, G. R., Proc. Eleventh Annual Meeting of the Institute of Nuclear Materials Management, Gatlinburg, Tenn. (1970) 316.

[12] MENLOVE, H. O., AUGUSTSON, R. H., and SMITH, D. B., Nucl. Technology 10 (1971) 366.

[13] JONES, D. W., MALMBERG, P. R., MAY, T. H., and STRAIN, C. V., Nucl. Appl. Technol. 8 (1970) 79.

[14] AUGUSTSON, R. H., MENLOVE, H. O., WALTON, R. B., EAST, L. V., EVANS, A. E., and KRICK, M. S., Proc. IAEA Symposium on Progress in Safeguards Techniques, II, Vienna (1970) 79.

[15] AUGUSTSON, R. H., and EVANS, A. E., LA-4523-MS (1970) 6.

[16] AUGUSTSON, R. H., and EVANS, A. E., LA-4605-MS (1970) 7.

[17] FORSYTH, R. S., "Burnup determination of fuels by fission product gamma-ray analysis", Trans. Am. Nucl. Soc., Seventeenth Annual Meeting, Boston, June 13-17, 1971.

[18] WEISBIN, C. R., and THORPE, M. M., LA-4605-MS (1970) 29.

[19] BRAMBLETT, R. L., GINAVEN, R. O., GOZANI, T., McMILLAN, J. I., REYNOLDS, G. M., and RUNDQUIST, D. E., Proc. IAEA Symposium on Progress in Safeguards Techniques I, Vienna (1970) 223.

[20] GOZANI, T., RUNDQUIST, D. E., GINAVEN, R. O., and BRAMBLETT, R. L., Proc. IAEA Symposium on Progress in Safeguards Techniques II, Vienna (1970) 143.

[21] BRAMBLETT, R. L., J. of Nondestructive Testing 2 (1970) 99.

[22] MENLOVE, H. O., "Experience with ^{252}Cf and subthreshold neutron sources for nondestructive assay", Trans. Am. Nucl. Soc., Seventeenth Annual Meeting, Boston, June 13-17, 1971.

[23] FORSTER, R. A., and MENLOVE, H. O., "Moderator investigation of ^{252}Cf for nondestructive assay of fissionable materials", Am. Nucl. Soc. National Topical Meeting, Neutron Sources and Applications, Augusta, Ga., April 1971 (to be published).

[24] MENLOVE, H. O., and FORSTER, R. A., LA-4605-MS (1970) 10.

[25] GOZANI, T., and COSTELLO, D. G., Trans. Am. Nucl. Soc. 13 2 (1970) 746.

[26] BRANDENBURG, R. W., BEYER, N. B., and PERRY, R. B., Materials Evaluation 28 4 (1970) 77.

[27] UNTERMEYER, S., J. of Nondestructive Testing 1 (1969) 223.

[28] DUKAT, A. J., GONSER, J., and JAMES, D. B., Trans. Am. Nucl. Soc. 13 2 (1970) 749.

[29] MENZEL, J. H., DENNIS, B. R., THORPE, M. M., WALTON, R. B., SMITH, D. B., and KEEPIN, G. R., Proc. IAEA Symposium on Progress in Safeguards Techniques I, Vienna (1970) 201.

[30] "International Safeguards", Joint Plenary Session of the American Nuclear Society/Atomic Industrial Forum; in Proceedings, International Conference on Constructive Uses of Atomic Energy, Washington, D. C., November 10-15, 1968.

[31] Proceedings, U. S. Atomic Energy Commission Symposium on Safeguards Research and Development, WASH-1147, Los Alamos, New Mexico, October 27-29, 1969.

[32] Proc. IAEA Symposium on Progress in Safeguards Techniques, Karlsruhe, Germany, July 6 - 10, 1970.

INSTRUMENTED NUCLEAR MATERIAL ASSAY FOR SAFEGUARDS

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Abstract–Résumé–Аннотация–Resumen

INSTRUMENTED NUCLEAR MATERIAL ASSAY FOR SAFEGUARDS.

This paper summarizes activities in the field of instrumented methods for nuclear material assay under way in the Federal Republic of Germany. Both destructive and non-destructive techniques are described.

The development of direct methods is dominated by (i) the automation of mass-spectrometrical determination of isotopic composition and concentrations of uranium and plutonium, (ii) the measurement of uranium and plutonium concentrations by X-ray fluorescence spectrometry. While a fully-automated system for X-ray fluorescence analysis will be available by the end of 1971, the development of the mass-spectrometric method has so far been limited to the automation of the spectrometer itself; automatic sampling and chemical processing is expected to be ready for use by 1973. A comprehensive comparative analytical program was performed in collaboration with numerous international laboratories. Results are indicated and discussed.

Indirect methods cover a variety of instruments that have been built for plant application, namely (i) calorimeters, (ii) slowing-down spectrometers, (iii) delayed-neutron analysers.

Several types of calorimeters for the assay of plutonium have been built and successfully used at the ALKEM fuel element fabrication plant. Calorimetry is supplemented by neutron measurements. For separate assay of ^{239}Pu and ^{235}U in fuel pins the slowing-down spectrometer is used. Equipment for routine industrial application has been developed at INTERATOM. The delayed-neutron technique for assay of highly enriched nuclear fuel has been applied at the University of Hannover. Operation, data acquisition and reduction is fully automated. Sub-threshold neutron-induced fission measurements using an Sb-Be neutron source can be used for non-destructive fissile material assay of low-enrichment fuel. Investigations on the feasibility of this technique were conducted. The fundamentals for assay of individual isotopes in nuclear fuel by gamma-ray spectrometry following neutron capture have been worked out at Karlsruhe.

METHODES DE DOSAGE DES MATIERES NUCLEAIRES POUR LES GARANTIES AU MOYEN D' APPAREILS SPECIAUX.

Le mémoire résume les travaux entrepris en République fédérale d' Allemagne sur les méthodes d' analyse des matières nucléaires, tant destructive que non destructive.

On s' efforce surtout de mettre au point des méthodes directes permettant 1) de déterminer par spectrométrie de masse automatique la composition isotopique et la concentration de l' uranium et du plutonium

et 2) de mesurer la concentration du plutonium et de l'uranium par spectrométrie à fluorescence X. Un ensemble entièrement automatique devait être prêt vers la fin de 1971 pour l'analyse par fluorescence X, mais la mise au point de la méthode par spectrométrie de masse dépend de l'automatisation du spectromètre lui-même. On pense que le prélèvement et le traitement chimique automatiques des échantillons seront possibles à partir de 1973. Un vaste programme d'analyses comportant des comparaisons de résultats a été exécuté en collaboration avec plusieurs laboratoires internationaux. Les auteurs donnent et discutent les résultats.

Les méthodes indirectes comportent plusieurs appareils construits pour l'utilisation industrielle, notamment des calorimètres, des spectromètres de ralentissement et des analyseurs de neutrons différés.

Plusieurs types de calorimètres servant au dosage du plutonium ont été construits et sont utilisés dans l'usine de fabrication d'éléments combustibles ALKEM. Les mesures calorimétriques sont complétées par des mesures de neutrons. Le spectromètre de ralentissement sert à doser séparément ^{239}Pu et ^{235}U dans les aiguilles de combustible. INTERATOM a mis au point un matériel à usage industriel. La méthode des neutrons différés pour l'analyse du combustible nucléaire fortement enrichi est appliquée à l'Université de Hanovre. Le calcul des variables ainsi que l'acquisition et l'exploitation des données sont complètement automatiques. Les mesures de la fission induite par des neutrons subcadmiques d'une source à Sb-Be peuvent être utilisées pour l'analyse non destructive de matières fissiles dans le combustible faiblement enrichi. On a étudié les possibilités de cette méthode. Les principes fondamentaux de l'analyse par spectrométrie gamma après capture de neutrons pour doser les différents isotopes présents dans le combustible nucléaire ont été mis au point à Karlsruhe.

АНАЛИЗ ЯДЕРНОГО МАТЕРИАЛА С ПРИМЕНЕНИЕМ КОНТРОЛЬНО-ИЗМЕРИТЕЛЬНЫХ ПРИБОРОВ ДЛЯ ЦЕЛЕЙ ГАРАНТИЙ.

В данном докладе кратко излагается деятельность в области разработки инструментальных методов анализа ядерных материалов, применяемых в Федеративной Республике Германии. Дается описание методов без разрушения и с разрушением образца.

Разработка прямых методов направлена, главным образом, на автоматизацию масс-спектрометрического определения изотопного состава и концентраций урана и плутония, а также на измерение урановых и плутониевых концентраций при помощи рентгеновской флюоресцентной спектрометрии. Поскольку полностью автоматизированная система для осуществления рентгеновского флюоресцентного анализа будет иметься в распоряжении к концу 1971 года, то разработка масс-спектрометрического метода до сих пор ограничивалась проблемой автоматизации самого спектрометра; ожидается, что автоматическое взятие проб и химическая обработка будут готовы для использования к 1973 году. В сотрудничестве с многочисленными международными лабораториями была выполнена обширная сравнительная аналитическая программа. В докладе приводятся и обсуждаются результаты выполнения этой программы.

Косвенные методы охватывают широкий ассортимент контрольно-измерительных приборов, разработанных для применения на установках, а именно: i) калориметры; ii) спектрометр замедления; iii) анализатор запаздывающих нейтронов.

Изготовлены и успешно используются несколько типов калориметров для анализа плутония на заводе по изготовлению топливных элементов ALKEM. Калориметрические измерения дополняются нейтронными измерениями. Для отдельного анализа плутония-239 и урана-235 в тепловыделяющих элементах, имеющих вид тонких стержней, используется спектрометр замедления. Оборудование для обычного промышленного применения было разработано ИНТЕРАТОМом. Анализатор запаздывающих нейтронов для анализа высокообогащенного ядерного топлива применялся в Университете в Ганновере. Эксплуатация, получение и обработка экспериментальных данных полностью автоматизирована. Подпороговые измерения деления под действием нейтронов с использованием нейтронного источника Sb-Be могут быть использованы для анализа без разрушения образца делящегося вещества для низкообогащенного топлива. Проведены исследования практического применения этого метода. Основные вопросы проведения анализа отдельных изотопов в ядерном топливе при помощи гамма-спектрометрии с последующим захватом нейтронов были разработаны в Карлсруэ.

METODOS INSTRUMENTALES PARA EL ANALISIS DE MATERIALES NUCLEARES EN RELACION CON LA APLICACION DE SALVAGUARDIAS.

En la presente memoria se resumen las actividades desarrolladas en la República Federal de Alemania en la esfera de los métodos instrumentales para el análisis de los materiales nucleares. Se describen tanto técnicas destructivas como no destructivas.

En el desarrollo de métodos directos representan un papel primordial: 1) la automatización de la determinación, por espectrometría de masas, de la composición isotópica y de las concentraciones de uranio y

plutonio, 2) la medición de las concentraciones de uranio y de plutonio mediante espectrometría por fluorescencia de rayos X. Si bien para fines de 1971 se dispondrá de un sistema totalmente automatizado para efectuar análisis por fluorescencia de rayos X, el desarrollo del método de la espectrometría de masas se ha limitado hasta la fecha a la automatización del espectrómetro propiamente dicho. Se espera poder disponer en 1973 de una toma de muestras y de un tratamiento químico automáticos. En colaboración con numerosos laboratorios internacionales, se ha llevado a cabo un amplio programa de análisis comparativos. Se dan a conocer y se examinan los resultados obtenidos.

Los métodos indirectos comprenden una serie de instrumentos construidos para utilizarlos en las instalaciones, a saber: 1) calorímetros, 2) espectrómetro de retardación, 3) analizador de neutrones retardados.

Se han construido diversos tipos de calorímetros para el análisis del plutonio, los cuales se emplean con éxito en la planta de fabricación de elementos combustibles de la ALKEM. La calorimetría se complementa con mediciones neutrónicas. El espectrómetro de retardación se utiliza para analizar por separado el ^{239}Pu y el ^{235}U en agujas de combustible. La INTERATOM ha construido un aparato que puede emplearse corrientemente en la industria. La técnica de los neutrones retardados se ha aplicado en la Universidad de Hannover para analizar combustibles nucleares altamente enriquecidos. El funcionamiento del dispositivo de medida y la obtención y reducción (simplificación) de los datos están totalmente automatizados. La medición de las fisiones inducidas por neutrones de energía inferior a la umbral, procedentes de una fuente de Sb-Be, se prestan al análisis no destructivo de los materiales fisionables contenidos en un combustible de bajo enriquecimiento. Se han llevado a cabo estudios sobre la viabilidad de este método. También se han establecido en Karlsruhe los principios fundamentales para el análisis de los distintos isótopos contenidos en los combustibles nucleares, por espectrometría de los rayos gamma emitidos a raíz de una captura neutrónica.

1. INTRODUCTION

A rational and objective nuclear material safeguards system requires, as a tool for material accountancy, non-destructive as well as direct methods for fissile material assay. In the Federal Republic of Germany a variety of techniques are under development. This paper describes instruments for direct analysis (mass spectrometer, X-ray fluorescence analyser) and non-destructive assay in plant application (calorimeter, slowing down spectrometer and delayed neutron analyser). Special emphasis is given to automation. Feasibility studies and check measurements pertinent to a variety of new methods (assay with Sb-Be neutrons and by neutron capture γ-ray spectroscopy) are also reported.

2. DESTRUCTIVE NUCLEAR MATERIAL ASSAY

In those parts of the nuclear fuel cycle in which the fissile material is present in the form of solutions or powders, direct methods of analysis are usually employed to determine concentrations and for isotope analyses. In order to fulfil the requirements of nuclear safeguards with respect to reliability, accuracy and tamperproofness of the analytical results, present efforts at the Karlsruhe Nuclear Research Centre are concentrated on the complete automation of both mass spectrometry and X-ray fluorescence spectroscopy.

2.1. Automatic isotope dilution analysis by mass spectrometry

Isotope dilution analysis by mass spectrometry is used for simultaneous determination of the isotopic composition and concentration of uranium and plutonium in nuclear fuel solutions [1].

The sampling procedure is followed by dilution of the sample solution to a concentration of about 1 μg Pu/ml. After addition of a spike (^{233}U, ^{242}Pu) a subsequent redox cycle ensures homogeneous mixing of the isotopes. Interfering elements are removed by ion exchange, and about 10 ng of the purified uranium and plutonium is put on the sample carriers of a mass spectrometer. The double filament technique is used to measure first the thermions of uranium and subsequently, at higher temperature, the thermions of plutonium with a single-stage mass spectrometer. The elemental concentrations and isotopic compositions are then calculated from the registered isotopic ratios and added quantities of spike material.

Systems analysis studies have shown that the system can be sub-divided into four consecutive processes, namely (1) sampling, (2) chemical processing of the samples and transfer to the sample carriers, (3) introduction into the spectrometer and mass spectrometric measurement, and (4) data processing [2].

Sampling problems due to the instability of the high radioactive solutions during storage are avoided by on-line analysis with immediate addition of the spikes. If later confirmation of the analytical data should be necessary, stabilized samples can be prepared in the following way: A known aliquot of the sample is pipetted into an aluminium capsule and weighed, carefully dried in the capsule, and the capsule with the residue is closed. In this form, the sample can be stored over a prolonged period of time and yet remain stable. The sampling stage and the chemical sample preparation unit are conceived as modules with very few types of design elements such as turntables, automatic grips, pipetting units, etc; this ensures flexibility and facilitates assembly and maintenance. Design and construction will be done by industrial firms.

A high-vacuum lock system consisting of three chambers was designed for increased capacity of the spectrometer. Several samples are simultaneously preheated in this lock before individual introduction into the ion source of the spectrometer. The lock is currently under construction. The automatic mass spectrometer will be controlled, and measured data will be evaluated by a process computer. The data reduction program has already been finished and checked.

Automating an analytical process of this complexity is not devoid of difficulties and problems. However, we think it is necessary because of the requirements of objectivity and tamperproofness for an instrumented nuclear safeguards system, and because large analytical capacity is needed at low operating cost.

Prototypes of the various components will probably be finished by 1972 (spectrometer control and high vacuum lock) and 1973 to 1974 (chemical processing and sampling process). After a transition period of continuous operation and subsequent improvement, the whole system will, hopefully, mature into some sort of 'black box'.

2.2. Automatic X-ray fluorescence analysis

In experiments described earlier [3] we were able to show that X-ray fluorescence spectrometry may successfully be used for the analysis of solutions of nuclear materials, both non-irradiated and irradiated. With concentrations around or above 100 μg plutonium per gram of solution, it is possible to achieve relative accuracies of better than 1% even in the presence

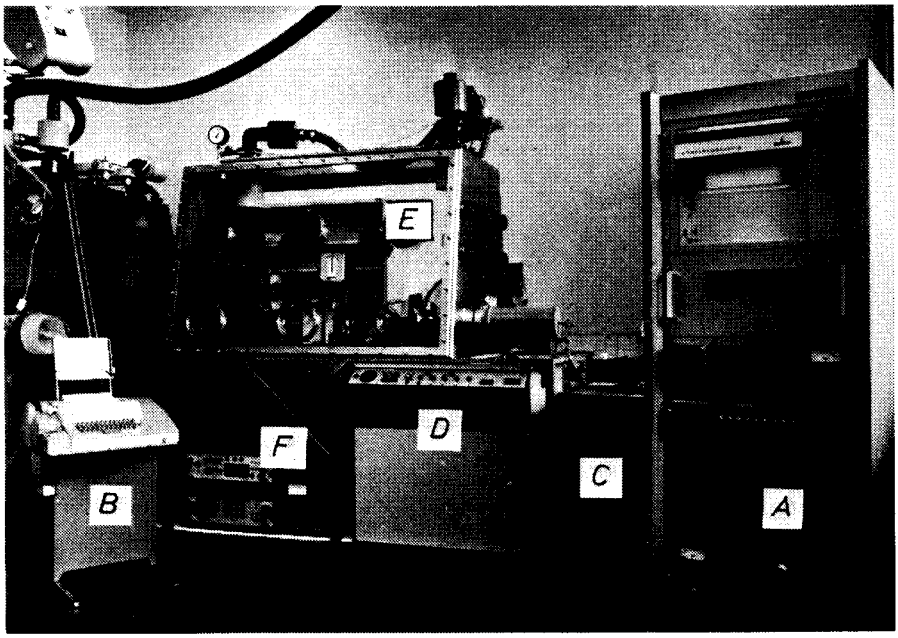

FIG.1. Automated X-ray fluorescence analysis system.
A: Counting electronics, spectrometer control unit; D: Spectrometer;
B: Tape puncher; E: Sample preparation;
C: High voltage supply F: Control electronics.

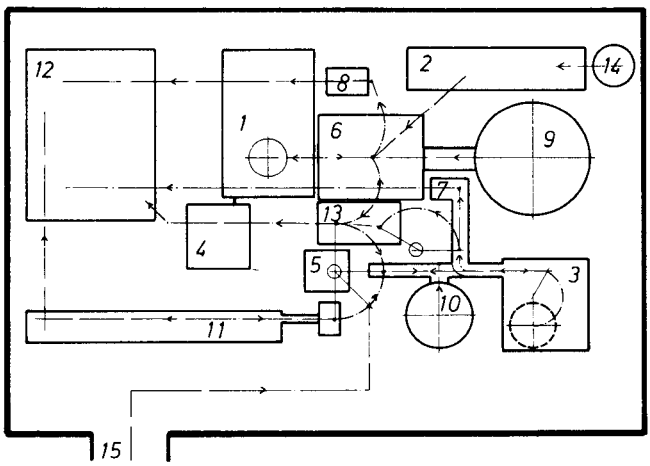

FIG.2. Automatic X-ray fluorescence analysis sample preparation.
1: Electronic balance H20E; 2: Dosing pumps; 3: Sample changer; 4: Balance adjusting device; 5: Pipetting device; 6+7: Beaker transport system; 8: Solid waste transport system; 9+10: Beaker magazines; 11: Pipette magazine; 12: Waste storage. 13: Liquid waste drain; 14: Storage for reagents and standard solutions; 15: Sample transport system.

of a 300-fold excess of uranium. This is sufficient for nuclear safeguards purposes. High fission product activities and other impurities can be tolerated. In these measurements we always employ the method of internal standards; as a rule, we use thorium as the internal standard.

In the past eighteen months we started automating the process. Our system consists of three main components:

(1) automatic sample preparation stage,
(2) automatic spectrometer, and
(3) computer for data evaluation.

A prototype of the automatic sample preparation stage and the automatic type SRS1 spectrometer have been completed and are on display at the exhibition of the Government of the Federal Republic of Germany organized on the occasion of this conference. The connection with the computer is currently made by punched tape, but a smaller computer will be connected on-line with the spectrometer later on. The entire system is shown in Fig. 1. Figure 2 is a schematic representation of the sample preparation system and its spatial arrangement.

The fully-developed system will operate automatically and be able to perform about four analyses per hour. About thirty minutes after introduction of a sample into the system the analytical result is available. After every fifty analyses the pipette and beaker magazines must be restocked. After this conference, the system will be installed at Karlsruhe and tested with highly radioactive samples under realistic conditions.

The automatic system just described for the determination of uranium and plutonium concentrations by X-ray fluorescence analysis largely fulfils the inspectors' requirements with respect to furnishing tamperproof and objective results, and also the requirements of plant operators for a fast and inexpensive analysis.

2.3. Capability of direct methods for nuclear safeguards

Safeguarding nuclear fuels by accountancy methods requires a clear-cut concept on the part of the inspectors about the capability of the analytical methods to apply. For this reason, the Karlsruhe Nuclear Safeguards Project carried out an extensive analytical intercomparison program in connection with a control experiment at Eurochemic at Mol, Belgium, with the co-operation of many international laboratories [4]. The samples were taken directly from the active feed solutions and the product solutions of the Eurochemic plant. The participating laboratories[1] were asked to carry out the analyses under routine conditions. Only conventional, non-automated methods were employed. The results communicated by the different laboratories were evaluated statistically with respect to the different error components.

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[1] The following organizations participated in a part of or all the interlaboratory tests mentioned: BCMN, EURATOM, Geel, Belgium; CCR EURATOM, Chemistry Division, Ispra, Italy; EUROCHEMIC, Analytical Laboratory, Mol, Belgium; European Institute of Transuranium Elements, EURATOM, Karlsruhe, Germany; IAEA Laboratory, Seibersdorf, Austria; Kernforschungszentrum Karlsruhe, Institut für Radiochemie, Karlsruhe, Germany; Oak Ridge National Laboratory, Oak Ridge, USA; Studiencentrum voor Kernenergie, Mol-Donc, Belgium.

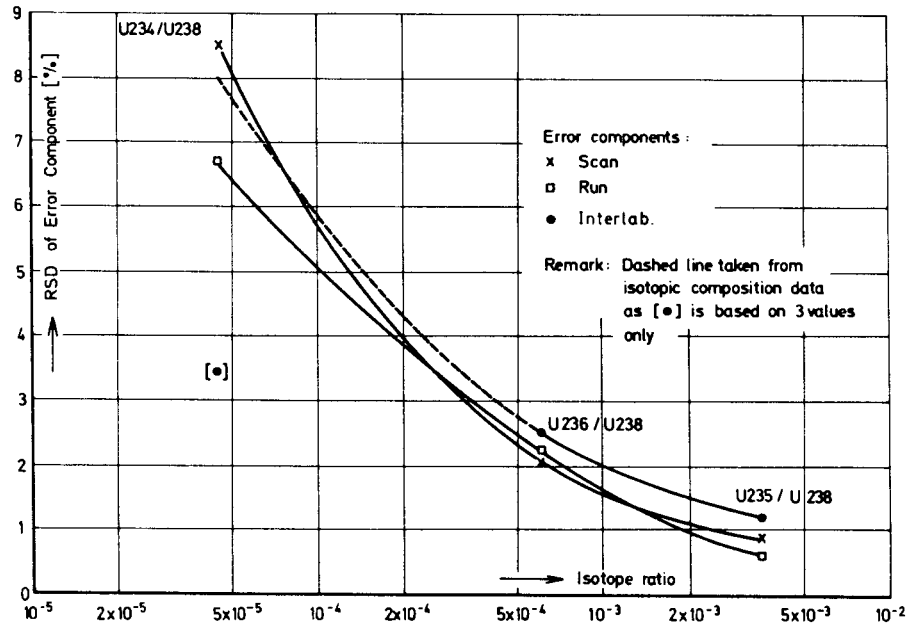

FIG.3. JEX 70 interlab test: determination of isotopic ratios of uranium. RSD of error components.

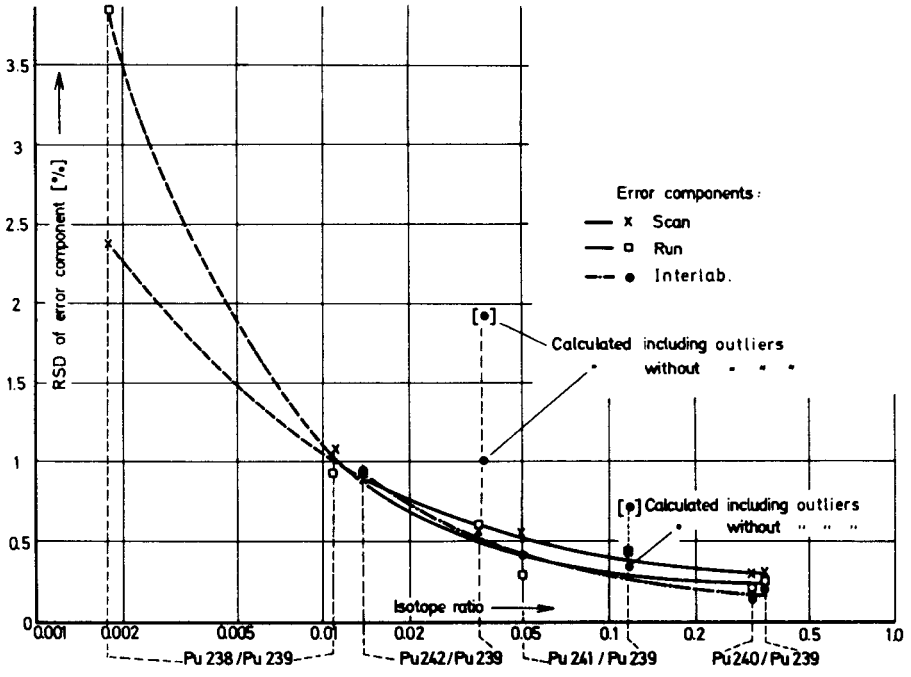

FIG.4. JEX 70 interlab test: mass spectrometric determination of isotopic ratios of plutonium. RSD of error components.

The following analytical comparisons were made:

(1) Assay for uranium and plutonium in product solutions acidified with nitric acid by wet chemical methods and X-ray fluorescence spectrometry.

(2) Determination of uranium and plutonium isotopic compositions by thermionic mass spectrometry and alpha spectrometry (^{238}Pu).

(3) Determination of uranium and plutonium concentrations in active feed solutions by using mass spectrometric isotope dilution analysis.

It turned out that sampling errors were insignificant for both uranium and plutonium in product solutions. The error due to sample preparation in laboratories and sample instability was not significant for uranium, but 0.49% RSD (Relative Standard Deviation) for plutonium. The deviations between laboratories and methods were 0.20% RSD and 0.25% RSD for uranium and plutonium, respectively, and the precision based on results of chemical methods only was 0.11% RSD for uranium and 0.24% RSD for plutonium.

The results of the isotopic analyses of uranium and plutonium are shown in Figs 3 and 4. Since the determination of isotopes present only in very low concentrations is of interest to the Minor Isotope Safeguards Technique (MIST), depleted uranium was used for this test. Moreover, two plutonium samples of different isotopic compositions were analysed. Although the ^{238}Pu concentration was relatively high in one case (about 0.7%), all laboratories preferred the alpha-spectrometric determination of ^{238}Pu.

In comparison with uranium analysis, the isotopic determinations of plutonium showed more outliers. If these are excluded, there is no indication of any significant differences in accuracy in the isotope determination of uranium and plutonium. The curves in Fig. 4 may be taken as direct extensions of the corresponding curves in Fig. 3.

The assays for uranium and plutonium by mass spectrometric isotopic dilution analysis were carried out on samples taken from feed solutions of Eurochemic which had been diluted to 250 times their original volume with $5\underline{N}$ HNO_3 to increase their stability. The plutonium concentrations were 3×10^{-3} mg/ml, the uranium concentrations 1 mg/ml. Since the participating laboratories merely carried out double analyses, no sufficiently secure values for reproducibility could be calculated from this experiment. For interlaboratory deviations, the relative standard deviations were about 2% for uranium and some 3% for plutonium.

2.4. Conclusion

While, until now, nuclear fuels present as solutions or powder have been safeguarded by conventional, non-automated or only partly automated direct methods of chemical analysis requiring the control of the operator's laboratories by an inspection agency or the installation of independent analytical laboratories for that inspection agency, it now appears to be possible in the near future to employ fully automated systems for the direct assessment of nuclear fuels in solutions and powders which requires far less expenditure in terms of control. The capability of the established conventional methods has been determined by interlaboratory tests.

3. NON-DESTRUCTIVE NUCLEAR MATERIAL ASSAY

3.1. Calorimetry

The calorimetric method for determination of plutonium has successfully been used at the fuel element fabrication plant ALKEM in Karlsruhe. In this method the thermal power generated by alpha decay of plutonium isotopes is measured. The accuracy of the result is limited by three factors, namely:

(i) accuracy of the isotopic analysis of the plutonium,
(ii) knowledge of the specific heat production of the individual plutonium isotopes, and
(iii) accuracy of the calorimetric measurement itself.

Inaccuracies in isotopic analysis and apparent inconsistencies of currently adopted values for specific heat production, especially of the isotopes ^{238}Pu and ^{241}Am which make the main contribution to the thermal power, result in an average error of about 0.5% for the first two factors. Absolute heat flux measurements can be made to ± 0.2% if suitably designed calorimeters are used. Thus, an overall accuracy of 0.6% is attainable. Examples are shown in Table I. A more detailed report about some of the results has already been published [5, 13].

However, calorimetry alone cannot be considered sufficiently tamper-proof, and additional determination of ^{240}Pu by neutron coincidence counting is valuable. With 16 BF_3 counters of 40 cm active length, installed in the water bath of a calorimeter for plutonium cans, a total neutron detection probability of about 6%, and average coincidence rates of 6 counts per minute and gram of PuO_2 were obtained. Measuring times in calorimeters, due to the slow adjustment of thermal equilibrium, will be of the order of 500 minutes. This time is sufficient to achieve neutron counting accuracies better than 0.7% even for small sub-assemblies. The system is currently being implemented by a data acquisition and processing system.

TABLE I. EXAMPLES FOR CALORIMETRIC DETERMINATIONS OF PLUTONIUM

Type of fuel shape and size (mm)		Pu per unit (g)	Measurement time (h)	Thermal power (mW)	Accuracy achieved	
					Heat flux (%)	Pu content (%)
UO_2/PuO_2 platelets						
Area	50.68^2	32	1.5	80	± 0.5	± 0.7
Thickness	6.25					
PuO_2 cans						
Diameter	190	1000	6	2500	± 0.5	± 0.7
Height	290					
UO_2/PuO_2 fuel pins						
Diameter	15	300	6	1500	± 0.5	± 0.7
Length	1200					

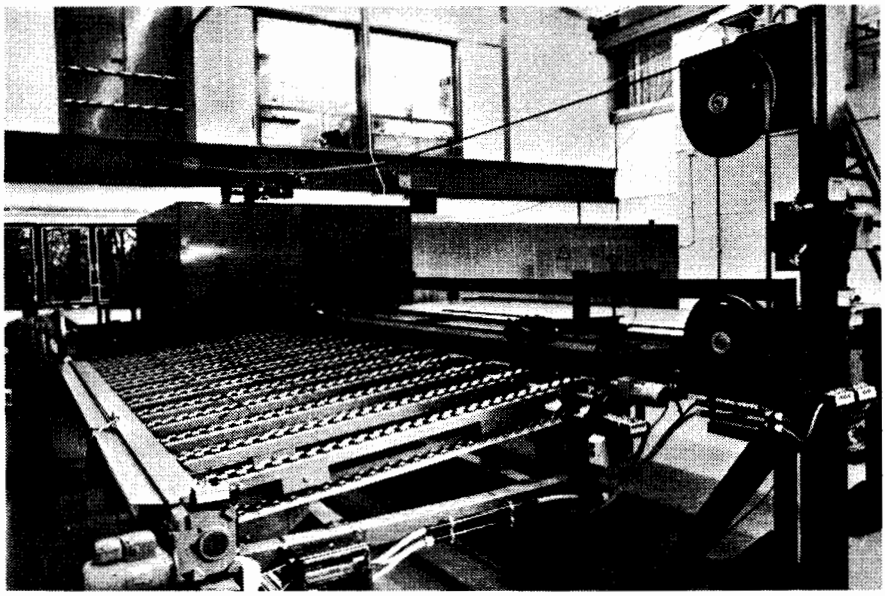

FIG. 5. Slowing-down spectrometer: general view.

A large-scale calorimeter is also being built at ALKEM for fuel elements with an overall length up to 4 m and a cross-section of 215 × 215 mm. This instrument will be the first to use thermocouples of an entirely new design which are characterized by high sensitivity (240 μV/°C per junction) and greatly increased ease of handling.

3.2. Slowing-down spectrometer

Another instrument particularly suited for the assay of fuel pins that contain mixtures of plutonium and ^{235}U is the slowing-down spectrometer [6-10]. An automatic lead pile spectrometer for LWR fuel pins has been built at INTERATOM with the following specifications:

Throughput	600 pins/day
Fuel enrichments	$^{235}U \leq 5\%$
	$^{239}Pu \leq 5\%$
Dimensions of fuel pins	length ≤ 4000 mm
	diameter ≤ 15 mm
Accuracy of measurement	≤ 2%

The most important components of the installation are:

Lead pile with shielding
Neutron generator
Automatic fuel rod transportation and storage machine
Computer system for automatic data acquisition, data processing, and process control
Fuel pin marker.

Figure 5 presents a general view of the assembly. The lead pile contains the necessary channels for insertion of the proton recoil counters and the target tube of the neutron generator. The lead pile is covered with shielding material (boron-loaded) which allows access to the fuel handling devices during operation of the system. A neutron generator with a source strength of about 4×10^{11} n/sec is used. The automatic transfer and storage machine acts as a buffer for arriving fuel pins, inserts pins into the lead pile and, depending on the result of the measurement, divides the pins into two categories which are stored separately. Before discharge the fuel rods are marked with the result of the measurement. The complete process is fully automated.

The slowing-down spectrometer has been in operation since July 1971. Combined with a long duration test, 15 fuel pins of KWO-type (Kraftwerk Obrigheim) were tested repeatedly (length: 2916.7-1.4 mm; diameter: 10.75 ± 0.06 mm; enrichment of ^{235}U: 3.1256%; weight of ^{235}U per pin: 50.129 g). A total weight of 82.56078 kg ^{235}U was measured, and the difference between the actual fuel content and the measured one was only 0.0182 kg, the accuracy thus being about 0.02%. The measuring time for one pin is 93 sec.

3.3. Delayed neutron analyser

At the Institute for Nuclear Engineering, University of Hannover, highly enriched fuel elements are investigated by the delayed neutron technique [11, 12] combined with γ-spectrometry. Special interest is devoted to HTGR pebble elements containing 1 g ^{235}U and 10 g ^{232}Th as coated particles in graphite.

The measuring system (Fig. 6) consists of a Cockcroft-Walton type neutron generator (Accelerator I, 150 kV, 3 mA deuteron current) using

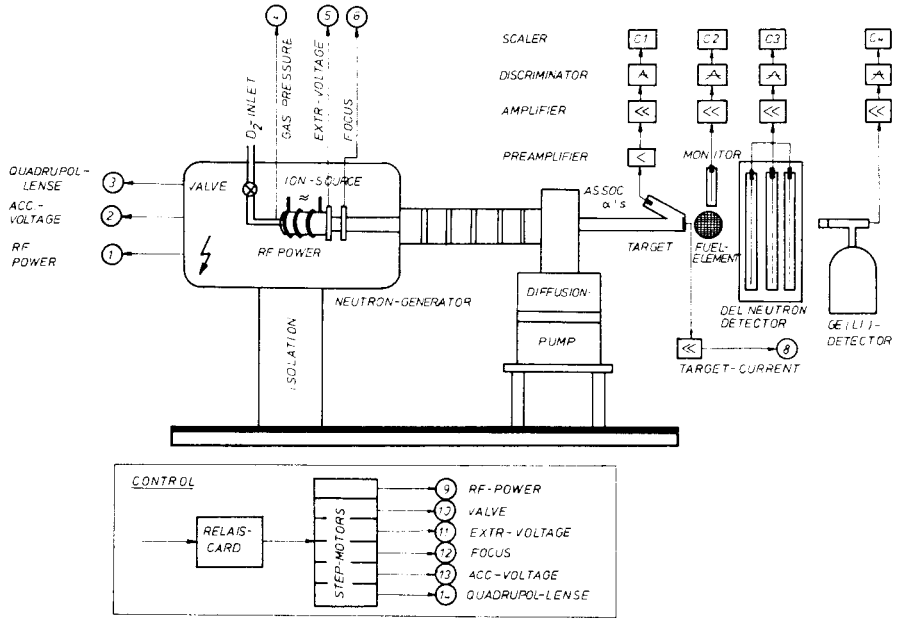

FIG. 6. Schematic diagram of the delayed neutron analyser.

the T(d, n)α reaction, and a neutron detection system. The source strength is determined by measuring the associated α-particles. For pebble elements the changing mechanism allows programmed insertion of a reference element into the pebble path. Measured pebbles are sorted into two containers according to their amount of nuclear material (Cinderella principle). The detection system consists of the delayed neutron detector (fifteen 6-atm ^{3}He-counters imbedded in moderating material, efficiency 18%), a Ge(Li)-detector for spectrometry of γ-rays, and fission chambers for monitoring the neutron flux near the element.

An on-line digital computer is used for data acquisition, data reduction and control. Special hardware features of the system include binary scalers for fast transfer of counts into the computer memory and a computer controlled pulse length generator for setting up neutron pulse length and repetition frequency. The parameter controlled is the neutron source strength. Actual values of deuterium gas flow, r.f. power of the ion source, extraction voltage and acceleration voltage are determined, multiplexed and digitized by a programmable analog-to-digital converter. The desired values are adjusted by step motors driven by relay outputs of the computer. The start-up and shut-down of the neutron generator is also automated.

3.4. Assay by antimony-beryllium neutrons

The assay of whole sub-assemblies requires a highly penetrative radiation. If neutron-induced fissions are used for fissile material detection, the energy of the neutrons should be above the ^{238}U-resonance region [10], but below the fission threshold of ^{238}U (10 keV < E < 500 keV).

Available at relatively high intensities (4×10^6 n/sec. Ci) and acceptable cost, the Sb-Be-source, which produces photoneutrons of 26 keV, seems to be well suited.

Measurements were performed at Karlsruhe which showed the feasibility of energy discrimination between interrogation and fission neutrons.

A 50 cm section of a BWR fuel element was used. The 49 fuel pins (650 g UO_2, 2% enrichment) could be replaced individually by pins of 2.5% enrichment or by lead dummies. First measurements were done with

TABLE II. FISSILE MATERIAL ASSAY WITH AN Sb-Be SOURCE ON A BWR FUEL-ELEMENT MOCK-UP

Number of pins			Geometry	Normalized counting rate	Mass of ^{235}U in g	
Pu	^{235}U					
	2%	2.5%			Real	Determined
5	23	21	2.5% central	1204 ± 4	565.3	566.6 ± 4.0
5	23	21	2.5% surface	1201 ± 5	565.3	565.4 ± 4.0
5	23	21	homogeneous	1204 ± 5	565.3	566.6 ± 4.0
0	49	0	homogeneous	1200 ± 5	564.85	standard
6	23	20	homogeneous	1167 ± 5	551.0	549.3 ± 4.0

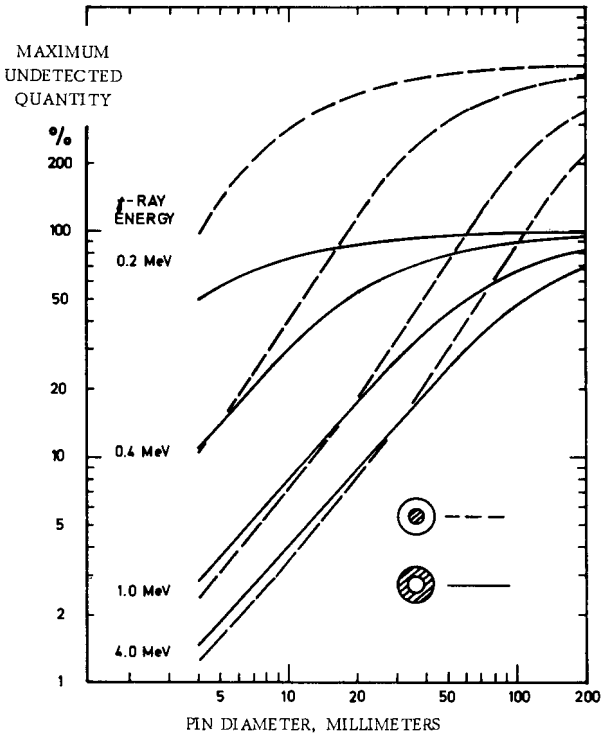

FIG.7. Quantity of material that can be removed from, or added to, a fuel pin (or a cylindrical fuel arrangement) without detection if γ-rays of different energies are used. Composition of the homogeneous pin is 15% PuO_2 with 85% UO_2, of density 10.35 g/cm^3, and local γ-source strength is assumed to be proportional to Pu concentration.
Solid lines: Pu concentrated at the periphery, broken lines: Pu concentrated in the centre. Quantities are given in per cent of Pu in the homogeneous pin.

3 proton recoil proportional counters (4.5 cm diameter, 3 atm methane) which were protected from the γ-radiation by a 15 cm lead shield surrounding the 35 Ci source. With the threshold set to 600 keV (thus discriminating higher energy source neutrons [14]) and a standard composition of the sub-assembly (564.9 g ^{235}U) a counting rate of 10 counts/sec was observed.

The influence of self-shielding was studied with pins of higher enrichment (2.5%) placed in the centre or at the surface of the sub-assembly, respectively. No difference in counting rate was observed (see Table II). The replacement of one pin by a lead dummy was clearly recognized.

Thus the method looks feasible for ^{235}U assay. In the case of plutonium (25% ^{240}Pu), a source of 230 Ci is required to get the induced fission rate equal to the spontaneous fission rate. For mixtures a second method must be used for separate assay of ^{235}U and plutonium [5].

3.5. Assay by neutron capture γ-rays

Most existing non-destructive methods measure a linear combination of the contents of different isotopes of uranium and plutonium and allow, at

best, the determination of ^{235}U and the sum of the fissionable plutonium isotopes. A new active method for the separate assay of all isotopes of interest is therefore being investigated in Karlsruhe. This method is based upon the measurement of γ-ray spectra following neutron capture. If γ-rays from neutron interaction with fissionable material are to be used for assay purposes, the knowledge of the nature of the individual γ-lines is very important. High-energy γ-rays due to neutron-induced fission do not allow quantitative analysis unless only one fissionable isotope is present. Therefore, the high-energy γ-ray spectra from neutron interaction with ^{235}U, ^{239}Pu and ^{241}Pu have been carefully examined to separate the capture from the fission components. Several capture lines were found with energies and intensities well suited for assay purposes; other γ-rays formerly ascribed to neutron capture could be shown to be due to fission events. Some results and a brief description of the measurements are given in Ref. [15].

Primary capture γ-rays are known to be of sufficiently high energy to ensure good transparence of the samples. Thus bundles of pins or small sub-assemblies can be measured. Calculations were made to investigate the influence of radial inhomogeneities of cylindrical fuel arrangements. The result, expressed in terms of the maximum amount of fissile material that can be added to (or removed from) the rod without detecting is plotted in Fig. 7 as a function of cylinder diameter.

REFERENCES

[1] KOCH, L., Radiochimica Acta 12 (1969) 160.

[2] WILHELMI, M., BEYRICH, W., GERIN, F., ROMKOSKI, M., TASMAN, H.A., Safeguards Techniques (Proc. Symp. Karlsruhe, 1970) 2, IAEA, Vienna (1970) 165.

[3] von BAECKMANN, A., Safeguards Techniques (Proc. Symp. Karlsruhe, 1970) 2, IAEA, Vienna (1970) 45.

[4] KRAEMER, R., et al., Vol. II Report on the JEX-70 Safeguard Experiment, to be published Rep. KFK (1971).

[5] SCHNEIDER, V.W., HILLE, F., KIY, M., GMELIN, W., Safeguards Techniques (Proc. Symp. Karlsruhe, 1970) 1, IAEA, Vienna (1970) 181.

[6] EINFELD, K., U.S. Patent No. 3.222.521, dated 17.10.59, Canadian Patent No. 666.943, dated 17.10.59, DAS No. 1.275.794.

[7] KRINNINGER, H., WIESNER, S., FABER, C., Nucl. Instrum. Meth. 73 (1969) 13.

[8] STEGEMANN, D., SEUFERT, H., Trans. Am. nucl. Soc. 11 2 (1968) 658.

[9] RUPPERT, E., KRINNINGER, H., "Die Anwendung einer Bremszeitspektrometeranordnung zur zerstörungsfreien Bestimmung des Spaltstoffgehaltes", Reaktortagung des Deutschen Atomforums, Berlin, April 1970.

[10] BAUMUNG, K., BÖHNEL, K., KLUNKER, J., KÜCHLE, M., WOLFF, J., Safeguards Techniques (Proc. Symp. Karlsruhe, 1970) 2, IAEA, Vienna (1970) 177.

[11] KEEPIN, G.R., et al., Safeguards Techniques (Proc. Symp. Karlsruhe, 1970) 2, IAEA, Vienna (1970) 79.

[12] AUGUSTSON, R.H., et al., Safeguards Techniques (Proc. Symp. Karlsruhe, 1970) 2, IAEA, Vienna (1970) 53.

[13] GMELIN, W., NENTWICH, B., OTTO, H.E., Rep. KFK 910 (1969).

[14] LALOVIC, M., WERLE, H., J. nucl. Energy 24 (1970) 123.

[15] MATUSSEK, P., MICHAELIS, W., WEITKAMP, C., WODA, H., Safeguards Techniques (Proc. Symp. Karlsruhe, 1970) II, IAEA, Vienna (1970) 113.

ASPECTS DU CONTROLE D'INSTALLATIONS REPRESENTATIVES DE L'INDUSTRIE

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Abstract–Résumé–Аннотация–Resumen

ASPECTS OF THE CONTROL OF REPRESENTATIVE INDUSTRIAL PLANTS.

The cases chosen for study relate to (1) the part of the (90%) enriched uranium fuel cycle located on Belgian territory, which unites the following three facilities on the basis of a yearly output of 50 $kg_{eff.}$: the MMN fuel fabrication plant, the BR2 test reactor, and the Eurochemic reprocessing plant; (2) the uranium fuel fabrication line for light-water reactors (LWR) which has been installed at the MMN plant and has an output of 250 $kg_{eff.}$; (3) the industrial-scale plutonium fuel fabrication plant belonging to Belgonucléaire, at present under construction. The same general principles have been applied to these cases in order to arrive at the following parameters: W = inventory in $kg_{eff.}$, and τ = mean residence time for the material in the facility. The choice of strategic points and measuring techniques has been made with a view to improving the cost/efficiency ratio. Non-destructive tests prove necessary, however, since most of the cases relate to the measurement of finished products; it is this type of control that is mainly covered by the paper. Analysis of the BR3 fuel cycle and the MMN uranium fuel fabrication line is based on the formalism of sampling functions and the graphical method, these being particularly useful in estimating such control parameters as the amount of material under safeguards at a strategic point at a given moment. The non-destructive tests applied are gamma spectrometry and thermal neutron interrogation. They are performed in a controlled beam geometry and the time variable has been intrinsically introduced into the measurements so as to incorporate them into the mathematical model proposed. Within the context of the Belgonucléaire plutonium fuel fabrication plant the operator describes the scope of the work he performs in controlling the fissionable and fertile material.

ASPECTS DU CONTROLE D' INSTALLATIONS REPRESENTATIVES DE L' INDUSTRIE.

Les cas choisis pour l' étude concernent: 1° la partie du cycle de combustible à uranium enrichi (90%) qui se situe sur le territoire belge et qui couple par un débit annuel de 50 kg_{eff} les trois installations: usine de fabrication MMN, réacteur d' essai BR2 et usine de retraitement Eurochemic; 2° la chaîne de fabrication de combustible uranifère pour réacteur du type à eau légère (LWR), installée à l' usine MMN, caractérisée par un débit de 250 kg_{eff}; 3° l' usine de fabrication de combustible plutonifère de taille industrielle de la Belgonucléaire, actuellement en construction. Les mêmes principes généraux ont été appliqués à ces cas pour déduire les grandeurs caractéristiques: W, inventaire en kg_{eff}, et τ, temps de séjour moyen de la matière dans l' installation. Le choix des points stratégiques et des méthodes de mesure a été fait en visant à améliorer le rapport coût/efficacité. Les tests non destructifs s' imposent toutefois puisqu' il s' agit dans la plupart des cas de mesurer des produits finis et c' est sur ce type de contrôle que porte l' essentiel du mémoire. L' analyse du cycle de combustible du réacteur BR3 et de la chaîne de fabrication de combustible uranifère de l' usine MMN utilise le formalisme des fonctions échantillonnées et la méthode des graphes qui se prêtent particulièrement bien à l' estimation des paramètres de contrôle comme la quantité de matières soumises aux garanties en un point stratégique à un moment donné. Les tests non destructifs utilisés sont la spectrométrie gamma et l' interrogation par faisceau de neutrons thermiques. Ils sont effectués en géométrie contrôlée et la variable du temps a été introduite de façon intrinsèque dans les mesures afin qu' elles

s' intègrent dans le modèle mathématique proposé. Dans le cadre de l' usine de fabrication de combustible plutonifère de la Belgonucléaire, l' exploitant décrit le cadre de ses travaux concernant le contrôle des matières fissiles et fertiles.

АСПЕКТЫ КОНТРОЛЯ РЕПРЕЗЕНТАТИВНЫХ ПРОМЫШЛЕННЫХ УСТАНОВОК.

Для данного исследования были выбраны следующие случаи: 1) секция топливного цикла с обогащенным ураном (обогащение 90%), которая находится на бельгийской территории, с годовой производительностью 50 $кг_{эфф}$, и которая соединяет следующие три установки: завод по изготовлению тепловыделяющих элементов фирмы "Металлуржи э меканик нуклеэр", испытательный реактор BR-2 и завод по переработке топлива "Еврокемик"; 2) линия по изготовлению тепловыделяющих элементов из урана для реакторов с легководным теплоносителем установлена на заводе фирмы "Металлуржи э меканик нуклеэр" и имеет производительность 250 $кг_{эфф}$; 3) установка промышленного масштаба для изготовления плутониевого топлива фирмы "Бельгонюклеэр" в настоящее время находится в стадии строительства. Одинаковые общие принципы были применены к этим случаям для выведения характеристических параметров: W – наличное количество, в $кг_{эфф}$, и τ – среднее время перемещения материала через установку. Выбор стратегических точек и методов измерения был сделан с целью улучшения отношения стоимость/эффективность. Тем не менее, использование неструктивных методов испытания необходимо, поскольку большинство случаев связано с измерением готовых продуктов, а в докладе освещается, в основном, этот тип контроля. При анализе топливного цикла реактора BR-3 и линии по изготовлению тепловыделяющих элементов из уранового топлива используются формализм z – функций отбора проб и графические методы, которые особенно подходят для оценки таких контрольных параметров, как количество материала, находящегося под контролем в соответствии с системой гарантий в одной стратегической точке в данный момент. К использованным недеструктивным методам испытаний относятся гамма-спектрометрия и пучки тепловых нейтронов. Они применяются в контролируемой геометрии, а временная переменная была введена в измерения, с тем чтобы включить ее в предложенную математическую модель. В рамках установки для изготовления плутониевого топлива фирмы "Бельгонюклеэр" исследователь дает описание своей работы, касающейся контроля за расщепляющимися и воспроизводящими материалами.

ASPECTOS DEL CONTROL DE INSTALACIONES INDUSTRIALES REPRESENTATIVAS.

Los casos elegidos para el presente estudio se refieren a: 1) la sección del ciclo del combustible de uranio enriquecido (90%) situada en territorio belga, cuya producción anual es de 50 kg efectivos y que consta de la instalación de fabricación MMN, el reactor de ensayo BR2 y la instalación de reelaboración de la Eurochemic; 2) la línea de fabricación de combustible de uranio para el reactor de agua ligera (LWR) instalado en la planta MMN, cuya producción es de 250 kg efectivos; 3) la fábrica de combustible de plutonio a escala industrial de la Belgonucléaire, que se está construyendo actualmente. En todos estos casos se han aplicado los mismos principios generales para deducir los parámetros característicos W, dotación de combustible en kilogramos efectivos y tiempo medio de tránsito del material a través de la instalación τ. Al elegir los puntos estratégicos y los métodos de medida se ha procurado mejorar la relación costo/eficacia. Sin embargo, es necesario efectuar ensayos no destructivos, puesto que en la mayor parte de los casos se trata de ensayar productos terminados, y esta memoria se ocupa principalmente de este tipo de ensayos. El análisis del ciclo del combustible en el reactor BR3 y de la línea de fabricación de combustible de uranio de la planta MMN se hace utilizando las funciones de muestreo y aquellos métodos gráficos que son particularmente apropiados para estimar los parámetros de control, tales como la cantidad de materiales sometidos a salvaguardias en un punto estratégico en un momento determinado. Los ensayos no destructivos aplicados consisten en la espectrometría gamma y el examen mediante haces de neutrones térmicos. Se llevan a cabo en una geometría controlada y se ha introducido intrínsecamente en las medidas la variable tiempo, de forma que intervengan en el modelo matemático propuesto. Ciñéndose a la fábrica de combustible de plutonio de la Belgonucléaire, el explotador describe el ámbito de sus trabajos relativos al control de los materiales fisionables y fértiles.

I. INTRODUCTION

Compte tenu de l'existence du Traité sur la non-prolifération des armes nucléaires et de l'importance éventuelle de son incidence en Belgique, des principes généraux de réalisation du contrôle des garanties ont été établis pour les niveaux national et de l'exploitation en fonction d'installations représentatives de l'industrie belge. [1]

(i) Généralités

Dans le préambule du Traité, les Etats Parties au Traité ont exprimé notamment :
"leur appui aux efforts de recherche, de mise au point et autres visant à favoriser l'application, dans le cadre du système des garanties de l'Agence internationale de l'énergie atomique, du principe d'une garantie efficace du flux des matières brutes et des produits fissiles spéciaux, grâce à l'emploi d'instruments et autres moyens techniques en certains points stratégiques." Ces efforts doivent finalement conduire à l'établissement d'un système de contrôle quantifié qui minimisera la discrimination économique entre les Etats dotés et non dotés d'armes nucléaires et qui respectera au maximum la propriété industrielle.

Les actions entreprises en Belgique s'inscrivent dans le cadre de cet appui témoigné par l'Etat. On s'est référé à la définition de l'objectif des garanties qui est "de déceler rapidement le détournement de quantités significatives des matières nucléaires des activités nucléaires pacifiques vers la fabrication d'armes nucléaires ou autres dispositifs nucléaires explosifs ou à des fins inconnues et de dissuader tout détournement par le risque d'une détection rapide" [2]. On a tenu compte de l'existence du système de contrôle des matières soumises aux garanties reconnu par l'Etat, système conforme au chapitre VII du Traité instituant la Communauté Européenne de l'Energie Atomique [3]. "L'Agence doit pouvoir vérifier les résultats obtenus par le système national en faisant usage de la comptabilité matières comme mesure d'importance essentielle, associée au confinement et à la surveillance comme mesures complémentaires importantes" [2]. L'Agence aussi bien que l'Etat baseront leur contrôle sur la comptabilité matières de l'Exploitant comme mesure d'importance essentielle.

(ii) Indice d'importance

Le maximum fixé pour les activités régulières d'inspection de l'Agence a été choisi comme indice de l'importance du contrôle des garanties appliqué à l'Etat dans le cadre du Traité. Cet indice, exprimé en journées d'inspecteur par an, a été estimé pour les réacteurs et les usines de fabrication qui constituent les installations les plus représentatives du cycle de combustible installé en Belgique.

Pour les réacteurs, les valeurs de l'indice pour 1975, 1980 et 1985 sont 300, 450 et 600 journées d'inspecteur par an. En les exprimant en fonction de la puissance électrique installée ces valeurs deviennent respectivement 0,18, 0,16 et 0,11 journée d'inspecteur par an et par MWe installé.

L'estimation de l'indice pour la fabrication du combustible uranifère faiblement enrichi ne peut être faite de façon sûre que jusqu'en 1975. On a admis pour l'extrapolation une croissance proportionnelle à la puissance installée dans le pays. Etant donné l'importance du terme constant dans la relation fixant le maximum autorisé dans cette catégorie, cette hypothèse paraît acceptable. Les valeurs obtenues sont 130, 150 et 190 jours d'inspecteur par an pour 1975, 1980 et 1985.

En ce qui concerne la fabrication d'uranium fortement enrichi, le maximum autorisé ne pouvant pas être inférieur à 450 jours d'inspecteur par an, l'indice n'est pas affecté par le débit de l'installation et reste égal à cette valeur.

Pour la recherche et le développement de même que pour la fabrication de combustibles enrichis au plutonium, l'indice est affecté par le débit de l'installation dès 1973. Les valeurs sont estimées à 670, 950 et 1350 jours d'inspecteur par an pour 1973, 1975 et 1980.

(iii) Définition des mesures de vérification

Les mesures de vérification sont appelées à résoudre des litiges concernant les déclarations pour chaque zone de bilan-matières, indiquant la différence d'inventaire pour une période déterminée et les limites d'exactitude des différences déclarées.

La crédibilité de ces mesures doit être rigoureusement établie. On a tenté d'atteindre cette crédibilité par la simplicité des mesures appliquées, par exemple des pesées, des comptages dans des conditions expérimentales facilement reproductibles etc.

Comme ces mesures doivent également dissuader l'entreprise de tout détournement par le risque d'une détection rapide, on a choisi d'utiliser de préférence les méthodes non destructives en les confirmant le cas échéant par des mesures destructives.

(iv) Contraintes

On a reconnu que la contrainte déterminante qui affecte le système des garanties en est le coût [4]. On a choisi comme indice de cette contrainte le rapport du coût des garanties au coût d'exploitation dans chaque zone de bilan-matières. Les procédures d'application des mesures envisagées dans une zone de bilan-matières sont classées selon cet indice auquel on a fixé une limite de 1 %. Au-delà de cette limite, les procédures envisagées devraient être rejetées du fait de cette contrainte.

On a considéré comme autre contrainte la possibilité de définir dans une installation une zone de bilan-matières spéciale qui inclurait dans ses limites un procédé dont les détails sont névralgiques du point de vue commercial, tout en respectant l'objectif des garanties [2]. Cette contrainte sera notamment introduite dans le cas de l'usine de fabrication de combustible plutonifère où l'on tiendra compte des mesures complémentaires de confinement et de surveillance.

(v) Choix des installations étudiées

On s'est attaché à l'étude des mesures de contrôle en vue de leur application dans les réacteurs et les usines de fabrication. Pour favoriser la diffusion des résultats et éviter des duplications, cette étude a été coordonnée dans le cadre de l'Association liant Euratom - G.f.K. - C.E.N. - C.N.E.N. et R.C.N. [5].

On a choisi d'étudier des cas précis.
Le premier se rapporte à une partie du cycle de combustible du réacteur BR2 (type MTR, uranium enrichi à 90 %) comprise entre la réception des plaquettes à l'usine Métallurgie et Mécanique Nucléaires (M.M.N.) et le dissolveur de l'usine de retraitement Eurochemic.

Le second cas, dont l'analyse n'est encore qu'au stade préliminaire, se rapporte à une campagne de fabrication, à l'usine M.M.N., de combustible faiblement enrichi pour la charge d'un réacteur de puissance de 390 MWe du type P.W.R.

Le troisième cas se rapporte à une installation de combustible plutonifère que construit actuellement Belgonucléaire.

(vi) Technique d'analyse - fonctions échantillonnées

Dans un mémoire précédent [6] se rapportant au premier et au troisième cas, l'estimation de l'efficacité des mesures dans les installations a été effectuée en se fondant sur les paramètres du contrôle : W inventaire total exprimé en kg $_{eff}$, T_c temps critique [7], et $\langle\tau\rangle$ temps de séjour moyen de la matière dans l'installation.

Les installations ont été divisées en régions de deux types, le premier type est celui où les matières sont accessibles et mesurables, le second type est celui où les matières sont en traitement.

Pour chacune des installations, on a établi un diagramme du mouvement régulier des matières fissiles en cours d'exploitation qui a conduit aux résultats présentés au tableau I.

TABLEAU I. PARAMETRES DU CONTROLE DES INSTALLATIONS

Paramètre	M.M.N.	BR2	BN
$\langle W \rangle$ (kg_{eff})	7,5	49,5	500
$\langle \tau \rangle$ dans l'installation	8,5 sem.	62 sem.	20 sem.
$\langle \tau \rangle$ en traitement	4,25 sem.	37 sem.	1,5 sem.
T_c	2 sem.	2 sem./14 sem.	2 sem.

L'influence de la relation entre $\langle \tau \rangle$ et T_c dans une zone de bilan-matières peut être mise en évidence en considérant les cas limites : $\langle \tau \rangle / T_c \to 0$ ou $\langle \tau \rangle / T_c \to \infty$, le contrôle est respectivement externe ou interne à l'installation.

Ce rapport $\langle \tau \rangle / T_c$ est important pour les zones de traitement où les matières sont inaccessibles et pour lesquelles les mesures de contrôle devraient éventuellement s'appuyer sur le confinement ou la surveillance.

On a introduit un formalisme mathématique qui tient compte de ce qu'en pratique le contrôle d'une installation s'effectue de façon discontinue, le plus souvent à des intervalles de temps réguliers. Il en résulte que les fonctions continues $S_i(t)$ et $X_i(t)$ représentant les stocks et les quantités en mouvement dans les différentes zones de bilan-matières ne sont connues qu'à certains moments bien déterminés, par exemple à $t = 0, T, 2T$, etc., T étant l'intervalle de temps qui sépare deux relevés consécutifs. Le contrôle ne peut porter que sur ces informations discrètes, c'est-à-dire sur des fonctions dites "échantillonnées".

On appelle fonction "échantillonnée" une fonction constituée par un train d'impulsions de la forme :

$$X^*(t) = \sum_r X(rT) \cdot \delta(t - rT)$$

où $X(t)$ est une fonction continue de t et T est l'intervalle de temps, supposé constant, entre deux échantillonnages.
Grâce à ses propriétés particulières, on a pu représenter très fidèlement les mouvements et les inventaires des matières dans une installation en faisant appel à la méthode des graphes [8]. L'analyse de système s'est ainsi développée dans cette voie, tandis qu'un examen détaillé de l'application des statistiques sur la prise des inventaires a été présenté en [9].

La suite de ce mémoire se limite à la description des résultats ou des conceptions retenues dans les trois cas précis étudiés.

II. CYCLE DE COMBUSTIBLE DU REACTEUR BR2 (type MTR, uranium enrichi à 90 %)

L'intérêt de l'étude de ce cycle est dû à la valeur stratégique élevée du combustible. Cette étude est facilitée par la proximité des installations principales : usine de fabrication, réacteur et usine de retraitement. Aussi, elle a été choisie comme premier domaine d'application du système proposé en [5]. Son développement actuel fait l'objet d'un contrat avec l'Agence Internationale de l'Energie Atomique.

Huit points clefs ont été retenus pour les mesures. Ce sont respectivement les stockages des plaquettes, des plaques laminées et des éléments frais à l'usine de fabrication ; les stockages des éléments frais, des éléments irradiés et des éléments avant envoi à l'usine de retraitement au réacteur BR2, et pour terminer le stockage des éléments et le dissolveur de l'usine de retraitement.

Pour chacun de ces points les objectifs sont :
1. les essais comparatifs des dispositifs expérimentaux
2. l'établissement d'une librarie des données acquises en spectrométrie gamma
3. le traitement des données sur place en utilisant un ordinateur compact
4. l'évolution du coût en fonction de l'exactitude et de la précision.

Pour indiquer la voie suivie, on présente des résultats relatifs au stockage des plaquettes, des éléments frais et des éléments irradiés.

(i) Description des entités à mesurer

Les éléments de combustible ont une structure cylindrique à trois, à cinq ou à six couches coaxiales. La longueur de la zone active est de 74,9 cm.
La composition de l'alliage UAl_4 + Al est la suivante :
- 24 % en poids d'uranium enrichi à 90 %
- 76 % en poids d'aluminium.

Les éléments à six couches (18 plaques laminées) et à cinq couches (15 plaques laminées) utilisant ce combustible contiennent respectivement : 244 ± 5 g et 223 ± 5 g d'uranium-235.

La densité superficielle de l'uranium-235 dans les plaques laminées est de 36,8 mg/cm^2.
Les tolérances de fabrication permettent des déviations locales de la densité superficielle pour autant que la densité moyenne observée sur 1 cm^2 ne dévie pas de plus de 20 % de la valeur nominale.

Les éléments à six couches utilisant le combustible cermet contiennent :
330 ± 5 g d'uranium-235
2,8 g de bore naturel sous forme de carbure B_4C
1,32 g de samarium naturel sous forme d'oxyde Sm_2O_3.

La densité superficielle en uranium-235 est de 49,7 mg/cm^2.
Les tolérances relatives sont les mêmes que pour les alliages.
Les plaquettes avant laminage ont une longueur de 10,5 cm, leur largeur est fonction du rayon des couches coaxiales.

(ii) Stockage des plaquettes avant laminage

Les spécifications de fabrication sont telles que les déclarations de poids, de dimensions, de teneurs en uranium et des enrichissements des plaquettes d'un même format sont très proches, pratiquement identiques. La procédure de vérification est fondée sur cette observation : pour les mesures les plaquettes sont groupées par dizaine, les résultats des groupes sont ensuite comparés.

On a en outre conçu la mesure de telle sorte qu'elle reflète le symbolisme mathématique des fonctions échantillonnées, en introduisant une composante temporelle. Elle s'appuie ainsi sur le modèle fondamental préconisé pour la vérification de l'installation.

Le groupe est d'abord pesé. Les plaquettes sont ensuite posées dans les cavités d'un disque support (fig. 1). Les tolérances de ces cavités sont légèrement supérieures aux tolérances de fabrication. Le disque porte un ensemble de sources isotopiques.

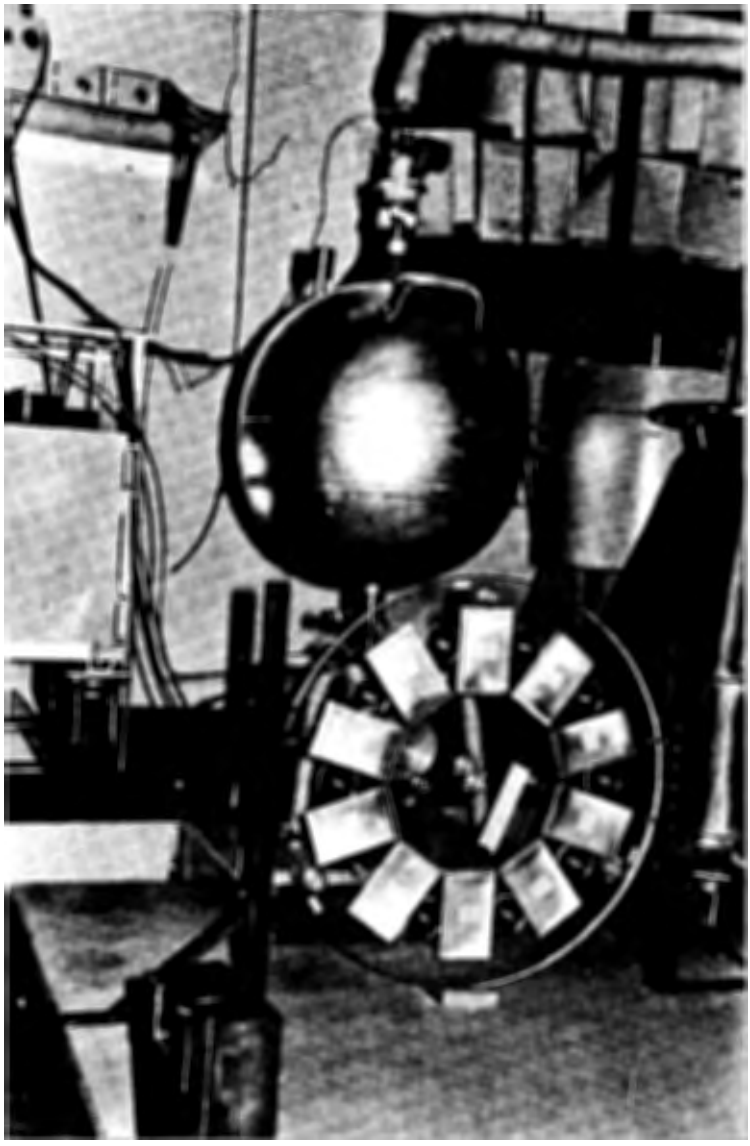


FIG. 1. Mesure autocontrôlée des plaquettes avant laminage.

On a choisi pour la source centrale le Na-22 ($E\gamma$ = 511 keV) et pour les trois sources périphériques le Cs-137 ($E\gamma$ = 661,6 keV).
Les énergies spécifiques de ces sources sont bien séparées du rayonnement des plaquettes ($E\gamma_{max}$ = 205 keV). Ces sources permettent le contrôle de la géométrie et de la durée effective du comptage.
On a ajouté une source Hg-203 ($E\gamma$ = 279,1 keV), dont la période de 46,9 jours adaptée à la durée de la campagne de fabrication permet de vérifier le moment de l'étalonnage.
Le rayonnement gamma de l'ensemble est mesuré au moyen d'un détecteur Ge(Li).

Les résultats caractéristiques d'une série de mesures sont résumés ci-dessous.
Dans cette série, sept groupes de dix plaquettes (dont la teneur moyenne déclarée était de 264 g d'uranium-235) ont été mesurés durant 300 s, avec un temps mort de 14 %.
Les résultats pour le pic de 185 keV ont été normalisés à l'unité, l'écart-type relatif calculé de ces sept mesures était égal à 0,005, le comptage moyen sous le pic après soustraction linéaire du fond parasite et du fond Compton était égal à 615.034.
En outre, une plaquette de teneur déclarée 26,40 g d'uranium-235 a été mesurée successivement dans les 10 positions avec un temps de comptage de 300 s et un temps mort de 3 %.
L'écart-type relatif calculé de ces mesures est égal à 0,01, le total des coups pour les dix mesures s'élève à 657.954. On constate que ce résultat est significativement différent de celui des mesures par groupe, alors qu'ils correspondent à une déclaration égale de la teneur en uranium-235. Le rapport des deux mesures est

$$0{,}935 \pm 0{,}015 \quad (1\ \sigma).$$

Les mesures ont été vérifiées par le comptage des sources de Cs-137, le rapport des mesures étant

$$0{,}926 \pm 0{,}025 \quad (1\ \sigma).$$

La différence significative peut donc être attribuée à une correction insuffisante du temps mort par l'analyseur. Ceci démontre l'utilité de la procédure de vérification introduite par les sources isotopiques.
Le comptage du pic de l'isotope Hg-203, dont la décroissance est caractérisée par une période de 46,9 jours, comparé au comptage du pic du Cs-137 permet un datage suffisant, vu le débit lent de la fabrication.

L'indice de contrainte du coût est de l'ordre du dollar par entité en ce point. Le débit est de 4 000 entités par année, le facteur d'utilisation est largement inférieur à 100 heures par année. Il est clair que tant pour l'amortissement que pour le rendement, le même appareillage doit être utilisé en d'autres points-clefs ou pour d'autres chaînes de fabrication.
Si on se limite à cette chaîne, la pesée et la mesure au détecteur NaI(Tl) paraissent les seules procédures à envisager avec sources isotopiques adéquates pour conserver la vérification des mesures et le datage.

(iii) Stockage des éléments frais

a) Méthode passive

Pour éprouver le principe de la méthode proposée, des mesures ont été effectuées sur un élément de combustible UO_2 d'enrichissement égal à 4 %. Cette expérience est décrite plus loin dans le paragraphe ayant trait au combustible P.W.R.

La sensibilité de l'essai passif appliqué à un élément de combustible de BR2 est telle qu'une perturbation de 3 % de la masse déclarée est mesurée par une variation du taux de comptage γ de 1,5 %.

b) Méthode active [10]

L'élément combustible à contrôler est placé perpendiculairement à un faisceau de neutrons thermiques de BR2. La fente de collimation est parallèle à l'axe de l'élément. Un compteur BF_3 mesure les neutrons transmis et un second, noyé dans la paraffine, mesure les neutrons de fission induits dans l'élément. Pour éliminer les neutrons thermiques diffusés, la paraffine est entourée de cadmium.

On peut déterminer la teneur de l'élément en uranium-235 à partir des deux paramètres mesurés, des rapports des taux de comptage

$\frac{F}{T_o}$ et $\frac{T}{T_o}$, et d'une constante d'étalonnage K.

F est le taux de comptage relevé au compteur de fission,
T_o le taux de comptage au compteur de transmission en l'absence de l'élément et
T le même taux en présence de l'élément.

Si M est la masse d'uranium-235 dans l'élément mesuré, on a :

$$M = K \cdot C \cdot \frac{F}{T_o} \cdot \frac{\left|\ln \frac{T}{T_o}\right|}{1 - \frac{T}{T_o}}$$

La constante K doit être déterminée par comparaison avec un élément de référence dont la teneur est supposée exacte. Lorsque l'élément mesuré n'a pas le même nombre de couches que l'élément de référence, sa surface active moyenne par plaque est différente et il y a lieu d'appliquer le facteur correctif C. L'intérêt de la méthode réside notamment dans le fait qu'elle ne fait pas intervenir explicitement la composition des plaques de combustible, ce qui est d'une importance particulière dans le cas du réacteur BR2, qui utilise des combustibles de plusieurs types.

En outre, en interrogeant les trois secteurs, par rotation de 120°, l'efficacité de l'interrogation varie peu pour les couches interne et externe ; on trouve respectivement une variation de 10 % et de 20 % pour les éléments à 6 couches à alliage et à cermet.
Du fait de la géométrie des éléments de combustible BR2, on a effectué les mesures dans trois orientations obtenues par rotation de 120°.

Le flux de neutrons du faisceau était de l'ordre de 2.10^5 n/cm^2.s, la fente de collimation de 2 cm × 0,5 cm et le temps de comptage de 1 à 5 minutes.

Les mesures ont été effectuées sur neuf points compris entre + 20 et - 20 cm, on dispose ainsi d'un ensemble de 27 mesures par élément examiné.
Ces 27 mesures de $\frac{F}{T_o}$ et $\frac{T}{T_o}$ présentent une certaine distribution, due à deux composantes : la dispersion propre des taux de comptage et la dispersion de la densité superficielle du combustible. On a constaté que la seconde composante l'emportait sur la première, compte tenu du nombre de coups mesuré.

Les résultats d'une série de mesures sont résumés ci-dessous

a) Constante d'étalonnage

L'élément de référence choisi est le M 545, de type V (cinq couches), dont la distribution des mesures $\frac{F}{T_o}$ et $\frac{T}{T_o}$ présentait le moins de dispersion. Sa teneur déclarée est de 219,14 g d'uranium-235.

La distribution des valeurs calculées de K au 27 points de mesure était normale et toutes les valeurs ont pu être retenues.

Il en est résulté :

$$\overline{K} = 3065$$

et $\Delta K_1 = \pm 10$ si M est exprimé en grammes

L'erreur de mesure ΔK_1, composée quadratiquement avec l'erreur de surface, estimée à partir des tolérances de fabrication donne finalement :

$$K = 3065 \pm 32$$

b) Masses en uranium-235

Les expériences ont porté sur quatre éléments de combustible représentatifs des différents types existants soit pour le combustible alliage trois, cinq et six couches et pour le combustible cermet six couches.

Le tableau II rassemble les résultats des mesures, les incertitudes ont été calculées à un intervalle de confiance de 95 %.

TABLEAU II. COMPARAISON DES MASSES DECLAREES ET DES MASSES MESUREES

Nombre de couches	Masse déclarée	Masse mesurée	Différence
3	154,2	145,9 ± 2,5	- 8,3
5	218,5	224,4 ± 3,4	+ 5,9
6	240,2	240,6 ± 3,4	+ 0,4
6 c*	329,3	323,0 ± 4,7	- 6,3

* Combustible de type cermet

On remarque immédiatement que les différences entre masses mesurées et masses déclarées dépassent sensiblement les incertitudes calculées.

Une source d'erreur, qui n'a pas pu être prise en considération dans le calcul, provient du fait que les conditions expérimentales ne conduisent pas à un échantillonnage statistique de la surface à examiner. En effet, la présence des trois raidisseurs longitudinaux exclut l'examen de la plage médiane des plaques, tandis qu'on a écarté de l'analyse les régions voisines des extrémités de la zone active, régions où les tolérances de fabrication sont notablement plus larges. Un échantillonnage plus complet de la surface active révélerait très certainement une dispersion accrue des mesures. Celle-ci, toutefois, ne rendrait pas compte des différences observées, car on a constaté que c'est l'erreur associée aux tolérances sur les surfaces actives qui est prépondérante.
Il est donc inutile d'augmenter le nombre de positions de mesure ou les temps de comptage : on n'y gagnerait pas en exactitude.
Dès lors pour améliorer la méthode, il faudrait mieux évaluer la surface active de chaque élément. Ceci peut se faire :

a) en utilisant un collimateur plus étroit et en admettant comme mesure de la longueur moyenne des plaques la distance entre les points pour lesquels l'intensité mesurée est la moitié de celle de la zone centrale, ou

b) en cours de fabrication, lors de l'examen aux rayons X.

Des conclusions définitives ne pourraient être présentées qu'après l'examen d'un lot plus étendu d'éléments, mais on peut admettre que la précision ultime s'approche de 1 % de la masse présente en uranium-235. La mesure s'applique qu'il y ait ou non des absorbants (poisons consommables).

Conclusion

Les deux méthodes présentées dans ce paragraphe, tout en arrivant à des précisions comparables, se fondent sur des propriétés très différentes. Couplées, elles fournissent un excellent outil pour la vérification.

(iv) Stockage des éléments de combustible avant envoi à l'usine de retraitement

Parmi les différentes mesures non destructives d'éléments irradiés on a retenu la spectrométrie gamma des produits de fission [11]. L'expérience décrite a pour but d'établir jusqu'à quel point il est possible de vérifier avec cette technique les déclarations de l'exploitant concernant la masse initiale, l'enrichissement et les données

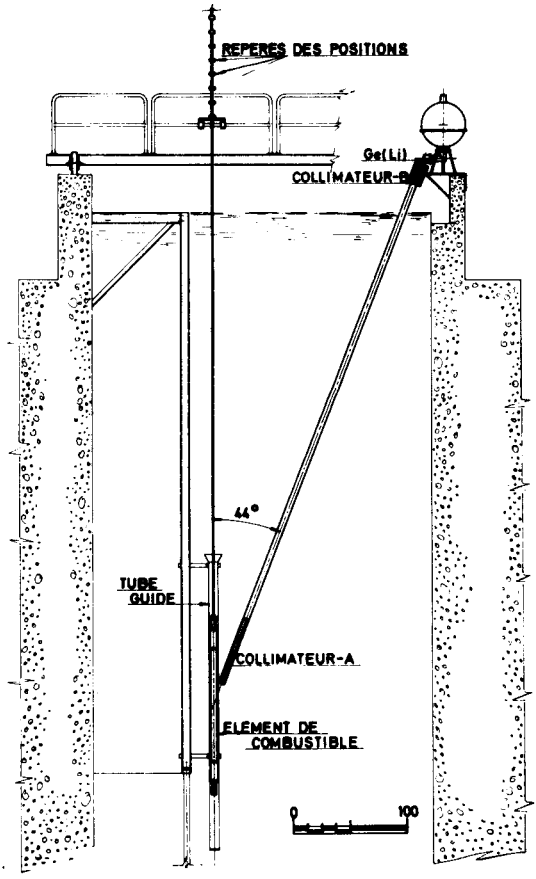


FIG. 2. Dispositif de mesure des éléments irradiés (spectrométrie gamma).

d'irradiation. On a choisi un lot de cinquante éléments de combustible ayant des taux d'épuisement très proches (37 à 41 %). Ce lot sera analysé au spectrographe de masse lors de la dissolution à l'usine de retraitement. On obtiendra alors un point de calibration résultant de l'analyse destructive.

La disposition du tube guide des éléments de combustible et du tube collimateur est présentée à la figure 2. L'angle d'immersion est de 44°. La collimation a été renforcée aux extrémités du tube par un collimateur large (diamètre 2 cm) près de l'élément et par un collimateur relativement étroit (diamètre 0,3 cm) près du détecteur Ge(Li). Un second tube collimateur a été ajouté pour obtenir simultanément des mesures au NaI (Tl).

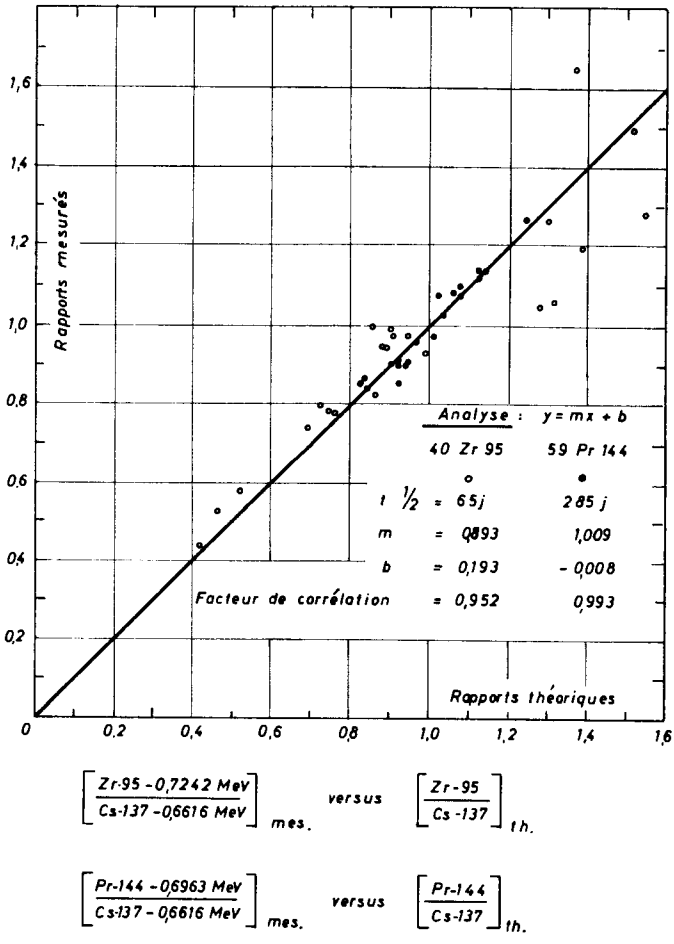


FIG. 3. Mesure des éléments irradiés: comparaison des rapports Pr-144/Cs-137 et Zr-95/Cs-137.

La résolution de la chaîne de spectrométrie Ge(Li) était de l'ordre de 3 keV. Pour chacun des éléments, cinq spectres ont été mesurés entre 450 keV et 3 MeV aux cotes - 25, - 5, + 5, + 25 cm. L'ensemble des mesures a été transféré sur bande magnétique ce qui fournit une librairie de données expérimentales facilement accessibles portant sur environ 250 spectres. L'analyse de ces données fournit les intensités gamma des isotopes : Cs-134, Cs-137, Zr-95, Nb-95, Rh-106, Pr-144. Certains isotopes (Pr-144, Rh-106) sont caractérisés par plusieurs pics bien espacés, ce qui permet d'appliquer aux intensités mesurées une correction qui tient compte de l'efficacité de détection et de l'atténuation dans l'élément de combustible pour toute la gamme des énergies.

Dans un premier stade, les données d'exploitation sont éprouvées par les rapports d'activité de deux isotopes. Des résultats caractéristiques sont présentés à la figure 3 pour le couple Pr-144 (696,3 keV) Cs-137 (661,6 keV) et pour le couple Zr-95 (724,2 keV) Cs-137 (661,6 keV). Ces résultats sont normalisés aux moyennes des rapports théoriques et mesurés. D'autres couples d'isotopes sont à l'étude.

III. CHAINE DE FABRICATION DE COMBUSTIBLE URANIFERE POUR REACTEUR DU TYPE L.W.R., INSTALLEE A L'USINE M.M.N.

La recherche et l'optimisation des méthodes de contrôle à l'usine de fabrication pour les combustibles du type L.W.R. sont importantes pour le développement du système de garanties, non seulement à cause des quantités mises en jeu mais également parce que les mesures faites dans cette partie du cycle de combustible fournissent les informations fondamentales sur lesquelles se fonderont les corrélations appliquées à l'entrée des réacteurs et des usines de retraitement.

La campagne de fabrication du combustible du réacteur de Doel (type P.W.R., 390 MWe) fournira un champ d'application de taille industrielle pour ces méthodes. Des résultats ont déjà été obtenus en laboratoire, notamment en ce qui concerne les poudres et l'interrogation des éléments de combustible avant assemblage.

(i) Mesure de poudre UO_2

Une méthode a été adaptée pour contrôler les enrichissements en uranium-235 dans les poudres d'oxyde d'uranium. La méthode se base sur les mesures de spectrométrie gamma pour une position définie de l'échantillon et du détecteur, de telle sorte que l'on obtienne une épaisseur "infinie" de l'échantillon pour le rayonnement gamma mesuré. Pour une géométrie ainsi définie, le taux de comptage du rayonnement gamma de l'uranium-235 est proportionnel à l'enrichissement en uranium-235 dans le matériau.

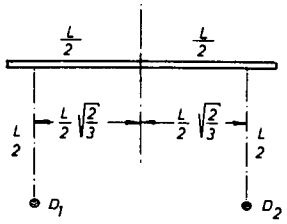
On peut utiliser comme détecteur un NaI(Tl) ou un semi-conducteur. Le NaI(Tl) présente l'avantage d'un haut rendement de détection ce qui conduit à des temps de comptage courts. Un semi-conducteur possède un meilleur pouvoir de résolution ce qui est requis dans le cas des matériaux contenant des éléments lourds autres que l'uranium.
Les résultats obtenus jusqu'à présent montrent que :

1) la méthode est simple, rapide et fiable : un temps de comptage de 5 minutes donne une précision meilleure que 0,5 %
2) l'utilisation de spectromètres avec stabilisateur de pics facilite les mesures.

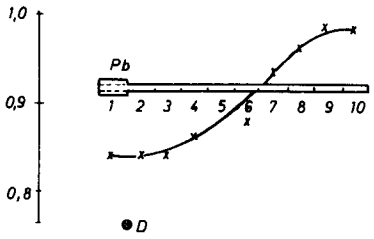
Des résultats préliminaires indiquent que la méthode pourrait être étendue à la détermination des concentrations d'autres isotopes fissiles dans divers types de matériaux homogènes.

(ii) Mesure des crayons avant assemblage

La méthode proposée a pour but de vérifier la teneur en uranium-235 des crayons. On effectue deux mesures intégrales dans des conditions géométriques décrites à la figure 4. L'activité est mesurée par deux détecteurs NaI (Tl) placés en D1 et D2. Les distances ont été calculées de telle sorte qu'en faisant la somme des deux mesures chaque pastille intervient pratiquement de la même façon dans le comptage.
Cette propriété a été démontrée sur un crayon de combustible UO_2 enrichi à 4 %, de 50 cm de longueur active et de 1 cm de diamètre. Une perturbation locale dans l'empilement des pastilles a été simulée en déplaçant le long du crayon un anneau de plomb de 2 mm d'épaisseur et de 5 cm de large. L'atténuation locale qui en résulte correspond à environ 8 % de l'intensité intégrée. L'anneau a été placé en 10 positions successives le long du barreau et les comptages ont été relevés par un seul compteur situé en D1. On a ensuite effectué la somme des comptages des positions symétriques (1 et 10, 2 et 9 etc.) ce qui correspond à l'activité qui aurait été mesurée au moyen des deux compteurs NaI placés en D1 et D2.
Les sommes successives étaient :
47 744, 47 910, 47 235, 47 135, 46 960 coups à comparer au comptage sans perturbation qui donnait 52 334 coups.
La perturbation mesurée est ainsi de 8,5 à 10,3 % selon la position, ce qui démontre suffisamment la propriété énoncée. L'avantage de cette méthode est sa simplicité. Elle ne requiert aucun élément mécanique mobile, mais seulement un support approprié et une chaîne de comptage.



a) CONDITIONS GEOMETRIQUES DE LA MESURE



b) ARRANGEMENT REEL : Intensité mesurée en fonction de la position de la perturbation (intensité en l'absence de perturbation = 1)

FIG. 4. Mesure des éléments de combustible non irradiés (spectromètre gamma).

IV. INSTALLATION DE FABRICATION DE COMBUSTIBLES AU PLUTONIUM DE BELGONUCLEAIRE A DESSEL

(i) Avant-propos

En vue de la coopération souhaitée entre l'Etat et l'Agence, il est essentiel que l'Etat et l'exploitant collaborent afin de faciliter la mise en oeuvre des garanties. Cette collaboration a permis de dégager des principes généraux communs aux différents types d'installations se trouvant en Belgique.

Pour permettre à l'Etat de mener à bien l'application des garanties, les caractéristiques spécifiques à une installation et les renseignements descriptifs particuliers seront tenus à disposition. Il s'agit principalement de l'identification de l'installation, l'aménagement général intérieur, les caractéristiques relatives au contrôle des matières fissiles et fertiles.

Dans l'analyse résumée ci-dessous, l'exploitant décrit le cadre de ses travaux concernant le contrôle des matières fissiles et fertiles.

En ce qui concerne l'installation de fabrication d'éléments combustibles enrichis au plutonium en cours de réalisation, les renseignements sus-mentionnés sont collectés à mesure de la disponibilité des données en vue de l'établissement d'un document complet. Le rapport doit permettre une compréhension suffisante de l'usine pour tout contrôleur extérieur afin qu'il puisse lui-même juger du degré de confiance qu'il peut attribuer au contrôle des matières, effectué par l'exploitant, et qu'il puisse optimiser le type et la fréquence des contrôles qu'il souhaiterait voir exécuter en sa présence.

(ii) Identification de l'installation

L'objet de l'installation est la fabrication de barreaux combustibles à pastilles enrichis au plutonium pour les types de réacteurs rapides et thermiques.

Etant donné qu'il s'agit d'une usine en cours de réalisation, les informations complémentaires d'identification générale seront basées sur les études spécifiques intéressant le contrôle des matières fissiles (inventaire, flux matière, temps de séjour) et sur les travaux effectués pour la production pouvant être exploités en vue de l'estimation de l'efficacité du modèle. L'identification générale sera complétée par la description des contraintes auxquelles est soumis l'exploitant, fixant par là même les limites contrôlables et contrôlées du point de vue qualité, quantité, manipulation ou stockage des matières dans l'installation.

a) Etudes

Parmi les études principales dont les conclusions feront partie du rapport descriptif, il faut citer les travaux devant aboutir à la représentation mathématique de l'installation, compte tenu de l'équipement, du procédé, et des moyens mis en oeuvre pour le contrôle. La formulation mathématique doit permettre entre autres :

- d'évaluer les flux de matières, les temps de séjour, etc.
- d'optimiser le nombre, les emplacements des points stratégiques, ainsi que les fréquences, précision, type de méthodes et technique de détection à choisir pour le contrôle des matières, de même que le rapport coût-efficacité,

- d'estimer la crédibilité du modèle proposé,
- de déterminer l'efficacité du système du point de vue de l'exploitant, de calculer ensuite la sensibilité de détection de détournement et les risques de l'exploitant en cas de détournement (cf. niveau national),
- de perfectionner le modèle,
- etc.

Il faut citer encore les travaux du Service Contrôle de l'usine qui, en parallèle à l'établissement par le fabricant des spécifications de référence, détermine les plans de contrôle de référence (plans d'échantillonnage et d'inspection) permettant de garantir, aux niveaux de l'exploitant et du client, les spécifications des matières premières ainsi que des produits demi-finis et finis.

b) Contraintes

Toujours dans le but de permettre au contrôleur extérieur d'estimer le degré de confiance qu'il peut attribuer à l'ensemble du système, il semble utile de lui communiquer les principales contraintes auxquelles l'exploitant est soumis.

- Forme des matières

Le permis de construction de l'usine a été accordé par un Arrêté Royal[1], rédigé par l'Autorité compétente, celle-ci étant le Ministère de l'Emploi et du Travail. Cet Arrêté autorise l'édification d'une installation de fabrication de combustibles mettant en oeuvre des oxydes d'uranium et de plutonium, principalement sous forme de poudre, de pastilles et de barreaux. Il est à souligner, étant donné le procédé de fabrication adopté, que dès la première étape de fabrication l'oxyde de plutonium est dilué dans l'oxyde d'uranium.

- Quantité maximale

L'Arrêté Royal précité stipule qu'une quantité maximum de 100 kg de plutonium (exception faite pour les halls "Analyses" et "Déchets" où la limite est de 2 kg) est admissible dans chaque hall de l'usine, compte tenu des études faites par l'exploitant, examinées par la Commission Spéciale des radiations ionisantes[2]. Les procédures relatives à la mise en oeuvre des quantités de plutonium dans l'établissement doivent être approuvées d'une part, par le Service du Contrôle Physique (Protection Sanitaire) attaché à l'établissement et d'autre part, par l'Organisme Agréé[3] chargé de vérifier si l'exploitation de l'installation s'effectue conformément aux prescriptions des arrêtés royaux précités.

- Criticité

L'exploitant devra, dans le cadre des études de sécurité, définir en particulier les prescriptions et les quantités maximales admissibles en opération et en stockage afin d'assurer la prévention à la criticité. Ces limites seront étudiées suivant le procédé, l'équipement, le contrôle et le type de combustible à fabriquer. Elles entreront en vigueur

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[1] Arrêté Royal N° S 3150 du 27.10.1970.

[2] Commission Spéciale en matière de radiations ionisantes instituée par l'Autorité Compétente conformément à l'Arrêté Royal du 28.2.1963 portant sur le règlement général de la protection de la population et des travailleurs contre les dangers des radiations ionisantes.

[3] Organisme Agréé en Classe 1 par Arrêté Ministériel.

après accord du Service du Contrôle Physique et de l'Organisme Agréé, en conformité avec les principes généraux définis par Arrêté Royal. Chaque unité opérationnelle établit un bilan-matières.

- Assurance

L'exploitant établira aussi des règles de travail tendant à limiter les quantités de matières en manipulation par rapport aux quantités de matières stockées, sans toutefois entraver la bonne marche de l'usine, étant donné que les montants des assurances des matières ne se payent que pour les quantités en circulation.

(iii) Description de l'aménagement intérieur

La philosophie générale s'appuie sur les critères principaux suivants : étanchéité du modèle et accessibilité des contrôleurs extérieurs aux halls relatifs aux contrôles et au stockage.

Confinement : la concentration de tous les halls en un bâtiment, ayant un passage entrée-sortie obligé et contrôlable, permettra de garantir le confinement de l'installation.

Procédé : l'installation est divisée en halls de stockage (U, Pu, déchets), de contrôle (destructifs et non destructifs), de fabrication. Chaque hall peut comprendre un ou plusieurs ensembles. Pour chaque hall un bilan de fabrication est établi.

Accessibilité : les halls sont regroupés suivant qu'ils contiennent des matières accessibles ou non au contrôle (stockage-contrôle d'une part et fabrication d'autre part). L'adoption d'un tel critère est issue de l'énoncé de l'Accord, qui prévoit que les garanties sont mises en oeuvre de manière à éviter d'entraver Ie développement économique et technologique, de gêner indûment les activités, de manière à être compatible avec une saine gestion. Le respect de ce dernier point est lui-même confirmé par les résultats des premières études mathématiques qui ont montré :
- la faible proportion des matières en manipulation par rapport aux matières stockées et en contrôle,
- le temps de séjour très court dans la partie de l'installation où la matière n'est pas accessible au contrôle et ce pour un pourcentage élevé de matière contenue.

Contrôle des matières : tout au long de la chaîne de fabrication se trouvent les endroits obligés de contrôle des matières (quantité et qualité). Entre deux points clefs de contrôle est établi le bilan-matières.

(iv) Caractéristiques relatives au contrôle des matières

La Section "contrôle des matières fissiles et fertiles" est en charge de la comptabilité et de l'inventaire physique des matières fissiles et fertiles. Cette section dépend du Service Contrôle. Ce dernier travaille parallèlement à la fabrication. En plus de cette tâche, le Service Contrôle participe à des programmes de qualification dont entre autres :
- "Umpire Qualification Program" organisé par Idaho Nuclear Corporation à la demande de l'U.S.A.E.C. et ayant pour objet aussi bien la qualification du point de vue "Safeguards" que du point de vue de la qualité chimique du produit nucléaire.
- le programme de qualification organisé à la demande de fabricants de combustibles par Battelle North-West ayant pour objet la qualification du point de vue contrôle de spécifications de fabrication.

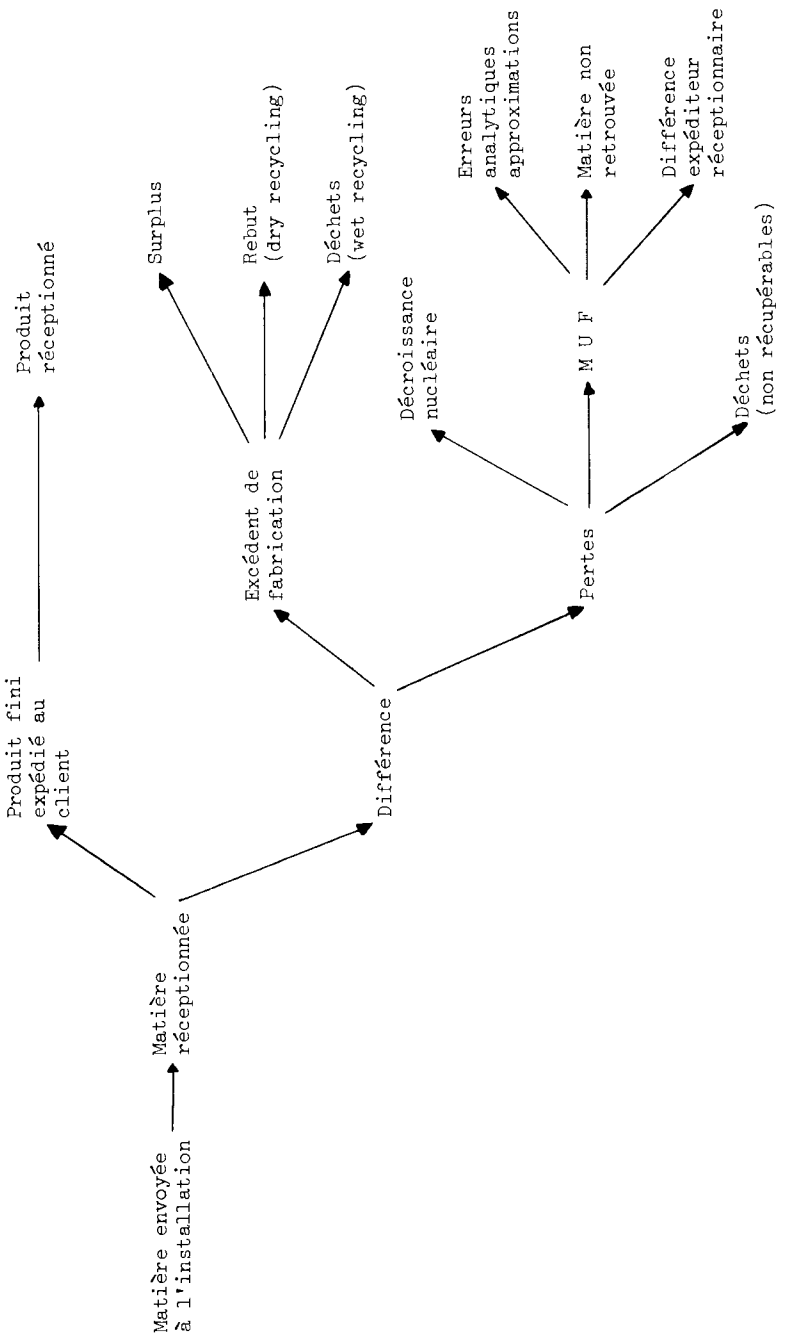

FIG. 5. Bilan-matières.

a) Inventaire comptable

L'inventaire comptable sera réalisé sur ordinateur par liaison télé-processing suivant un programme actuellement en rodage dans la chaîne pilote de fabrication. L'application fournit un inventaire permanent, mis à jour continuellement, permettant aux intéressés de connaître de façon détaillée et précise les quantités, qualités, localisations, mouvements, transformations et usages des matières. Ce bilan-matières est unique pour l'installation et par là même les valeurs qu'il traite sont indépendantes du but (gestion, fabrication, sécurité, contrôle matières) pour lequel elles y ont été introduites.

Le principe de réalisation peut se résumer comme suit : l'application est divisée en trois parties consacrées respectivement aux lots (quantité et qualité), aux localisations et aux mouvements. Les programmes qui permettent l'exécution des transferts en constituent les points importants. Ces programmes exécutent en outre le nouveau bilan-matières suivant la classification reprise à la figure 5. L'édition principale est celle des relevés des postes créditeurs pour les "zones physiques" et débiteurs pour les "zones comptables".

Le principe de fonctionnement est repris ci-après : toute entrée de matières fissiles du "LOT INITIAL" correspondant à un transfert externe est enregistrée au poste "TRANSIT" ou au poste "RECEPTION" selon que la matière arrive dans l'installation avant ou après réception officielle. La matière est introduite physiquement dans des unités de stockage. Toute matière prélevée entraîne un transfert interne du stockage vers les halls. Les fabrications exigent un "MELANGE" de matières appartenant à des lots différents engendrant de la sorte un "LOT SECONDAIRE". La matière traitée sera stockée à la sortie de l'installation suivant sa qualité. La sortie se fait par le poste "EXPEDITION". Le poste "PERTES" apure la comptabilité bilan-matières.

b) Inventaire physique

L'inventaire comptable est doublé d'un contrôle physique permanent de la matière, exécuté en des points clefs de contrôle, pouvant être complété, en cas de nécessité, par un inventaire physique complet. Ces endroits délimitent des unités bilan-matières. Ils correspondent aux postes réception-expédition de la matière vers l'extérieur (faisant l'objet d'un transfert externe, critère : confinement) entrée-sortie des halls de stockage et de contrôle (faisant l'objet d'un transfert interne de la matière, critère : accessibilité), entrée-sortie du mélange (faisant l'objet d'un transfert interne, critère : dilution de plutonium).

En ces points clefs de contrôle, la matière est vérifiée du point de vue qualité-quantité. Ces contrôles permettent de préciser le MUF du bilan. De plus, en ces postes les quantités de matières introduites dans la comptabilité sont exprimées directement en unités de compte.

Les contrôles sont effectués soit par échantillonnage soit sur la matière elle-même.

Le type et la fréquence des contrôles sont choisis en fonction des proportions relatives des diverses classes de la matière (fig. 5) pour garantir la précision requise sur le bilan total de l'installation. Pour tout ce qui concerne les transferts externes un contrôle à 100 % est effectué. Les types de contrôle choisis sont principalement des tests destructifs tels que composition isotopique et coulométrie à potentiel contrôlé pour les contrôles nécessitant des valeurs absolues.

V. CONCLUSIONS

Ce mémoire montre les voies suivies pour atteindre les objectifs du contrôle des garanties. Les travaux sont axés sur les points suivants :

1. l'analyse par simulation et développement du symbolisme mathématique adéquat correspondant aux installations considérées ;
2. le développement des techniques de mesure non destructives et éventuellement destructives pour l'évaluation du bilan-matières ;
3. l'estimation de l'efficacité des procédures en se fondant sur les résultats des deux premiers points ;
4. l'application dans le cadre d'expériences intégrales limitées aux points clefs de contrôle.

Pour renforcer la crédibilité du système de contrôle des garanties basé sur l'emploi d'instruments et autres moyens techniques en certains points stratégiques, on a poursuivi l'effort entrepris dans le cadre de la campagne "Umpire Qualified Laboratories". On dispose ainsi de techniques de mesure déjà éprouvées et comparables au niveau international.

REFERENCES

[1] Agence Internationale de l'Energie Atomique INFCIRC/140, 22 avril 1970

[2] Agence Internationale de l'Energie Atomique
INFCIRC/153, mai 1971, alinéas : 28, 29, 46 b iv

[3] ERRERA J., SYMON E., VANDERMEULEN J., VERNAEVE L.
Euratom - Analyse et commentaires du Traité
Institut Belge de Science Politique
Les Editions de la Librairie Encyclopédique S.P.R.L. Bruxelles (1958)

[4] KOUTS H.
System Studies for Nuclear Material Safeguards
Technical support Organization
Brookhaven National Laboratories (1969)

[5] ARSENAULT F.J.
Research and Development for Safeguards
WASH-1122 (Safeguards and Nuclear Material Management)
December 1, 1968 p. 73

[6] BEETS C., GOOSSENS H., MOSTIN N.
Safeguards Techniques
Proceedings of a symposium
Karlsruhe, 6-10 July 1970, vol.I p. 251-270 I.A.E.A., Vienna(1970)

[7] MORGAN F. (in consultation with BENETT C.A.)
Report to the Director General of the International Atomic Energy Agency, by the Consultants on Criteria for Safeguards Procedures
Topic 3, Part 1 p. 35 (1969)

[8] COCQUYT G., HECQ R. Blg 445 (1970)

[9] SERVAIS F., GOLDSCHMIDT P.
Transactions of the American Nuclear Society 1970 Winter Meeting
Washington, November 15-19, 1970
Vol. 13, N° 2, pp. 478-479 (1970)

[10] LENDERS H. Blg 443 (1970)

[11] RASMUSSEN N.C.
WASH 1076 pp. 130-134 USAEC (1967)

OPTIMIZED MATERIAL ACCOUNTING SYSTEM FOR SAFEGUARDS IN A NUCLEAR FUEL CYCLE

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Abstract–Résumé–Аннотация–Resumen

OPTIMIZED MATERIAL ACCOUNTING SYSTEM FOR SAFEGUARDS IN A NUCLEAR FUEL CYCLE.

Of the three basic safeguards measures, material balance accountancy, containment and surveillance, the first-mentioned enables a safeguards organization to make objective and quantifiable statements with regard to a diversion. This measure is therefore expected to form an important component of any modern safeguards system. However, activities associated with the establishment of a credible material balance in different facilities of a nuclear fuel cycle are mainly manpower- and capital-intensive. Manpower and capital investments are interrelated with respect to the tamper-resistance of the system for generation, registration, processing and reception of information. For this reason, it is anticipated that many of the research and development activities in the field of safeguards will be directed towards investigation of means and methods to reduce efforts in establishing credible fissile material balances in a fuel cycle.

In the present paper, a number of parameters are analysed which may cause such a reduction in efforts for safeguards. These parameters are: use of historical data, transparency of the safeguards information system, fuel cycle correlation and containment measures. They are so applied that the uncertainties in the material balance established are not increased. It has been shown with the help of some numerical examples that a significant reduction of efforts in establishing such a material balance can be achieved by using these parameters in a nuclear fuel cycle consisting of reactors, reprocessing plants and fabrication plants as well as conversion and isotope separation facilities.

SYSTEME OPTIMALISE DE COMPTABILITE MATIERES D' UN CYCLE DE COMBUSTIBLE NUCLEAIRE AUX FINS DES GARANTIES.

Parmi les trois opérations fondamentales de garanties, à savoir le calcul du bilan matières, le confinement et la surveillance, la première est celle qui permet à l' organisme chargé des garanties de consigner des observations objectives et quantifiées de détournements éventuels. Il faut donc s' attendre qu' elle constitue un élément important d' un système moderne de garanties. En revanche, les activités afférentes à l' établissement d' un bilan matières digne de foi dans les différentes installations d' un même cycle de combustible nucléaire demandent beaucoup de personnel et des dépenses considérables. Le personnel et les fonds sont étroitement liés en ce qui concerne la rigueur du système de collecte, d' enregistrement, de traitement et de réception de l' information. Pour cette raison, on prévoit que la plus grande partie des recherches et des études en matière de garanties seront orientées vers les moyens et méthodes capables de faciliter l' établissement de bilans de matières fissiles dignes de foi à l' intérieur d' un cycle de combustible.

Les auteurs analysent un certain nombre de paramètres qui peuvent permettre de réduire les moyens mis en œuvre pour les garanties. Ces paramètres sont les suivants: les données «historiques», la transparence du système d' acquisition et de traitement de l' information aux fins de garanties, la corrélation du cycle des combustibles et les mesures de confinement. Ils sont appliqués de manière à ne pas augmenter les incertitudes du bilan matières établi. Les auteurs montrent, avec l' aide de quelques exemples numériques, que l' on peut simplifier considérablement l' établissement de ce bilan matières en utilisant ces paramètres pour un cycle de combustible comprenant des réacteurs, des usines de retraitement et des usines de fabrication, ainsi que des installations de transformation et de séparation isotopique.

ОПТИМАЛЬНАЯ СИСТЕМА УЧЕТА ЯДЕРНЫХ МАТЕРИАЛОВ ДЛЯ СИСТЕМЫ ГАРАНТИЙ В ОТНОШЕНИИ ЯДЕРНЫХ ТОПЛИВНЫХ ЦИКЛОВ.

Из трех основных мер системы гарантий – баланс делящихся материалов, опечатывание и надзор – только первая мера позволяет организации системы гарантий делать объективные и достоверные в количественном отношении заявления о переключении материала на

другие цели. Поэтому ожидают, что эта мера явится важной составной частью любой современной системы гарантий. Однако работа по подведению наиболее вероятного баланса в установках с различным топливным циклом определяется,в основном, численностью личного состава и капиталовложениями. Личный состав и капиталовложения, в свою очередь, зависят от степени организованности системы выборки, регистрации, обработки и хранения информации. По этой причине ожидается, что большая часть исследовательской и организаторской работы в области системы гарантий будет направлена на разработку способов и методов, с помощью которых можно будет уменьшить усилия, затрачиваемые на подведение баланса делящихся материалов в топливном цикле.

В представленном докладе анализируются некоторые параметры, которые могут помочь уменьшить такие усилия для целей гарантий. Такими параметрами являются: временные последовательности, простота информационной системы гарантий, корреляция между топливными циклами и измерения сохранности делящихся материалов. Их применение должно происходить таким образом, чтобы неопределенности в подведенном балансе делящихся материалов не возрастали. На нескольких численных примерах показано, как можно существенно сократить затраты усилий на подведение такого баланса, если топливный цикл, включающий реактор, заводы по изготовлению и переработке ядерного топлива, а также обогатительные установки и установки по разделению изотопов рассчитать с помощью указанных параметров.

SISTEMA OPTIMIZADO DE CONTABILIDAD DE MATERIALES PARA LA APLICACION DE SALVAGUARDIAS EN UN CICLO DE COMBUSTIBLE NUCLEAR.

De las tres medidas básicas de salvaguardias, contabilidad de balances de materiales, contención y vigilancia, la contabilidad de balances permite a una organización de salvaguardias efectuar comprobaciones objetivas y susceptibles de expresión cuantitativa en relación con una desviación de material. Se espera, por tanto, que esta medida constituya un componente importante de un sistema moderno de salvaguardias. Sin embargo, las actividades asociadas con el establecimiento de balances verosímiles de materiales en las diversas instalaciones de un ciclo de combustible nuclear dependen fundamentalmente del número de personal y de las inversiones. El número de personal y las inversiones están relacionados con el grado de confianza del sistema en la producción, registro, tratamiento y recepción de la información. Por ese motivo, se espera que una gran parte de las actividades de investigación y desarrollo en el campo de las salvaguardias esté dirigida hacia la búsqueda de procedimientos y métodos que reduzcan los esfuerzos para establecer balances verosímiles de materiales fisionables en un ciclo de combustible.

En la presente memoria se analiza una serie de parámetros que puede producir una disminución de esos esfuerzos. Estos parámetros son la utilización de datos del historial, la claridad y simplicidad del sistema de información para salvaguardias, correlación del ciclo del combustible y medidas de contención. Se aplican de manera tal que no aumente el grado de incertidumbre en el balance de materiales fijado. Se ha demostrado, con ayuda de algunos ejemplos numéricos, que puede conseguirse una reducción considerable de los esfuerzos para fijar tal balance de material mediante la utilización de esos parámetros en un ciclo del combustible nuclear constituido por reactores, instalaciones de reelaboración de combustibles irradiados y de fabricación de combustibles, así como instalaciones de tratamiento y de separación de isótopos.

1. INTRODUCTION

It is a well accepted fact that, of the three basic safeguards measures, material balance, containment and surveillance, the first mentioned will find maximum application in a modern safeguards system. At present, it is the only measure which enables a safeguards authority to make quantifiable statements in the case of a possible diversion. However, activities associated with the establishment of a credible material balance in different facilities of a nuclear fuel cycle are mainly manpower- and capital-intensive. For this reason, it is anticipated that many of the R. and D. activities in the field of safeguards will be directed towards the investigation of means and methods to reduce efforts in establishing credible fissile material balances in a fuel cycle. These investigations include, among others, definitions of the effectivity of a safeguards system [1] and optimization with varying degrees of uncertainties in the material balance or by the use of game theoretical considerations [2]. In the present paper, a number of ad-

ditional parameters have been investigated which may be considered for the same purpose. The main idea behind this approach is the utilization of those characteristics of a fuel cycle which are inherent, or which can be made inherent to it, and cannot be influenced easily or significantly by the operators of individual facilities in such a cycle. The results, which should be regarded as preliminary, indicate that, by using these parameters, not only is the credibility of the information improved, but the efforts required to establish a credible material balance are reduced.

It is to be noted in this connection that, in contrast to the investigations described in Ref. [2], in which a reduction in efforts was achieved mainly by increasing the uncertainties in the established material balance, the present investigation is directed towards reducing efforts without increasing such uncertainties.

2. INPUT DATA

2.1. Fuel-cycle data

To establish an optimized material accounting system for a given fuel cycle, the basic characteristics of the fuel cycle (e.g. number and characteristics of facilities, throughputs, inventories) have to be known to the safeguards organization.

The reference fuel cycle considered here is shown in Fig. 1. The cycle data required for this purpose are presented in Table I. It is to be noted that the cycle consists of 12 light-water type reactors, and one reprocessing, one fabrication, one isotope separation and three conversion plants for the uranium cycle. The plutonium produced has been assumed to be fabricated in an additional Pu fabrication plant with the fuel pins produced being stored in casks.

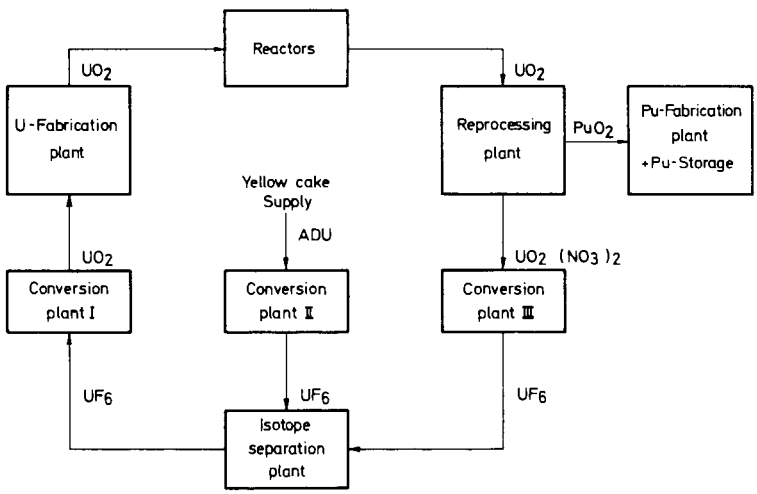


FIG. 1. Reference fuel cycle with nuclear facilities.

TABLE I. NUCLEAR FACILITIES CONSTITUTING THE REFERENCE FUEL CYCLE FOR 6 GW(e) INSTALLED POWER

Facility Characteristics	Reactors	Reprocessing plant + Pu storage	U fabrication plant	Isotope separation plant	Conversion plant I	Conversion plant II	Conversion plant III	Pu fabrication plant
No.	12	1	1	1	1	1	1	1
Type	LWR; batch loading; 1/3 core loading/ unloading/year Init. U-235 conc. 3% Final U-235 conc. 1% Final Pu conc. 1%	Purex, Pu produced stored	UO_2 pellet in pins and sub-assemblies	UF_6-enriehm. Feed 1% and 0.71% U-235 Product 3% U-235	Chemical UF_6-UO_2 (3% U-235)	Chemical $(NH_4)_2 U_2 O_7$ (ADU)-UF_6 (nat. U)	Chemical $UO_2(NO_3)_2$-UF_6 (1% U-235)	PuO_2 pellets, pins for fast breeder sub-assemblies
Capacity	500 GW(e)/reactor or 112 sub-assemblies							
Throughput per month (input)	18 t UO_2/reactor (3% U-235)	18 t UO_2 (1% U-235) + 170 kg PuO_2	18.02 t UO_2 (3% U-235)	23.1 t UF_6 (1% U-235) 105.9 t UF_6 (0.71% U-235)	23.6 t UF_6 (3% U-235)	103 t ADU (nat. U)	26 t $UO_2(NO_3)_2$ (1% U-235)	146.7 kg Pu
Inventory Input process		10 t U 100 kg Pu	16 t U 14 t U	92 t 1% U-235	16 t U 10 t U	42 t U	10 t U	500 kg Pu 330 kg Pu 500 kg Pu (prod.)
No. of working days/yr		200	200	200	200	200	200	200

Furthermore, the basic safeguards measures in the cycle (e.g. safeguards units, numbers of units per year, random and systematic errors of the single measurement) and the time and costs for these measures should also be known so that the effect of the application of the parameters can be assessed. The relevant data are summarized in Table III. The details on the inventory taking data presented here are the same as those in Ref. [2].

It is to be noted that the inspection time for other safeguards measures (data collection, verification of reports, etc.) for each facility has been taken to be 80 man-hours/yr. The only exception made is for conversion plants II and III. Because of the simplicity of the system, 40 man-hours for each of these facilities have been assumed.

2.2. Records

A prerequisite for a material accounting system is the records system kept by the plant operators at the facilities. The requirements for safeguards purposes are laid down in Ref. [3]. The purpose of the record system for safeguards is twofold:

(a) Verification of the data submitted in the accounting reports with those of the accounting records.
(b) Verification and use of source data kept in the operating records for calibration of instruments, etc., by the inspectors.

The integral experiments [4, 5] carried out so far indicate that the records normally kept by the operators are also sufficient for safeguards purposes.

The operating records for chemical analyses are a special case, as they are open-ended with respect to the primary information. They have to be kept within reasonable limits so as not to impose an undue burden on the plant operators.

Records for reference cases, for example a reprocessing plant [6], a heavy water type reactor [7] and a fabrication plant [8], have been developed.

3. CRITERIA FOR OPTIMIZATION AND RELEVANT PARAMETERS

According to Ref. [3], the purpose of the accounting system is to come to a statement, in respect of each material balance area, of the amount of material unaccounted for over a specific period, giving the limits of accuracy of the amounts stated.

To reach this objective in an optimum manner means reducing all the efforts required in establishing a material balance without reducing the quality of the statement. Two categories of parameters which are of importance in this connection have been considered.

3.1. Category I

Parameters which reduce the efforts in a definite but, so far, not fully quantifiable manner, for example:

(a) historical data on material unaccounted for, measurement accuracies, etc.;
(b) transparency of the information system.

3.2. Category II

Parameters which reduce the efforts in a quantifiable manner, such as:

(a) correlation techniques with the help of minor isotopes;
(b) shipper-receiver-correlations.

3.1. Category I – details

(a) Historical data

A safeguards organization must be in a position to assess the implication of MUF (material unaccounted for) after the establishment of a material balance in a nuclear facility. The MUF, as defined in Ref. [3], gives the difference between the book and the physical inventory. The safeguards organization should be able to decide whether the difference can be explained by the normal operating conditions of the facility in question or whether it corresponds to some unusual situation. For this purpose, the relevant historical operation data of a facility may be of great value. In case no such data are available, additional safeguards efforts may be required to explain the existence of the MUF. These historical data, besides adding credibility to the material balance, also lead to a reduction in effort in establishing a credible material balance.

In a previous study [9], an attempt was made to analyse the MUF values for completed campaigns in various nuclear facilities. Such analyses have to be continued to up-date the MUF investigation results. Table II shows some of the results given in Ref. [9]. It can be seen that one of the most important factors influencing the MUF values is the systematic component of the measurement errors. Usually, such a component cannot be established in a single campaign. A possible method of establishing it is inter-lab tests. Such tests carried out on the international level [10] have shown that valuable information on the various error components of a method can be established. However, they have to be repeated continuously to generate historical data on different measurement methods.

Although there is a clear indication that such historical data will reduce the efforts, at present no information is available to the authors on how such a reduction could be quantified. Therefore, the contribution of this parameter has not been taken into consideration in the numerical example given later.

(b) Transparent information system

A number of integral exercises [4, 5, 11] and analyses of lay-outs of some existing fabrication and reprocessing facilities [11, 12] have shown that a modern safeguards system based on material balance accountancy, supplemented by containment and some surveillance measures, can be applied to these plants with certain restrictions. However, the intensity and the extent of safeguards efforts could be reduced if transparent information network systems were introduced in the plant lay-outs. Figures 2

TABLE II. NORMALIZED MUF VALUES (M_{is}) FROM A KNOWN TYPE OF FACILITY AND MATERIAL NORMALIZED WITH RESPECT TO INPUT/FEED AND SYSTEMATIC ERRORS [a]

Mean value (μ): + 0.29

Standard deviation (σ): ± 0.91

Serial No.	M_{is}	Serial No.	M_{is}
1	5.86	46	0.40
2	4.78	47	0.39
3	3.77	48	0.38
4	3.15	49	0.38
5	2.47	50	0.36
6	2.46	51	0.35
7	2.46	52	0.32
8	2.42	53	0.26
9	2.35	54	0.25
10	2.28	55	0.21
11	2.28	56	0.21
12	2.23	57	0.19
13	2.09	58	0.19
14	2.08	59	0.18
15	1.89	60	0.15
16	1.85	61	0.13
17	1.75	62	0.13
18	1.67	63	0.12
19	1.63	64	0.12
20	1.62	65	0.11
21	1.58	66	0.11
22	1.49	67	0.09
23	1.46	68	0.04
24	1.44	69	0.02
25	1.30	70	-0.11
26	1.22	71	-0.16
27	1.09	72	-0.19
28	0.98	73	-0.27
29	0.98	74	-0.28
30	0.85	75	-0.81
31	0.79	76	-0.86
32	0.78	77	-1.04
33	0.74	78	-1.09
34	0.71	79	-1.28
35	0.67	80	-1.37
36	0.66	81	-1.47
37	0.65	82	-1.82
38	0.63	83	-1.82
39	0.56	84	-1.89
40	0.56	85	-2.35
41	0.54	86	-2.46
42	0.48	87	-2.62
43	0.47	88	-4.05
44	0.47	89	-4.61
45	0.42		

[a] M_{is} values greater than 2 and less than -2 are not within the systematic error range with 95% confidence.

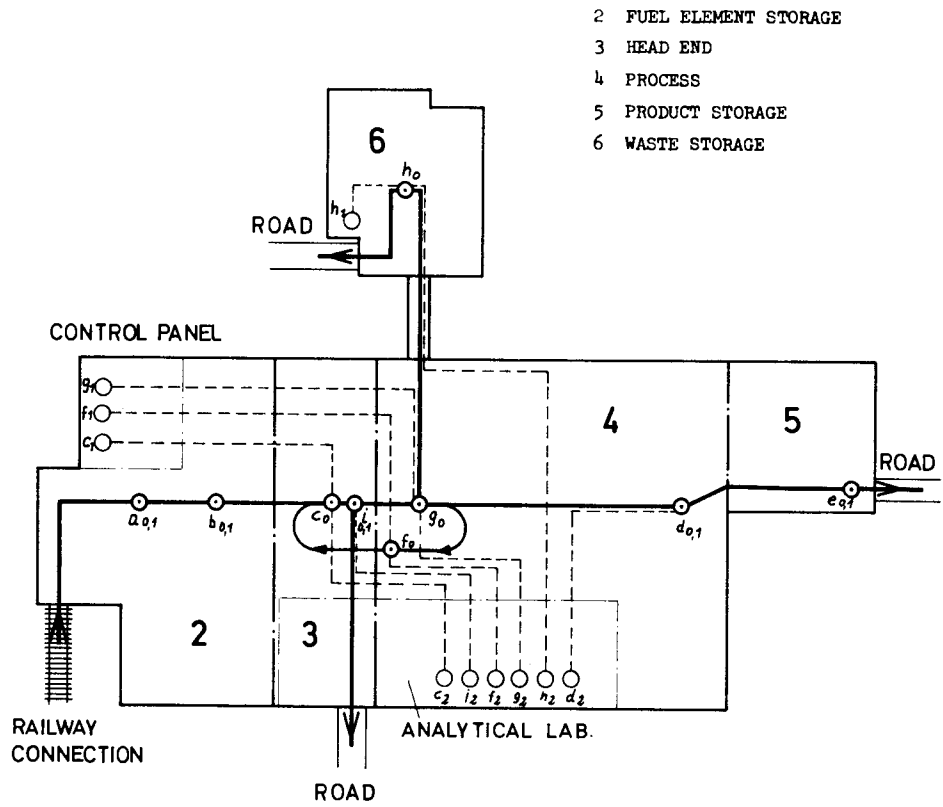
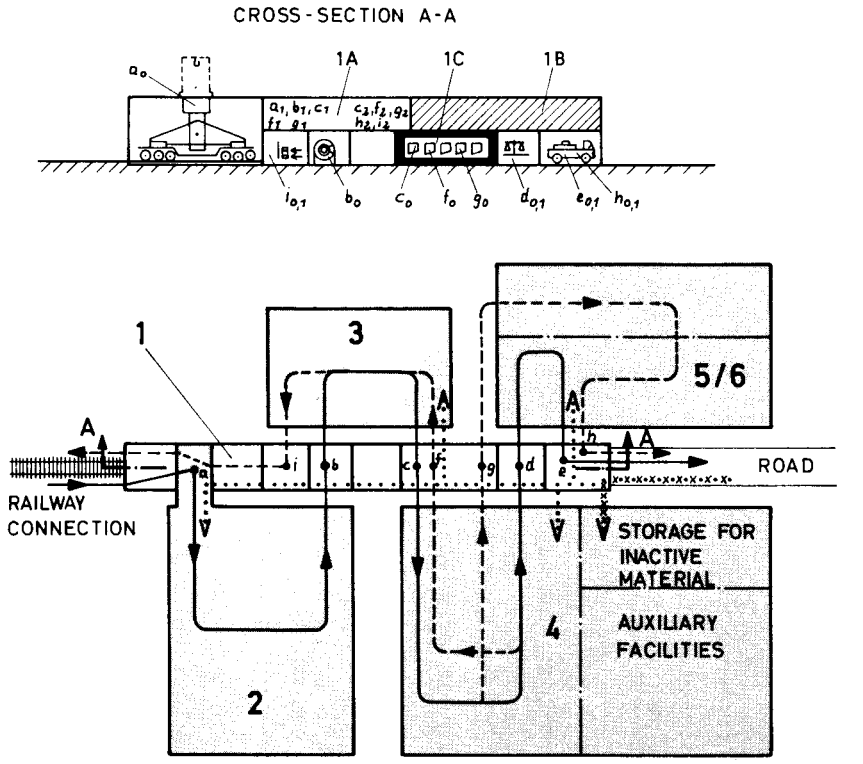


FIG. 2. Information network important to safeguards in an existing reprocessing plant of the WAK type.



ACTIVITIES AT STRATEGIC POINTS

a REGISTRATION, IDENTIFICATION AND INTACTNESS TEST OF FUEL ELEMENTS ARRIVING

b REGISTRATION, IDENTIFICATION AND INTACTNESS TEST OF FUEL ELEMENTS LEAVING THE STORAGE

c INPUT MEASUREMENT OF DISSOLVER SOLUTION

d REGISTRATION, SAMPLING, SEALING AND INSPECTION OF PRODUCT CONTAINERS TO BE STORED

e REGISTRATION, IDENTIFICATION AND INSPECTION OF PRODUCT CONTAINERS TO BE SHIPPED

f RECYCLED ACID MEASUREMENT

g MEASUREMENT OF LIQUID WASTE TO BE SENT TO STORAGE

h MEASUREMENT OF LIQUID WASTE TO BE SHIPPED

i REGISTRATION, MEASUREMENT, SEALING AND INSPECTION OF SOLID WASTE TO BE STORED OR SHIPPED

FLOW OF FISSILE MATERIAL

xxxxxxxx FLOW OF INACTIVE MATERIAL

..... PERSONNEL

1 INFORMATION BUILDING WITH STRATEGIC POINTS
1A PANEL AND ANALYTICAL ROOM FOR SAFEGUARDS AND ACCOUNTABILITY
1B PANEL AND ANALYTICAL ROOM FOR OPERATION
1C ACCOUNTABILITY CELL
2 FUEL STORAGE
3 HEAD END
4 PROCESS
5/6 PRODUCT AND WASTE STORAGE

(For indices see Fig.2)

FIG.3. Arrangement of a modified WAK type plant.

and 3 are reproduced from Ref. [12] to illustrate this point. Figure 2 shows the information system in an existing reprocessing plant of the WAK (Wiederaufarbeitungsanlage für Kernbrennstoffe, Karlsruhe) type. It is seen from this figure that:

(1) the information relevant to safeguards (e. g. fissile material amounts for the feed, product and waste streams) is generated almost throughout the plant;
(2) part of the information is received at the point of its generation (product stream) but a major part is registered at the control panel of the plant (e. g. level indication for feed and waste streams);
(3) all information on fissile material concentration is generated in or near the relevant process steps (e. g. input accountability, product, waste tanks and sampling from the relevant tanks), processed at the laboratory and sometimes received at some other point (e. g. inspection room);
(4) the different accountability tanks, sampling stations, the control panel, the analytical laboratory and the inspection room are located on different floors of the process building and are physically widely apart.

In short, it appears that, in an existing reprocessing plant, the various process steps are arranged in a centralized manner, whereas the information system is decentralized. Particularly this point leads to a complicated, widespread information network system, which requires considerable efforts in ensuring the credibility of the information and accepting it for safeguards purposes.

A conceptual reprocessing plant of the same type is shown in Fig. 3. The main characteristics of this plant are the decentralization of the different process buildings and a centrally located information building.

Under normal conditions, all the safeguards measures can be executed by the inspectorate personnel in the various strategic areas in this building. The building is divided into two floors. The ground floor is partitioned into a number of strategic points (a) to (i), at which all the streams relevant to fissile material balance accountancy are identified, measured or stored. The upper floor is divided broadly into two areas 1A and 1B. In 1A instrument panels, analytical facilities and records and reports offices are provided for safeguards and plant accountability purposes. Area 1B contains control panels and analytical facilities required mainly for plant operation or process control purposes.

It has been estimated that the actual inspection hours (not those required for analysis, identification, sealing, etc.) could be reduced by about 30% with this type of information system as compared with that required for the original lay-out.

It is to be noted that part of the reduction caused by the transparency of an information system can be assessed, for example, by estimating the reduction in the inspection hours. However, another part in the reduction, which expresses itself in the advantages of non-intrusiveness to the plant operator, is difficult to quantify at the present stage.

For optimization purposes, the reduction of 30% in inspection man-hours in the reprocessing and Pu fabrication plant has been taken into consideration.

3.2. Category II – details

(a) Shipper-receiver correlations

In a fuel cycle, the output from a given facility normally forms the input for the next facility. The transfers of nuclear materials usually take place in well defined containers. Typical containers are birdcages or bottles for plutonium transferred from the reprocessing to the fabrication plant, cylinders containing UF_6 for isotope separation in a conversion or fabrication plant, and fuel elements transferred from the fabrication plant to the reactor and also from the reactor to the reprocessing plant. In such cases, when the nuclear material in a container has been measured to establish a material balance at the input or output of a facility, it need not be measured again at the output or input of the adjacent facility. The measurements at the latter points can be replaced completely by containment measures, i.e. sealing and identification of the containers. As is shown in the numerical example, there is thus a significant reduction in effort. The quality of statement on a material balance is not reduced in any way, since the measurement values from the previous stage of the fuel cycle can be taken over fully.

It should be noted that, with containment measures of this type, the information on material balance can be frozen in time and space for a limited period. This means that the information generated at one point of the cycle can be used without any significantly large additional effort at some other point of the cycle at a later date, and the credibility of the information remains intact during this period.

(b) Isotopic correlations

The inherent relationships between the formation of Pu and some reactor parameters such as depletion of U or built-up of ^{236}U and Pu isotopes have great potential for safeguards purposes. In some recent studies [13, 14], empirical relationships of the following types have been recommended ($W^{235}_{0,1}$ = initial, final enrichment):

$$\frac{Pu/U}{W_0^{235} - W_1^{235}}, \quad \frac{Pu/U}{\Delta^{236}U}, \quad \frac{Pu/U}{^{240}Pu/^{239}Pu}$$

It was found from Yankee data [13] that the first relationship could be reproduced for similar cores (e.g. same reactor, cladding material, initial enrichment, moderator to fuel ratio, fuel geometry) within 1%. Even different initial enrichments can be covered with an empirical equation of the type

$$R = \frac{Pu/U}{W_0^{235} - W_1^{235}} = a_0 - a_1 \times \text{initial enrichment}$$

Investigations on reactors other than Yankee show similar results [14, 15].

Several potentially valuable safeguards applications are:

(1) Verification of operation data by a consistency check at the reprocessing plant

The safeguards authority may evaluate operators' data with respect to the above-mentioned relationships and compare actual data with historical data which should correspond within the associated error bars if similar cores are reprocessed. The possibility of tampering with the reported input data consistently can be detected with high confidence by correlating input and product data on isotopic quantities of U and Pu. The recovery of each isotope should be equal and should correspond to historical recovery factors within accepted error bars. In addition, the so-called 'fingerprint' aspect (correlation of isotopic abundances of input and product batches, e. g. product isotopic abundances must lie within the maximum possible mixing area given by the isotope vectors of input batches) may help to detect uncontrolled blending operations.

(2) Verification of operators' data by measurement

If the safeguards authority intends to perform independent measurements for comparison of reactor batches for which such relations have already been established and the initial total uranium amount and enrichment are known, the use of this technique is advantageous, since considerable reduction in the efforts can be achieved in the case of analyses and transfer measurements.

Only relative measurements such as isotopic abundances of U and Pu are required. No isotopic dilution analysis (for the determination of plutonium amounts), with all associated problems such as spiking, sample stability, activity and transportation, is necessary, since the absolute plutonium amounts can be obtained with the help of the relation

$$Pu = U_0 \times R \frac{U}{U_0} (W_0^{235} - W_1^{235})$$

The ratio U/U_0 is a correction term and is a function of the burn-up. The value of this ratio can be established either by using very rough burn-up data ($\sim\pm$ 15%) supplied by the reactor operator or by using some other burn-up parameters, such as the amounts of fission products measured at the reprocessing plant.

The use of this relation enables the safeguards authority to dispense with the verification of tank volume and density measurements completely under certain conditions [16]. If the inspector finds an outlier in the course of the consistency check as described under (1), he should pay special attention to it. Analysis of such outliers can provide valuable additional information.

A further method of reducing efforts is provided by the composite sample technique [17, 18]. This method will permit verification of the U and Pu content and the isotopic abundances with (in principle) only one analysis without a significant reduction of the accuracy of the result, provided that the problems which are still associated with this method (e. g. aging effects) can be solved. However, an inspection regime should maintain its capability of establishing a material balance by independent measurements, particularly at this point in a fuel cycle.

4. OPTIMIZATION OF THE MATERIAL ACCOUNTING SYSTEM

To optimize safeguards efforts, the criteria for the optimization have to be defined. Among the several concepts developed so far, the more important ones are:

(1) The safeguards efforts have to be distributed in such a way that, for any facility for a given hazard value (in effective kilograms of fissile material), a given probability of detection will be reached [1, 19].
(2) The safeguards efforts have to be distributed in such a way that the operators of the different facilities are induced to act legally. This will be achieved if the expectation value of the gain in the case of any diversion is smaller than the expectation value in the case of no diversion [20, 21].

In the case of (2), some assumptions on the fraction of the pay-off parameters in case of successful (denoted as d) and unsuccessful (denoted as c) diversion have to be made.

TABLE III. REDUCTION OF SAFEGUARDS EFFORTS BY USE OF INHERENT CYCLE PROPERTIES FOR THE REFERENCE FUEL CYCLE AND FULL COVERAGE
(1 insp. yr $\hat{=}$ 1600 insp. h $\hat{=}$ 50 000 DM)

	Costs of safeguards measures (10^6 DM/yr)	Net insp. time (10^3 h/yr)	Total safeguards costs (10^6 DM/yr)
1. Conventional accountability	3.119	39.41	4.292
2. Use of S-R correlations	2.115	27.36	2.968
3. Use of S-R correlations + isotopic correlations	1.99	27.11	2.834
4. Use of S-R correlations + isotopic correlations + transparency	1.99	26.43	2.814
5. Percentage reduction from 1 to 4	36	33	35

TABLE IV. SAFEGUARDS MEASURES IN THE NUCLEAR FACILITIES IN THE REFERENCE FUEL CYCLE

Facility / Measures	Reactors	Reprocessing	U fabrication	Isotope sep.	Convers. I	Convers. II	Convers. III	Pu fabrication
1. Input Measures	1.1 Ident. and counting of sub-ass.	1.1 Ident. and counting of sub-ass. 1.2 Volume det. 1.3 Taking of samples 1.4 Mass-spectr. anal. of samples [a]	1.1 Ident. of seals of containers 1.2 Weigh. of cont. 1.3 Taking of samples 1.4 Chem. anal. of samples	1.1 Ident. of seals of cylinders 1.2 Weigh. of cyl. 1.3 Taking of samples 1.4 Chem. anal. of samples	1.1 Ident. of seals of cylinders 1.2, 1.3, 1.4 Same as is. sep. output 2.1, 2, 3	1.1 Weigh. of waggons 1.2 Taking of samp. 1.3 Chem. anal. of samp.	1.1 Ident. of seals of cont. 1.2, 1.3, 1.4 Same as repr. output 2.1, 2, 3	1.1 Ident. of seals of birdcages 1.2 Calorimetry
Time/ meas. (h)	1.1 0.25	1.1 0.5 1.2, 1.3 2 1.4 2 [a]	1.1 0.166 1.2, 1.3 0.5 1.4 1	1.1 0.166 1.2, 1.3 1 1.4 2	1.1 0.166 1.2, 1.3 1 1.4 2	1.1, 1.2 2 1.3 2	1.1 0.166 1.2, 1.3 2 1.4 2	1.1 0.166 1.2 3
Cost/ meas. (DM)	1.1 20 for full coverage	1.1 Incl. in reactor 1.2 - 1.3 10 1.4 1000 [a]	1.1 4 for full coverage 1.2, 1.3 5 1.4 150	1.1 Incl. in fabr. 1.2, 1.3 5 1.4 150	1.1 Incl. in fabr. 1.2, 1.3 4 1.4 400	1.1 Incl. in input 1.2 5 1.3 50	1.1 Incl. in fabr. 1.2, 1.3 5 1.4 400	1.1 Incl. in repr. 1.2 90
No. of meas./yr	112 × 12 = 1344	1344 sub-ass. + 14 × 12 = 168 batch.	320 × 12 = 3840	(15 + 70) × 12 = 1020	16 × 12 = 192	5 × 12 = 60	21 × 12 = 252	710
2. Output Measures	2.1 Ident. and counting of sub-ass.	2.1 Weigh. of contain. 2.2 Taking of samples 2.3 Mass-spectr. anal. 2.4 Sealing	2.1 Measurement of pins 2.2 Sealing of sub-ass.	2.1 Weigh. of cyl. 2.2 Taking of samples 2.3 Mass-spectr. anal. 2.4 Sealing	2.1 Sealing of containers 2.2, 2.3, 2.4 Same as fabr. input 1.2, 3, 4	2.1 Sealing of cyl. 2.2, 2.3, 2.4 Same as is. sep. input 1.2, 3, 4	2.1 Sealing of cyl. 2.2, 2.3, 2.4 Same as is. sep. input 1.2, 3, 4	2.1 Calorimetry (100 pins/ measurement) 2.2 Sealing of casks
Time/ meas. (h)	2.1 0.5	2.1, 2.2 U 2, Pu 2 2.3 2 2 2.4 0.25 0.25	2.1 0.166 2.2 0.5	2.1, 2.2 1 2.3 2 2.4 0.25	2.1 0.25 2.2, 2.3 0.5 2.4 1	2.1 0.25 2.2, 2.3 1 2.4 2	2.1 0.25 2.2, 2.3 1 2.4 2	2.1 3 2.2 0.5

TABLE IV (cont.)

Facility / Measures	Reactors	Reprocessing	U fabrication	Isotope sep.	Convers. I	Convers. II	Convers. III	Pu fabrication
Cost/ meas. (DM)	2.1 40 for full coverage	2.1 } 5 2.2 } 5 2.3 400 400 2.4 2 2	2.1 1 2.2 Incl. in insp.	2.1 } 5 2.2 } 5 2.3 400 2.4 2	2.1 2 2.2 } 5 2.3 } 5 2.4 150	2.1 2 2.2 } 5 2.3 } 5 2.4 150	2.1 2 2.2 } 5 2.3 } 5 2.4 150	2.1 160 2.2 Incl. in insp. costs
No. of meas./yr	112 x 12 = 1344	(21 + 100) 12 = 1452	12×10^4 pins + 1344 sub-ass.	16 x 12 = 192	320 x 12 = 3840	70 x 12 = 840	15 x 12 = 180	396
3. Waste Measures		3.1 Volume det. 3.2 Taking of samples 3.3 Chemical analysis	3.1 Volume det. 3.2 Taking of samples 3.3 Chemical analysis	3.1 Volume det. 3.2 Taking of samples 3.3 Mass-spectr. analysis	3.1 Volume det. 3.2 Taking of samples 3.3 Chemical analysis	3.1 Volume det. 3.2 Taking of samples 3.3 Chemical analysis	3.1 Volume det. 3.2 Taking of samples 3.3 Chemical analysis	3.1 Volume det. 3.2 Taking of samples 3.3 Chemical analysis
4. Inventories								
No. of inv./yr		2 (1 PI + 1 WO) [b]	2	1	2	1	1	2
Total time (h/yr)		600	1483	180	120	82	40	686
Total costs (10^3 DM/yr)		52	50	40	32	5	11	27.4
5. Other measures	(a) Collection of data (activity measure., registr. integr. power, cooling time, etc.) (b) c,d as in other facilities		(a) Calibration of measuring instruments. (b) Preparation of periodic reports on material balance. (c) Auditing of operators' reports. (d) Miscellaneous activities (checking of containment, validity of design data, etc.).					

[a] For isotope correlation, 1 h/measurement and DM 250/measurement have been assumed.
[b] PI : process inventory, WO: washout.

In Ref. [2], a comparison between the case of full coverage, that is, the case where the hazard value is reduced to the measurement errors, case (1) for a hazard value of 10 eff kg and a probability of detection of 0.95 and case (2) for a fraction d/c smaller than one has been carried out. It was shown quantitatively how the safeguards efforts were reduced by proceeding from the first case over the second to the third.

As mentioned earlier, in this paper some inherent characteristics of the fuel cycle have been used to reduce the efforts in establishing a material accountancy system without reducing the quality of the material balance established. The results are given in Table III. The unit costs and safeguards efforts which have been used to estimate the total amount of effort are given in Table IV. In arriving at the relevant figures, a number of assumptions were made:

(a) In the conventional accounting system, full measurements of fissile material have been assumed to be carried out at all the strategic points in the reference fuel cycle of Fig. 1. No sealing and identification will be carried out except for fresh sub-assemblies at the exit of a fabrication plant.
(b) To estimate the influence of shipper-receiver correlations, the fissile material measurements were assumed to be replaced by sealing and identification measures at the exit of conversion plant II and at the exits and entrances of conversion plants I and III, respectively, and at the entrance of the Pu fabrication plant. The remaining measures were the same as in the conventional method.
(c) To assess the reduction through the isotopic correlation technique, it was assumed that some of the isotopic dilution analyses at the input of the reprocessing plant were replaced by simple isotopic abundance measurements and that some of the volume and density measurements were not verified by the inspector as they were in the preceding cases.
(d) For the transparency of the information system, only the inspection hours were reduced by 30% and not the time and costs for analyses and containment measures.
(e) The additional safeguards efforts for the plutonium storage at the fabrication facility and for the waste streams at all the nuclear facilities are negligible compared with the total safeguards efforts. They have not been considered for the optimization.

It is to be noted from Table III that the total costs of the measures are reduced by 36% and the inspection efforts by 33%. However, it should be mentioned that the use of identification and seals is normal practice in safeguards even today. This component alone (point 2) causes a reduction of 32% in the costs of safeguards measures. Therefore, the actual reduction resulting from the use of isotopic correlation techniques and transparency of information is about 4% for the costs of safeguards measures and about 3% in the case of inspection efforts.

5. CONCLUSIONS

An effort has been made in this paper to investigate the influence of some cycle-inherent parameters on the optimization of a material accountancy system for safeguards purposes. It has been shown that, by using

containment measures (identification and sealing) instead of duplicating material balance measures at a number of points in the fuel cycle, a significant reduction in the efforts may be achieved. Further reductions may be obtained by using isotopic correlations in the system and making the information system transparent at the facilities. Although the use of historical data on MUF and measurement accuracy may also cause a further reduction in the efforts, collection of data on an international level is required before their use can be quantified.

A further point in this connection is the recognition of the fact that these reductions in efforts do not reduce the quality of the material balance established. The uncertainty in the material balance remains the same.

ACKNOWLEDGEMENT

The authors would like to thank W. Häfele for his interest in this work.

REFERENCES

[1] AVENHAUS, R., GUPTA, D., Effectivity and Cost Optimization of Safeguards Systems, Part 2, KFK 1101 (1969).
[2] AVENHAUS, R., GUPTA, D., "Effective application of safeguards manpower and other techniques in nuclear fuel cycles", Safeguards Techniques (Proc. Symp. Karlsruhe, 1970) 1, IAEA, Vienna (1970) 345.
[3] IAEA Document INFCIRC/153 (1971).
[4] GMELIN, W., NENTWICH, D., OTTO, H.E., Safeguard Exercise at the Fabrication Plant ALKEM, KFK 901 (1969).
[5] KRAEMER, R., et al., Beschreibung eines Kontrollexperimentes in der Wiederaufarbeitungs-Anlage Eurochemic, KFK 907 (1969).
[6] HAGEN, A., et al., Development of Safeguards Procedures for a Reprocessing Plant Similar to the WAK Type, KFK 1102 (1970).
[7] SCHRÖDER, R., et al., Development of Safeguards Procedures for Heavy Water Moderated, Cooled and Reflected Pressurized Water Type Reactors, KFK 804 (1970).
[8] Contract IAEA 790/RB, to be published.
[9] SINGH, H., Analysis of Some Available Data on Material Unaccounted For (MUF), KFK 1106 (1971).
[10] KRAEMER, R., BEYRICH, W., JEX 70, KFK 1100 (1971) Chapter 7.
[11] GMELIN, W., et al., Safeguards Measures and Efforts in Conceptual Fabrication Plants for Uranium and Plutonium Containing Fuel Elements, KFK 910 (1969).
[12] HAGEN, A., HERRE, F., GUPTA, D., Design Criteria for Reprocessing Plants, KFK 1107 (1970).
[13] CHRISTENSEN, D.E., et al., The Safeguards Value of Chemical Plant Measurements Relating to Burn up – Yankee Cores V and VI, BNWL-1473 (1970).
[14] MOEKEN, H.H.P., Some developments in input accountability at Eurochemic, Safeguards Techniques (Proc. Symp. Karlsruhe, 1970) 1, IAEA, Vienna (1970) 551.
[15] KRAEMER, R., BEYRICH, W.: JEX 70, KFK 1100 (1971) Chapter 6.
[16] HÄFELE, W., NENTWICH, D., "Modern safeguards at reprocessing plants and reactors", Safeguards Techniques (Proc. Symp. Karlsruhe, 1970) 1, IAEA, Vienna (1970) 3.
[17] JUMP, M.J., et al., Evaluation of Methods for Input Accountability in a Reprocessing Plant – Preparation and Analysis of Composite Samples, NFS-AL-127 (Dec. 1967).
[18] AVENHAUS, R., et al., Verification of Input Analysis of a Reprocessing Plant by Means of Composite Sample Technique, KFK 1104 (1970).
[19] STEWART, K.B., "A cost effectiveness approach to inventory verification", Safeguards Techniques (Proc. Symp. Karlsruhe, 1970) 2, IAEA, Vienna (1970) 387.
[20] BIERLEIN, D., Direkte Überwachungssysteme, Operations Research Verfahren 6 (1968) 57-68.
[21] BEINHAUER, R., BIERLEIN, D., "Game theoretical models for inspection procedures", Safeguards Techniques (Proc. Symp. Karlsruhe, 1970) 2, IAEA, Vienna (1970) 425.

A TECHNICAL BASIS FOR INTERNATIONAL SAFEGUARDS

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Abstract–Résumé–Аннотация–Resumen

A TECHNICAL BASIS FOR INTERNATIONAL SAFEGUARDS.

Based on the consideration of light-water reactor fuel cycles of different levels of completeness, the major safeguards problems are singled out for the fuel cycle which includes thermal recycle of plutonium, namely verification that all plutonium produced in a power reactor reaches the reprocessing plant and is accounted for there; verification of plutonium quantities in fabrication facilities; verification of large amounts of plutonium in sealed stores or other contained uses, such as fast critical facilities.

It can be shown that these problems will require by far the greater part of the safeguards inspection efforts, notwithstanding the remaining requirements for verification of uranium in conversion and fabrication plants and reactors, under the fundamental assumption of absence of enrichment facilities. The paper presents the technical safeguards objectives and the resulting verification requirements of an international system to meet the obligations arising from the Non-Proliferation Treaty. The paper considers the verification procedures which can be applied by the international system to material flows and inventories of the three types of facilities, using independent measurements complemented by containment and surveillance measures to achieve quantified accountability levels. The technical impact of the desired level of accountability, the amounts of flow and inventory, the point and the use of statistical sampling on the allocation of inspection effort is considered. The paper finally points out safeguards techniques that need to be developed, in particular in connection with independent measurements, the application of item accountability based on reactor fuel assemblies and the use of isotopic correlation data.

LES BASES TECHNIQUES DE L' APPLICATION DE GARANTIES INTERNATIONALES.

En partant d' une analyse de cycles du combustible plus ou moins complets pour réacteurs à eau légère, les auteurs cernent les principaux problèmes de garanties indiqués ci-après pour le cycle du combustible qui comprend le recyclage du plutonium dans des réacteurs thermiques: vérifier que tout le plutonium produit dans un réacteur de puissance parvient à l' usine de retraitement et qu' il n' y a pas d' écart entre les quantités produites et les quantités reçues; vérifier les quantités de plutonium dans les installations de fabrication; vérifier les grandes quantités de plutonium contenues dans des magasins sous scellés ou dans d' autres installations où elles sont confinées, telles que les assemblages critiques à neutrons rapides.

On peut montrer que ces vérifications constitueront de loin la plus grande partie des activités d' inspection, même compte tenu des autres tâches qu' impose la vérification de l' uranium dans les usines de transformation et de fabrication et dans les réacteurs, si l' on fait l' hypothèse fondamentale d' une absence d' usines de séparation. Les auteurs présentent les objectifs techniques des garanties et les opérations de vérification qu' exige un système international visant à permettre à l' Agence de s' acquitter des obligations qui découlent pour elle du Traité sur la non-prolifération. Ils examinent les modalités de vérification que l' on peut appliquer aux flux et aux stocks de matières des trois types d' installations, en faisant appel à des mesures indépendantes, complétées par le confinement et la surveillance, pour atteindre des objectifs comptables définis quantitativement. Ils examinent les incidences techniques de l' objectif fixé, des quantités de matières dans le flux et les stocks et de l' utilisation de l' échantillonnage statistique, sur l' allocation de moyens d' inspection. Pour conclure, ils indiquent les techniques de garanties qui doivent être étudiées, notamment en ce qui concerne les mesures indépendantes, l' application de la comptabilité par article fondée sur les assemblages combustibles et l' utilisation de la corrélation avec les données isotopiques.

ТЕХНИЧЕСКАЯ ОСНОВА ДЛЯ ПРИМЕНЕНИЯ МЕЖДУНАРОДНЫХ ГАРАНТИЙ.

Ниже перечисляются основные проблемы по гарантиям, основанные на рассмотрении топливных циклов различных уровней законченности разработки легководного реактора, выделенные для топливного цикла с повторным использованием плутония: проверка того, что весь плутоний, произведенный в энергетическом реакторе, доходит до завода по переработке топлива и учитывается там; проверка количества плутония на установках по изготовлению топлива; проверка больших количеств плутония в опечатанных складах или других применяемых установках, таких, как быстрые критические сборки.

Можно показать, что эти проблемы потребуют гораздо больших инспекционных усилий по гарантиям, несмотря на остающиеся требования к проверке урана, находящегося на заводах по обработке и изготовлению топлива, и на реакторах, при основном предположении, что отсутствуют обогатительные установки. В докладе излагаются технические цели гарантий и вытекающие из этого требования международной системы по проверке, для того чтобы соответствовать обязательствам, вытекающим из Договора о нераспространении ядерного оружия. В докладе рассматриваются процедуры проверки, которые могут применяться международной системой к потоку материала, и инвентаризация на трех типах установок с применением независимых измерений, дополненными мерами по сохранению и наблюдению для достижения учетных уровней определенных количеств. Также рассматривается техническое влияние желаемого уровня учета, количества потока и инвентарного количества, места и применения статистического взятия проб на размещение инспекционных усилий. И, наконец, в докладе указываются методы гарантий, которые необходимо разрабатывать, особенно в связи с независимыми измерениями, приминение отчетностей по пунктам, основанное на топливных сборках реактора и применении данных по изотопной корреляции.

FUNDAMENTOS TECNICOS DE LAS SALVAGUARDIAS INTERNACIONALES.

Basándose en el estudio de los ciclos del combustible de los reactores de agua ligera, completos en distinto grado, se abordan individualmente los siguientes problemas capitales referentes a las salvaguardias en el caso del ciclo del combustible que comprende el reciclado térmico del plutonio: la verificación de que todo el plutonio producido en un reactor de potencia llega a la planta de reelaboración y se contabiliza en la misma; la verificación de las cantidades de plutonio en las instalaciones de fabricación; la verificación de grandes cantidades de plutonio en instalaciones de almacenamiento precintadas, o utilizadas en otras condiciones de confinamiento como, por ejemplo, en instalaciones críticas rápidas.

Puede demostrarse que esos problemas exigirán, con mucho, la mayor parte del volumen de las actividades de inspección relacionadas con las salvaguardias, pese a las restantes necesidades de verificar el uranio en las plantas de transformación y de fabricación y en los reactores, partiendo siempre del supuesto básico de la ausencia de instalaciones de enriquecimiento. En la presente memoria se exponen los objetivos técnicos de las salvaguardias y los consiguientes requisitos de verificación que debe reunir un sistema internacional para satisfacer las obligaciones dimanantes del Tratado sobre la no proliferación. Se examinan los procedimientos de verificación que pueden aplicarse en este sistema al movimiento y existencias de materiales en los tres tipos de instalaciones, sirviéndose de mediciones independientes complementadas por medidas de contención y vigilancia para lograr resultados cuantitativos. Se consideran asimismo las consecuencias técnicas que sobre el volumen que deben alcanzar las actividades de inspección imponen el grado de contabilización deseado, la magnitud del movimiento y de las existencias de materiales, el punto en que se efectúe el muestreo estadístico, y el uso que se haga del mismo. Finalmente, se señalan las técnicas de salvaguardia que han de desarrollarse, en especial en relación con las mediciones independientes, con la aplicación de la contabilización por partidas basada en los conjuntos de combustible de los reactores, y con el empleo de los datos de correlación isotópica.

1. INTRODUCTION

The objective of IAEA safeguards has been defined [1] as "the timely detection of diversion of significant quantities of nuclear material from peaceful nuclear activities .....". It has been agreed that this objective be attained by "..... the use of material accountancy as a safeguards measure of fundamental importance with containment and surveillance as important complementary measures" and that "..... the technical conclusion of the Agency's verification activities shall be a statement, in respect of each material balance area (MBA) of the amount of material unaccounted for (MUF) over a specific period giving the limits of accuracy of the amounts stated".

The terms of primary importance for the technical implementation of Agency safeguards are:

(a) 'Timely'. This term leads to requirements concerning the timings of provision of reports, the closing of the material balances, and the allocation of inspections.

(b) 'Detection'. This term means the ability to distinguish between the normal causes of MUF and large losses of material which could be diversions.

(c) 'Significant quantities'. This term requires, in conjunction with a definition of the quantified 'levels of accountability', an analysis to determine the 'limits of accuracy' actually achieved in various situations with a given level of verification effort.

In this paper a complete light-water reactor cycle (except isotope separation) has been chosen as an example to demonstrate the application of Agency safeguards. This example involves those facilities which will constitute a major part of the safeguards work-load for the Agency in the Non-Proliferation Treaty (NPT) situation. While other facilities, e.g. research centres and enrichment plants, may present different problems, the safeguards procedures described here should be typical for these other facilities and the safeguards objectives and general requirements will remain the same.

All Agency safeguards applications involve the following fundamental elements: provision and review of design information, maintenance of accounting and operating records, the provision of accounting reports and inspections. These elements, with detailed requirements and constraints, are defined in the NPT Agreement [2] and have been described and analysed in another paper presented at this Conference [3]. This paper will thus concentrate on the verification procedures needed to meet the stated safeguards objective. In general, only the normal inspection situation is described, in which there is no particular reason to believe that significant material quantities have been lost.

2. REFERENCE FUEL CYCLES

For the purpose of this paper, a fuel cycle has been analysed (see Fig. 1) consisting of light-water reactors (LWR) together with a uranium conversion/fabrication plant, a reprocessing plant, a plutonium storage and a mixed plutonium-uranium oxide fabrication plant for the production of recycle fuel. It is assumed that isotope separation is carried out in another State. The data for these facilities are given in Table I and a flow diagram is given in Fig. 1.

The assumed designation of material balance areas and key measurement points (KMP) are shown in simplified form in Table II and the following remarks can be made:

(a) An LWR comprises one MBA only with three key measurement points identified therein.

(b) The conversion and fabrication plant consists of two MBAs: a feed storage (shipper-receiver difference) MBA, where the input material (UF_6) is kept on shipper's values, and a process (MUF) MBA, comprising the remaining part of the plant; five KMPs for flow verification are designated. It may be noted that the verification of the product accountability would be carried out at two points, C and D. At C the weights and concentrations would be verified whereas at D the necessary supplementary containment measures would be carried out as indicated in Table IV. This is due to the momentary lack of sufficiently accurate and reliable non-destructive technique (NDT) instruments. Once these instruments are

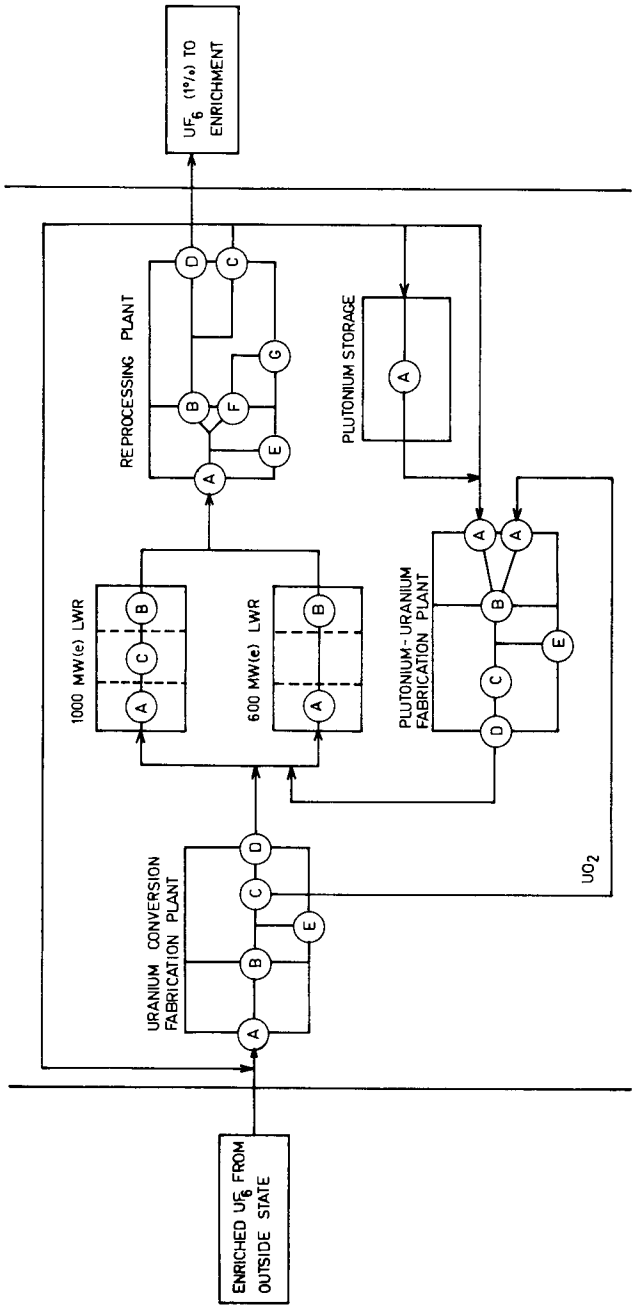

FIG. 1. Structure of LWR fuel cycle.

TABLE I. DATA OF FACILITIES USED IN REFERENCE CASE[a]

	LW reactors 600 MW(e) BWR	LW reactors 600 MW(e) PWR	LW reactors 1000 MW(e) BWR	LW reactors 1000 MW(e) PWR	Uranium conversion fabrication		Reprocessing		Plutonium storage	Mixed oxide fabrication	
INPUT											
Type of item	Assemblies				Cylinders		Assemblies		Bird cages	Drums	Bird cages
Items per year	150	50	225	64	170	23	540 - 1815		544	23	544
Tonnes or kg per item					1.5 t	1.5 t			4 kg Pu	1.5 t	4 kg Pu
Total tonnes or kg	19 t	19 t	38 t	29 t	225 t U	34.5 t U	250 t U	2200 kg	2178 kg Pu	34.5 t U	2178 kg Pu
% enrichment	2.65	3.0	2.0	2.7	3.0	Depl.	1			Depl.	
Material	UO_2	UO_2 or	UO_2	UO_2	UF_6	UF_6	UO_2	Pu	PuO_2	UO_2	PuO_2
Total tonnes UO_2	16	16	33.2	24.2							
Total tonnes PuO_2/UO_2	2.9	2.9	4.8	4.8							
% Pu	6	6	6	6							
OUTPUT											
Type of item	Same as input UO_2 reduced by burn-up				Assemblies	Drums	Cylinders	Bird cages	Same as input	Assemblies	
Items per year					540-1815	23	165	544		95-340	
Tonnes or kg per item						1.5 t	1.5 t	4 kg Pu		2158 kg	
Total tonnes or kg					225 t U	34.5 t U	247.5 t U	2178 kg Pu		2158 kg	34.1 t
% enrichment					3.0	Depl.	1			6	Depl.
Material					UO_2	UO_2	UF_6	PuO_2		PuO_2	UO_2
Pu (kg)/year	180	180	300	300							
Discards (kg)/year					100		2500 U	22 Pu		20 Pu	370 U
Annual throughput Ekg[b]					48		2225			2189	
Working days	365				220 (2 shift)		350 (3 shift)			250 (2 shift)	
Frequency of physical inventory taking per year	1				1		2 (wash-out)			2 (clean-out)	

[a] Reference case throughput is assumed to be 255 tonnes uranium.

[b] Ekg = effective kilograms.

TABLE II. DESIGNATION OF KMP, BATCHES AND MAXIMUM ROUTINE INSPECTION EFFORT FOR REFERENCE CASE

	LWR	Uranium conversion and fabrication	Reprocessing	Pu-storage	Mixed oxide fabrication plant
Number of KMPs for flow verification (FKMP)	2	5	7	1	5
Location of FKMP	Fresh fuel storage (A) Spent fuel storage (B)	Storage of UF_6 cylinders (A) Feed point UF_6 (B) Loading point pellets (C) Storage assemblies (D) Storage discards (E)	Spent fuel storage (A) Input accountability (B) Hull measurements (E) Waste transfer point (F) Product load-out Pu (C) Product load-out U (D) Waste accountability (G)	Whole storage (A)	Storage for input (A) Input accountability (B) Loading point pellets (C) or before assembling (C) if NDT Storage of assemblies (D) Waste storage (E)
Number of KMPs for inventory verification (IKMP)	3	about 5	7	1	5
Location of IKMP	Same as FKMP plus core (C)	Cannot be determined in advance	Same as FKMP (clean-out only)	Same as FKMP	Same as FKMP (clean-out only)
Input batch	All assemblies for one refuelling	1 UF_6 cylinder	1 assembly, 1 filling of input accountability tank	1 bird cage	1 bird cage total amount of depleted U/month
Output batch product	1 assembly	Each shipment	1 bird cage 1 UF_6 cylinder	1 bird cage	1 assembly
Output batch waste		Total amount of waste in one month	Total amount of different wastes in a month		Total amount of waste in a month
Maximum routine inspection effort (man-days/year) single plant	50	179	1415	50	1374

available, these weights and concentrations would be verified at a point where the fuel rods or the assemblies are finished.
The KMPs for inventory taking (IKMP) can hardly be fixed in advance, since their location depends on the procedures of the operator's physical inventory taking. Based on the results of a recent integral test, the number of points can be estimated to be about five.

(c) The reprocessing plant comprises two MBAs: a feed storage (shipper-receiver difference) MBA and a process (MUF) MBA; seven KMPs are designated. The IKMPs are, in the case of a wash-out of the plant, the same as the FKMPs (Flow Key Measurement Points). If, however, the inventory taking is done without complete clean-out of the plant, the appropriate accountability tanks would be designated as IKMPs for this inventory taking.

(d) The storage comprises one MBA where item accountability and other containment measures may be applied and the whole area of this storage may be regarded as one KMP.

(e) The mixed-oxide fabrication plant comprises two MBAs: a feed storage (shipper-receiver difference) MBA and a process (MUF) MBA; five KMPs are designated. If, however, instruments are available which enable the measuring of fuel rods or fuel assemblies, the verification of the plutonium concentrations and the weight of the pellets would not be needed. The KMPs for inventory taking are, in the case of a clean-out of the process, the same as the FKMPs. If, however, the inventory taking is carried out by partial clean-out of the process, the appropriate inventory locations where material remains would be designated as the IKMPs for the time of this inventory taking only.

The batches identified for safeguards purposes are shown in Table II. These batches are the units for the inventory change reports to be provided to the Agency. The size of the inventory batches normally cannot be determined in advance but depends on the actual situation.

3. DESCRIPTION OF VERIFICATION PROCEDURES

The purpose, scope and types of inspections are defined in the Agreement and possibilities for the technical implementation thereof are discussed in the following paragraphs.

Following Articles 76 to 82 of the Agreement, the inspections at facilities are to be carried out under two types of limitation: access limitations and limitations in the inspection effort. In respect of the access limitations to the strategic points and to the records, procedures for effective verification during inspections have been developed and described in several papers [4]. The Maximum Routine Inspection Effort (MRIE) can be calculated for facilities according to Article 80 of the Agreement and is shown in Table II. With this upper limit, an actual inspection effort may be determined taking into account the four criteria stated in Article 81. For the purpose of the technical implementation of Agency safeguards it is necessary to define and specify the lower limit of routine inspection effort which may be called the 'Lowest Routine Inspection Effort' (LRIE) required for effective Agency safeguards. The LRIE can be generally defined to be that lower limit of inspection effort which enables the Agency to make a statement (see Fig. 2) in respect

of each MBA, of MUF and its related limits of error over a specific period of time under the ideal conditions that:

(a) the Agency is able to make maximum possible use of the State's system of safeguards according to the criteria laid down in Article 81 of the Agreement, i.e. the form of nuclear material, the effectiveness of the State's accounting and control system, characteristics of the State's fuel cycle, international independence and other technical developments;

(b) the State fulfils all Agency information requirements by provision of design information, maintenance of records and timely provision of reports.

According to this definition of the LRIE the following observations can be made:

(a) The actual routine inspection effort will be higher than the LRIE whenever the conditions mentioned above are not fulfilled.

(b) If the actual routine inspection effort is below the LRIE the Agency will not be able to make a statement that effective safeguards have been applied.

(c) The specification of the LRIE is to be made separately for each particular facility taking into account the actual situation.

In the following paragraphs verification procedures to be applied during inspections are described and figures for the LRIE are evaluated for each of the facilities considered. The figures given are preliminary estimates based on the Agency's experience and publications and must be improved in the course of the future application of Agency safeguards. This provides one of the reasons for the need to carry out integral tests. During such integral tests, an investigation has to be made, for instance, of whether the LRIE required for the verification of the input in a LWR can be assumed to be four man-days per year only, as shown in Table III.

3.1. Light-water reactors

The safeguards procedures to be carried out at LWRs are summarized in Table III and are designed to enable the Agency to meet two objectives:

(a) detection of unauthorized removal of single fuel sub-assemblies within the limit of the level of accountability of the nuclear material by application of item accountability;

(b) acquisition of calculated burn-up, nuclear loss and production, and isotopic data for the purpose of application of isotopic correlations which are particularly useful for the safeguards of the reprocessing plant. It may be noted, however, that these data are not verified at the reactor by reviewing the burn-up calculations, etc., but the data are simply taken from the records or reports.

In Table III an estimate has been made for the LRIE, as mentioned above in Sections 3 and 3.1, which turned out to be 23 man-days per year. This figure has been estimated under the following assumptions:

(a) Only one shutdown with subsequent opening of the vessel takes place during a year. For each additional shutdown an additional 12 days are required.

(b) No seal is damaged in the intervals between inspections.

(c) The records and reports are kept and provided according to the agreed requirements.

TABLE III. SUMMARY OF VERIFICATION PROCEDURES FOR LW REACTORS

Principle of verification: item accountability
Time between re-fuelling: 12 months
Time for re-fuelling: 21 days
MRIE: 50 man-days/year

Event	Task	LRIE
After receipt of fresh fuel (one visit)	Removal of seals at assemblies and identification, records audit, check of seal at vessel, routine identification of irradiated assemblies, maintenance of camera	4
After shut-down but before re-fuelling	Removal of seal at vessel; identification and counting of fuel at reactor vessel, fresh fuel storage and spent fuel storage; records audit; maintenance of camera	6
After re-fuelling but before start-up	Identification and counting of fuel at vessel and storages, fixing of seal to vessel, maintenance of camera, records audit	6
Intermediate inspections (three visits)	Identification and counting of fuel at storages, check of seal at vessel, records audit, maintenance of camera	3
After completion of shipment of irradiated fuel	Identification and counting of fuel at storages, check of seal at vessel, records audit, isotopic data acquisition; maintenance of camera	4
TOTAL	7 inspections/year	23 man-days/year
% of MRIE		46

(d) The fuel assemblies can be uniquely identified either directly by appropriate seals or instruments or indirectly by surveillance cameras.

It may be noted that the stated inspection effort is independent of the reactor power, i.e. the same effort is required for 600-MW(e) and 1000-MW(e) reactors.

3.2. Conversion and fabrication plant

Two types of verification procedure would be applied at the uranium conversion and fabrication plant, those for flow verification and those for

TABLE IV. PROCEDURES FOR FLOW VERIFICATION AT A CONVERSION AND FABRICATION PLANT FOR LOW ENRICHED MATERIAL
Material: UO_2 - 3% enriched
Maximum Routine Inspection Effort (MRIE): 175 man-days/year

Flow	From	KMP	Batch type	Number of batches/year	Verification procedures	LRIE (man-days/year)
Input	Enrichment	A, B	UF_6 cylinders	100	Records audit, identification of cylinders and removal of seals,	5
					evaluation of SRDs	7[a]
						0[b]
Discards	Process	E	Drums	330	Records audit, sealing of drums plus measuring of some drums by gamma spectrometry	0[c]
						0
Output	Process	D	One shipment within a reactor load	10 reactor loads per year (19 to 34 t UO_2 each)	Records audit, identification and sealing of assemblies	10
						12
		C			Records audit, sampling of 8 pellets from each re-load, maintenance of automatic scale	5
						50[d]
Others	Review of operator's measurement quality control data					5
Total flow						94
LRIE needed for physical inventory taking[d]						30
Total						124
% of MRIE						69

[a] 0.5 hours/cylinders. [b] Not to be carried out at plant. [c] Included in input, output and physical inventory effort. [d] See text.

inventory verification. The flow verification procedures are described in Table IV and the following remarks can be made:

(a) For the verification of the input stream, no sampling, weighing and chemical analysis would be carried out in the presence of inspectors but only the identification of the UF_6 cylinders, the removal of seals and the necessary records audit. This procedure is based on the assumption that the measurement data of both shipper and receiver can be considered as mutually independent. Bias or diversions can be detected by the evaluation of the shipper-receiver difference using standard statistical techniques.

(b) The verification of the product would be carried out at KMPs C and D. At C the pellets are loaded into the rods and D is the exit point of the plant. The verification at C would require the major fraction of the verification activity according to the following:

(i) The overall objective is to enable the inspector to verify the UO_2 product: the weights, the concentration of uranium in the UO_2, and the isotopic composition.

(ii) The weight can be verified by using the print-outs of automatic scales, and the design of these scales is assumed to be such that a tamper-resistant print-out would be made for the inspector recording each transfer.

(iii) For verification of the uranium concentration, use can be made of the fact that this concentration normally varies within narrow limits of tolerance for each re-load of a reactor. According to data available to the Agency, it is assumed that the concentration of uranium in the UO_2 may vary between 88.06 and 88.14%. The aim of the verification of that concentration is thus to verify that within a re-load of a reactor the average concentration lies de facto between these assumed limits. The number of pellets to be sampled can be determined using standard statistical techniques.

For the plant considered in this paper the number of pellets to be sampled from each re-load would be eight. The man-days required to observe this sampling and weighing for the assumed throughput of ten re-loads have been estimated to be 50 man-days per year. This takes into account, firstly, that, even if only 1 man-hour will be spent at the facility, a full man-day will be accounted for, and, secondly, that this sampling can take place parallel with the records audit and calibrations carried out at the other key measurement points.

(c) The verification of the discards assumed to be assembled in 330 drums per year would consist of records audit, selection of a random sample for gamma measurements of the selected drums, and sealing of all drums. According to the Agency's experience, no additional inspection man-days are required for this activity.

The verification of the physical inventory taking would consist of observing the physical inventory taking as carried out by the operator and the taking of samples on a random basis. Based on recent integral tests, the manpower needed is estimated to be at least 30 man-days.

It may be noted that a reduction of the above-mentioned inspection effort required for effective safeguards may be achieved in this type of plant once advanced and automated NDT instruments are available for the determination of the product stream. In the absence of such instruments, the LRIE required for verification of the nuclear material in this type of facility will thus be at least 124 man-days per year.

TABLE V. PROCEDURES FOR VERIFICATION AT A REPROCESSING PLANT

Material: Input spent fuel, output PuO_2 and UF_6
MRIE: 1415 man-days/year

Flow	From	KMP	Batch type	Number of batches/year	Verification procedures	LRIE
Input	Reactor	A	Assembly	540-1815	Records audit; identification and counting of fuel; acquisition of isotopic data	0[a]
Input	Spent fuel storage	B	1 input tank filling	250	Records audit; observation of volume measurements and sampling; analysis of samples on random basis; evaluation of isotope correlation; observation of calibration; review of quality control program. One activity per day assumed.	0[a]
Pu-output	Process	C	1 bird cage	544	Observation of weighing and sampling; analysis of sample from each bird cage on random basis; sealing of bird cages. Two activities per day assumed.	750 man-days per year assuming 3 shift operation[b]
U-output	Process	D	1 cylinder	169	Records audit; sealing and identification of cylinders.	0[a]
Waste-output	Process	E, F G	Unspecified	Unspecified	Records audit; observation of operator's measurements	0[a]
Total LRIE - Flow						750
LRIE - Inventory[b]						14
Total for facility						764 man-days/year
% of MRIE						54

[a] Covered by the 750 man-days/year.

[b] See text.

3.3. Reprocessing plant

The verification procedures of the nuclear material in the reprocessing plant can be sub-divided again into those for flow verification and inventory verification. The verification procedures are given in Table V and the following remarks can be made:

(a) For the verification of the input, use is made of isotopic correlations in conjunction with observation of the volume measurements and sampling (to ensure representative sampling) as carried out by the operator. From each dissolver batch, one duplicate of the operator's sample is taken for the inspector. The inspector would keep these samples and would select those to be independently analysed, during random tables.

(b) For plutonium product, a fraction or duplicate of the operator's sample of each batch would be taken for the inspector. From these samples, a random selection would be made for independent analysis.

(c) For the determination of the LRIE, it should be noted that at least one inspection activity is needed for the dissolver, product, and waste batches. Assuming at least one input, product or waste verification activity per shift, 3 man-days per day would be required for the flow verification. During that time the verification of the wastes can also be carried out. The LRIE is estimated to be 759 man-days per year for flow verification.

Two types of inventory taking are considered: firstly, clean-out of the plant at intervals of about half a year, and, secondly, inventory taking by the 'isotope step function technique'. The procedures for verifying the clean-out of the plant would be:

(a) Review of the clean-out procedures to be used.

(b) Verification of the resulting effluents washed out from the process.

During the time interval between clean-outs, additional inventory takings would be arranged using the isotopic step function technique if the differences in the isotopic compositions between the different reactor campaigns are significant. For this paper, it has been assumed that about six inventory takings per year of this kind could be arranged. In addition to the randomly selected samples to be analysed for the normal flow verification, a sample from the batches immediately preceding and following the isotopic step change would have to be analysed. Further, a certain operating mode during the passage of the isotopic step would be arranged with the operator in order to keep the mixing of the material of the different campaigns at a minimum level.

The inspection effort for inventory verification can be estimated to be 7 man-days in the case of wash-out and 0 man-days in the case of inventory taking by means of the isotopic step function technique. The LRIE therefore adds up to 764 man-days per year for this reprocessing plant.

3.4. Plutonium storage

The procedures applied for the verification of the nuclear material in plutonium storage are based on item accountability. These procedures would consist of records audit and identifying and counting sealed bird cages containing plutonium. Assuming that not more than 2 man-days are required for item accountability for all items in the storage, the estimate of the total number of man-days required for the verification of the storage

TABLE VI. PROCEDURES FOR FLOW VERIFICATION AT A MIXED OXIDE FABRICATION PLANT

Material: UO_2 and PuO_2
MRIE: 1374 man-days/year

Flow	From	KMP	Batch type	Number of batches/year	Verification procedures Accountability system based on		LRIE
					Sampling and chemical analysis	NDT measurements	
Input-Pu	Reprocessing	A	Bird cage	544	Records audit; removal of seals; observation of operator's weighing and sampling; analysis of samples taken on random basis for verification of operator's measurement accuracy; review of quality control program	observation of calorimetric measurements; checking of calibrations; sealing of calorimeter and neutron counter	500 man-days/year
					about two activities per day		
Input-U	Fabrication plant	A	Drums	28	Records audit		0[a]
Output-pellets	Process	B	Amount of pellets corresponding to 1 assembly (8 or 26 kg Pu)	76 PWR or 255 BWR	Verification of weighing, taking 3 pellets per batch, records audit. One activity per day.		0[a]
Output-rods	Process	C	Number of fuel rods corresponding to an assembly	76 PWR or 255 BWR		Observation of calorimetric measurements and neutron measurements, calibration checks, sealing of instruments. About one activity per day.	0[a]

TABLE VI (cont.)

Output-assemblies	Process	D	1 Fuel assembly	76 PWR or 255 BWR	Records audit, sealing and identification. One activity per day.	0[a]
Output waste	Process	E	Total amount of waste during a month	ca. 200-400 drums	Records audit, observation of measurements, checking of calibration. About one activity per day.	0[a]
Total LRIE - Flow						500
LRIE - Inventory[b]						14
Total LRIE						514 man-days/year
% of MRIE						37

[a] Assumed to be carried out during input verification.

[b] See text.

is about 16 to 20 man-days per year. The same time intervals apply as for the reprocessing plant.

3.5. Mixed oxide fabrication plant

The verification procedures for the plutonium fabrication plant again consist of those for flow verification and those for inventory verification. The flow verification procedures are summarized in Table VI. It is shown there that one of two different types of procedure may be applied, depending on the operator's measurement system and the safeguarding technology, i.e. either procedures following the operator's weighing, sampling and chemical analysis or procedures to verify NDT measurements. The former procedures are based on the following objectives:

(a) Verification of the operator's measurement uncertainty which can be done by random sampling and analysis and review of the quality control program. This random sampling would be carried out by taking a fraction or duplicate of the operator's sample for each input batch for the inspector. The inspector would randomly select from these samples for analysis.

(b) Verification of the product output as mentioned for the product of the uranium fabrication plant in Section 3.2.(b). Assuming that an automatic scale is used at the point where the pellets are loaded into the fuel rods, the inspector would require the print-outs of the weights and arrange for sampling of three pellets from each batch (8 or 30 kg plutonium per batch) to verify the concentration of plutonium in the mixed oxide.

The safeguards procedures used when the operator's accountancy system is based on NDT techniques is also given in Table VI. The following remarks can be made:

(a) In using NDT techniques, stress is mainly placed on verification of the calibration runs (to avoid biases) and on obtaining accurate isotopic data. The actual measurements can be assumed to be automatically recorded.

(b) The actual verification procedure as applied by the inspector varies with the NDT instruments available. The procedures given in Table VI may be regarded as an example only.

When comparing the different procedures for verification of the plutonium fabrication plant, the main advantage of the accountancy system with NDT techniques is that, except for the preparation of standards, no independent sampling and expensive analyses are required. The accuracy of MUF is not expected to be different between the different procedures and it may be noted that, when the operator's accountancy system is based on NDT techniques, better cost/effectiveness of Agency safeguards can be obtained. As far as the inspection effort is concerned, no difference between the procedures is expected, since the number of inspector's activities per day needed for sampling in the first case mentioned will approximately match those needed for the calibrations of the instruments in the second case. The inspection effort required for effective safeguards of the material flow has been estimated on the basis that, for measurements of the input and the product, at least 2 man-days per day are required, assuming two shift operations. Two input batches and one output batch would be processed per day and at least one inspector's activity is required per batch.

The physical inventory taking is assumed to consist of clean-outs of the lines where plutonium is processed, identification of the bird cages

containing plutonium and audit of the depleted uranium kept in the feed storage MBA. Clean-outs are normally performed after the end of a particular campaign, and thus the verification activities consist of weighing, sampling and analysing the scraps and clean-outs and identifying the items kept in storage as applicable. According to experience, these activities would require at least 10 additional man-days. For this facility, the LRIE adds up to 510 man-days per year.

4. LEVEL OF ACCOUNTABILITY

Material accountancy, which leads to the closing of material balances in the majority of material balance areas by a complete physical inventory taking, is the measure of fundamental importance for Agency safeguards [5]. The result of a material balance over a given period of time and for a particular MBA can be expressed by MUF and its related limits of accuracy. Limits of accuracy of MUF could be defined in several ways. In this paper, the accuracy of MUF is defined as the (absolute) standard deviation (σ_{MUF}) comprising the random and systematic measurement error components propagated over the time interval between two physical inventory takings. The limits of accuracy are consequently defined as the confidence limits of MUF at a chosen confidence level. From the design information provided to the Agency, the expected standard deviations of the batches and of MUF are known. The actual standard deviation, S_{MUF}, would be verified by the comparison with the expected standard deviations and by the inspection procedures as described in former paragraphs in conjunction with the verification of MUF itself. Thus MUF and S_{MUF} denote verified numerical values. The evaluation and analysis of MUF can be carried out following the different branches of the structure presented in Fig. 2 with the aim of detecting unusual losses or diversions of significant quantities.

As shown in Fig. 2, two kinds of input are required for the evaluation of MUF: firstly, the results of the material balance MUF and S_{MUF}, and, secondly, the information block which has been designated 'level of accountability'. The level of accountability consists of four components, two probabilities, a threshold amount and a time interval. The following general rules can be given for the quantification of these components:

(a) The two probabilities are the confidence level and the power or probability of detection associated with the errors of type I and II, respectively, as used in standard statistical inference techniques. These define both the risk of a false statement in the case of no material loss and the probability of detection of a given amount lost. Efforts have been made to introduce game theory and other statistical techniques, but, as a result, the quantification of the essential probabilities seems obscured by the problem of assuming pay-off functions. The simple approach suggested here has proved so successful in several integral tests that it is proposed to use it in future with a 95% confidence level or 5% probability of the type I error and at least 95% detection probability (type II probability 5%) for a given threshold amount.

(b) The quantification of this threshold amount will give different numerical values depending on whether item accountability can be used or not:

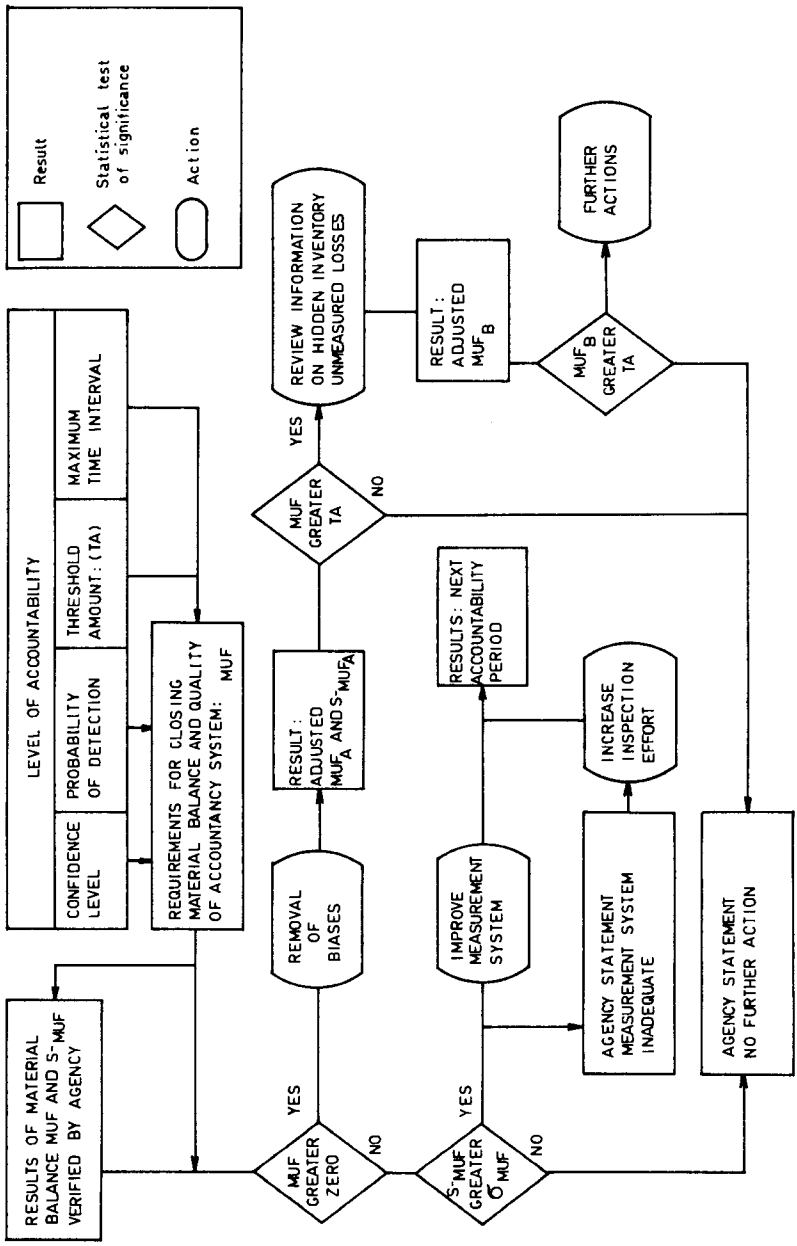

FIG. 2. Evaluation of MUF

(i) If accountability of sealed or otherwise identifiable items with proven integrity can be carried out in an MBA, no measurement error should occur and consequently the threshold amount should be one item and the probability of detection will come out as 100% for that item. This applies, for instance, to LWR fuel assemblies, sealed items in storage and normally in MBAs where shipper-receiver differences occur.

(ii) In a process MBA the threshold amount, together with the two probabilities, will define the uncertainty in the MUF which can be accepted as adequate for international safeguards. One approach[1] is that the threshold amount should be of the same order of magnitude as the fast critical masses for plutonium and uranium-235, i.e. 8 kg and 25 kg [6], respectively. These values are used in this paper. Thus the expected standard deviation for MUF, obtainable from the operator's routine accountability measurements, must be checked for each particular case during the examination of the design information, and the actual MUF and S_{MUF} obtained during operation must be verified by the inspection procedures. Besides the quality of the measurement system, the operator's proposed timing of physical inventory takings in order to close the material balance is of particular importance, since the uncertainty in MUF is influenced by the frequency of inventory taking.

(c) As stated earlier, timely detection is one of the safeguards objectives and thus a maximum time interval must be part of the level of accountability. This requirement for a time interval determines the minimum frequency of physical inventory taking where the throughput or inventory is not small in comparison with the threshold amount. The need to define a time interval has led to the concept of critical time, which was defined as a function of the physical and chemical state of the nuclear material. Several problems have arisen from this concept. For plutonium or highly enriched uranium, a critical time of ten days has been proposed; this would then require a physical inventory taking every ten days for MBAs processing these materials and such a verification requirement clearly hampers the operation of a plant. For low enriched or highly irradiated material, a critical time of one month is proposed, requiring a frequency of physical inventory taking of one per month; this may also be too burdensome.

The critical time concept is thus not directly useful for defining a maximum time interval for closing material balances for international safeguards. Two possibilities can be identified to define a maximum time interval: the approach of partial physical inventory taking and a pragmatic approach, both of which are described below.

(i) A partial physical inventory of materials kept in intermediate storages in a process MBA can be carried out if the remaining fraction of the material in process is less than the measurement uncertainty (times a probability factor) of MUF, and if the amount of material in the intermediate storage can be measured so that an independent

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[1] Other approaches could be used. This paper does not examine the factors, both political and technical, that could be taken into account. The point to be stressed here is the importance of the level of accountability in the application of accountancy and the strong effect it can have on the inspection effort and the effectiveness of safeguards.

TABLE VII. COMPARISON BETWEEN PRESENT CAPABILITIES OF ACCOUNTANCY SYSTEMS AND THE LEVELS OF ACCOUNTABILITY FOR SPECIAL FISSILE MATERIAL[a]

	Light-water reactors[f]	U-conversion and fabrication	Reprocessing inventory[c] carried out by		Pu-storage	Mixed oxide fabrication
			wash-out	step function		
INPUT[b]						
Total amount (kg)	780	7650	1250/1100	420/370	362[d]	1088[e]
Number of batches	64	170	125	42	91	272
Random error (%)		0.1	0.5/0.5	0.5/0.5		0.2
Systematic error (%)		0.1	0.15/0.2	0.15/0.2		0.1
Standard deviation (kg)		7.6	1.95/2.2	0.71/0.96		1.1
OUTPUT - PRODUCT[b]						
Total amount (kg)	290/300	7647	1237/1088	413/362	(362)	1076
Number of batches	64	10	83/272	28/91	(91)	48
Random error (%)		0.1	0.1/0.2	0.1/0.2		0.5
Systematic error (%)		0.04	0.1/0.1	0.1/0.1		0.1
Standard deviation (kg)		3.9	1.2/1.1	0.42/0.36		1.3
OUTPUT WASTE[b]						
Total amount (kg)		3	12.5/11	4/3.7		10
Number of batches		310	100	20		100
Random error (%)		50	20	20		8
Systematic error (%)		10	5	5		2
Standard deviation (kg)		0.3	0.7/0.6	0.3/0.25		0.2
INVENTORY TAKING						
Amount	2300	700	5/5	25/25	varying	40
Number of batches	320	100	1	2/6		10
Random error (%)		1	5	10		0.5
Systematic error (%)		0.1	1	5		0.1
Standard deviation (kg)		1	0.3/0.3	2.1/1.6		0.1

TABLE VII (cont.)

MUF						
Resulting standard deviation, S_{MUF} (kg)	0	8.6	2.4/2.5	2.5/2.74		1.72
Confidence limit (95% in kg)	0	14.2	3.9/4.1	4.1/4.5		2.83
LEVEL OF ACCOUNTABILITY						
Assumed time interval	12	12	6	2	2	6
Assumed probability of detection	100	95	95	95	100	95
Amount of loss that would be detected with the assumed probability and S_{MUF} (kg)	1 assembly	28.4	7.9/8.3	8.3/9.1	1 bird cage	5.7

a ^{235}U and plutonium are considered.
b All figures are related to one interval between subsequent physical inventory takings of the same type.
c Data uranium/data plutonium.
d Item accountability.
e For plutonium only.
f Data taken for 1000 MW(e) PWR reactor.

estimate of that storage inventory can be achieved. Under these conditions the in-process inventory can be roughly estimated and the material balance can be closed with this approach more frequently than when based on a complete physical inventory. The practical cases, however, where these conditions are fulfilled seem to be rather limited. Partial inventories may still be useful as an aid to evaluation of the running book inventory.

(ii) The pragmatic approach is to verify a physical inventory taking whenever the plant operator takes a physical inventory for his own purposes but with a minimum frequency of at least once or twice per year. In a reprocessing plant, additional use can be made of the isotopic step function technique, which allows determination of the physical inventory by formation of so-called 'super batches' which have significantly different (in a statistical sense) isotopic compositions. This technique reduces the burden on the operator compared with normal physical inventory taking but requires a slightly greater number of samples to be analysed by the Agency. The practical frequency for this method of inventory taking is estimated to be about six times per year.

The level of accountability thus describes a defined amount of special fissile material, the loss or diversion of which is to be detected at a given level of confidence with a given probability of detection and within a given period of time. Following this definition of the level of accountability it is not possible to combine several MBAs and determine the level of accountability for that combination. Thus the level of accountability applies to single MBAs only. Proposals have been made to combine a number of facilities to a super MBA, e.g. a whole fuel cycle or all facilities in a State, and to detect diversion on the basis of a 'global' MUF or 'country' MUF. This is not practical, since the physical inventory of that super MBA cannot normally be obtained at the same point of time and hence MUF and S_{MUF} cannot be determined.

Table VII shows the assumed limits of accuracy for each KMP and for MUF, the assumed time interval, the assumed probability of detection and the resulting amount of loss that would be detected under these assumptions. The measurement uncertainties stated are based on the literature and the Agency's experience and are representative of performance expected in industry today. The magnitude of the systematic errors is very critical with respect to the amount of loss that can be detected and these errors will have to be investigated further in the course of integral tests and inter-laboratory comparison tests to determine whether they can be made in practice. There are indications that many of the accuracies stated in the literature tend to understate the accuracies that are actually achieved by industry. This appears to be largely due to a lack of data on and understanding of the systematic errors associated with sampling, calibration of methods and instruments and reference standards and whether reference standards are representative of process materials measured at KMPs.

For reactors and sealed storages the loss of one item should, of course, be detectable with a probability of 100%. For uranium conversion and fabrication, a loss of 28.4 kg ^{235}U could be detected with a probability of 95%. For reprocessing, the corresponding amount of plutonium detectable with 95% probability is 8.3 kg or 9.1 kg, depending on the inventory

taking technique and the assumed time intervals of six and two months, respectively. For the mixed oxide fabrication plant, a loss of 5.7 kg plutonium would be detected with a probability of 95%. These results, which would seem to be of the right order of magnitude, are extremely sensitive to the magnitude of the systematic errors and it is clear that, if the stated errors are not made in practice, the level of accountability will increase significantly. In such cases the Agency would have to increase the inspection effort to be able to explain the causes of the systematic errors and to make the relevant statements concerning the adequacy of the measurement system.

5. DEVELOPMENT OF SAFEGUARDS TECHNIQUES

The major development problems yet to be solved are associated with the verification of accounting data which have been reported. Inspections should be as unintrusive as possible. It would thus be of general interest to reduce the inspection effort needed to verify the limits of accuracy achieved by the operator and to ensure that the limits of accuracy are consistent with the level of accountability accepted as adequate for international safeguards. This may be obtainable by developing measurement methods of higher accuracy or which require less effort for the same accuracy and by developing other advanced techniques that reduce the uncertainty in evaluations of MUF.

The following areas should merit special attention at the present time:

(a) The development of sealing and identification methods in order to establish item accountability in transit, storage areas and reactors. The requirements are for simple verification of item integrity and unique identification, which are in principle – though not always in practice – easy to achieve. There is still no easy way to meet these requirements for reactor fuel assemblies, although several promising lines of development work are being pursued.

(b) The development of automatic and tamper-resistant, non-destructive measurement techniques. In many cases, non-destructive techniques are the only ones available, and, in other cases, they have obvious advantages; for instance, in permitting concentration of flow measurements to a single process stage. Such techniques may reduce intrusiveness but may not reduce the inspection effort, and this is pointed out in the preceding text. In some cases, automated measurements may achieve some reduction in inspection effort, e.g. automatic scales with sealed print-outs. Some instruments may actually increase costs for the same accuracy and this will have to be balanced with the reduction of intrusiveness.

(c) One important recent development has been the experimental proofs of the value of isotopic correlation techniques. At present there are still too few data available, but the technique gives hope that it will be possible to simplify the verification task, particularly at the head-end of reprocessing plants.

The basic safeguards objective, that of achieving a quantitative statement of the results of the verification work, requires that both the safeguards procedures and any new techniques be proven in an initial application under completely realistic conditions. This would mean that, for each new type of facility or each new facility which has significant differences

from others of the same type, a greater verification effort would have to be spent in the initial safeguards stage in order to obtain the required assurances. Only after that will it be possible to define the LRIE for that plant. Part of this work can be performed in integral tests in which all elements of a safeguards system are applied. The experience gained from integral tests has been very important, not only in proving specific techniques in a practical case and providing clear statements of the performance of the verification procedures, but also by providing important information on the feasibility of inter-laboratory tests designed to determine the comparability of analytical results obtained at different laboratories. These tests indicate that great problems exist in achieving comparability between laboratories and in obtaining rapid transport and analysis of samples.

REFERENCES

[1] INTERNATIONAL ATOMIC ENERGY AGENCY, Articles 28, 29 and 30 of INFCIRC/153.
[2] INTERNATIONAL ATOMIC ENERGY AGENCY, INFCIRC/153.
[3] ROMETSCH, R., "Development of IAEA safeguards system for NPT", these Proceedings, Paper 770, Vol. 9.
[4] INTERNATIONAL ATOMIC ENERGY AGENCY, Safeguards Techniques (Proc. Symp. Karlsruhe, 1970), IAEA, Vienna (1970) 2 vols.
[5] INTERNATIONAL ATOMIC ENERGY AGENCY, Article 29, INFCIRC/153.
[6] UNITED NATIONS, "Effects of the possible use of nuclear weapons and the security and economic implications for states of the acquisition and further development of these weapons", Report of the Secretary General transmitting the study of his consultative group, UN Doc. A/6858 (1968).

EXPERIENCE WITH A NUCLEAR MATERIALS INFORMATION SYSTEM

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Abstract–Résumé–Аннотация–Resumen

EXPERIENCE WITH A NUCLEAR MATERIALS INFORMATION SYSTEM.

This paper describes the data-gathering functions and applications of the USAEC Nuclear Materials Information System (NMIS). The system encompasses the activities of approximately 1200 entities. The central system is highly automated but is sufficiently flexible in its accumulation of information and production of usable reports to accommodate and interface with USAEC contractor data, commercial lincensee data, and international data. The sources and types of information input into the system and the data elements concerning them are discussed. Distinction is made between elements essential for safeguards alone, those for nuclear-materials management alone, and those common to both functions. Areas where the system is designed to provide safeguards control include the timely recognition and analysis of shipper-receiver differences, loss data, and inventory variations. Safeguarding materials being transported relies completely on physical protection techniques; however, responding to incidents requires rapid communication about the shipment. This requirement will be dependent upon capabilities built into the NMIS system. Identifying the amount of nuclear materials in a transaction may be one of the strongest tools of an information system. The paper discusses the methods used by the USA and describes some evaluations being programmed into the NMIS. In the same vein, the identification of material unaccounted for (MUF) and normal operating loss (NOL) is essential. These will be reviewed, and some statistical evaluations will be presented. The paper discusses the evolutionary process that is envisioned and planned to bring reporting and subsequent analysis of safeguards data into the system on a more timely basis, together with related attention indicators and action limits. It concludes with a summary description of the many different types of reports presently generated by the NMIS and of how they serve many different aspects of materials management and safeguards. And, with a look to the future, the paper touches on the many new programs scheduled for NMIS.

EXPERIENCE ACQUISE AVEC UN SYSTEME DE TRAITEMENT DE L' INFORMATION CONCERNANT LES MATIERES NUCLEAIRES.

Les auteurs décrivent les fonctions et les applications du NMIS (système de traitement des informations sur les matières nucléaires) de l' USAEC, qui porte sur les activités d' environ 1200 établissements. Le système central est extrêmement automatisé tout en restant suffisamment souple quant à la manière dont il stocke les informations et produit des rapports pour recevoir les données des adjudications de l' USAEC, les données des établissements privés et les données internationales. Ils énumèrent les sources et les types de données d'entrée et décrivent les éléments qu'elles comportent. Parmi les éléments, on distingue ceux qui sont indispensables pour les garanties seulement, ceux qui sont destinés uniquement à la gestion des matières nucléaires et ceux qui sont communs à ces deux catégories. Les opérations de ce système qui intéressent le contrôle aux fins des garanties consistent notamment à connaître suffisamment à temps et à analyser les différences entre les chiffres de l' expéditeur et ceux du destinataire, les pertes et les variations du stock. L' application de garanties aux matières transportées est entièrement tributaire des méthodes de protection physique; toutefois, en cas d' accident, une communication rapide est nécessaire. Cette exigence dépendra des possibilités incorporées dans le système NMIS. L' identification des quantités de matières nucléaires faisant l' objet d' une transaction peut être un instrument puissant d' un système d' information. Les auteurs étudient les méthodes utilisées aux Etats-Unis et décrivent certains calculs programmés pour NMIS. De même, il importe d' identifier les différent cas d' inventaire et les pertes en cours d' exploitation normale. Les auteurs passent ces méthodes en revue et présentent certaines évaluations statistiques. Ils décrivent ensuite le processus d'évolution qui est envisagé

pour faire entrer plus rapidement dans le système les rapports concernant les garanties et leur analyse, ainsi que les signaleurs correspondants et les limites d'action. Ils donnent enfin une description succincte de différents types de rapports actuellement produits par NMIS et montrent comment ils servent à différentes fins de gestion et de garantie des matières. En ce qui concerne l'avenir, les auteurs disent quelques mots des nombreux nouveaux programmes préparés pour NMIS.

ОПЫТ РАБОТЫ С ИНФОРМАЦИОННОЙ СИСТЕМОЙ ПО ЯДЕРНЫМ МАТЕРИАЛАМ.

В докладе дается описание назначения и применений Информационной системы по ядерным материалам Комиссии по атомной энергии США. Эта система охватывает деятельность примерно 1200 объектов. Централизованная исключительно автоматизированная система обладает достаточной гибкостью в накоплении информации и получении отчетов, пригодных для согласования и разграничения с данными поставщиков Комиссии по атомной энергии США, данными промышленных организаций, которым были выданы лицензии, и международными данными. Рассматриваются источники и виды информации, вводимые в систему, и связанные с ними элементы данных. Проводится разграничение между элементами, которые имеют важное значение только для системы гарантий, и элементами, относящимися лишь к учету и снабжению ядерными материалами, а также элементами, несущими на себе функции как первых, так и вторых. Области, для которых система должна обеспечивать контроль по гарантиям, включают в себя своевременное установление и анализ расхождений между данными грузоотправителя и грузополучателя, данные о потерях и изменениях материально-производственных запасов. Охрана перевозимых материалов полностью основана на физических методах защиты; однако в случае инцидентов требуется быстро сообщить данные о грузе. Это требование будет зависеть от возможностей, заложенных в Информационную систему по ядерным материалам. Установление количества ядерных материалов, связанных со сделкой, может быть одной из самых сильных сторон информационной системы. В докладе рассматриваются используемые в США методы и описываются некоторые оценки, запрограммированные в эту систему. Таким же образом важное значение имеет установление неучтенного материала и нормальных потерь в процессе производства. Для последних дается обзор и представлены некоторые статистические оценки. В докладе рассматривается эволюционный процесс, предусматриваемый и запланированный для того, чтобы вводить в систему отчетные данные и данные последующего анализа по гарантиям на более своевременной основе, вместе с соответствующими сигнальными индикаторами и пределами действий. В заключение дается краткое описание многих различных видов отчетов, которые в настоящее время выдаются Информационной системой по ядерным материалам, и каким образом они отвечают многим различным аспектам гарантий, учета и снабжения материалами. Что касается перспектив на будущее, то доклад затрагивает также много новых программ, запланированных для Информационной системы по ядерным материалам.

EXPERIENCIA ADQUIRIDA CON UN SISTEMA DE INFORMACION SOBRE MATERIALES NUCLEARES.

En esta memoria se describen las funciones y aplicaciones del sistema de acopio de información sobre materiales nucleares (Nuclear Materials Information System, NMIS) de la Comisión de Energía Atómica de los Estados Unidos (USAEC). El sistema abarca las actividades de aproximadamente 1200 entidades. El sistema central está altamente automatizado, pero es suficientemente flexible en su acumulación de información y producción de informes para adaptarse e integrar en sí datos procedentes de los contratistas de la USAEC, de los usufructuarios de licencias comerciales, así como datos internacionales. Se estudian las fuentes y tipos de información recogida en el sistema y los elementos de los datos a ellos concernientes. Se distinguen los elementos esenciales exclusivamente para las salvaguardias, sólo para la gestión de materiales nucleares y los que son comunes a ambas funciones. Entre los sectores en los que el sistema sirve para el control de salvaguardias, se incluyen el reconocimiento oportuno y el análisis de las diferencias remitente-destinatario, los datos de pérdidas y los cambios en el inventario. La salvaguardia de los materiales en tránsito se basan enteramente en técnicas de protección física; sin embargo, para hacer frente a los incidentes se requiere una comunicación rápida acerca del envío. La satisfacción de esta necesidad dependerá de la capacidad que posea el sistema NMIS. La identificación de la cantidad de materiales nucleares que intervienen en una transacción puede ser uno de los elementos más importantes de un sistema de información. El trabajo describe los métodos empleados en los Estados Unidos y presenta algunas valoraciones que se han introducido en el programa NMIS. En el mismo sentido, es esencial la identificación de las diferencias inexplicadas y de las pérdidas normales de explotación. Estos elementos son objeto de un estudio general, presentándose además algunas evaluacionnes estadísticas. La memoria estudia el proceso evolutivo tal como se concibe y proyecta para conseguir que la recopilación y ulterior análisis de los datos sobre salvaguardias se ajusten a límites temporales más adecuados, incluyendo los apropiados indicadores y sus correspondientes límites de acción. Se concluye con un resumen descriptivo de los numerosos y diferentes tipos de informes suministrados actualmente por el NMIS, y de la manera en que son útiles para muy diferentes aspectos de la gestión de materiales y de las salvaguardias. Por lo que se refiere al futuro, el trabajo menciona también numerosos programas previstos para el NMIS.

GENERAL

The United States Atomic Energy Commission's development of its Nuclear Materials Information System (NMIS) is now well into its third decade. And while this paper will describe the system as it has evolved to date, it can readily be seen that the future will introduce many changes.

As a point of reference, it is the NMIS, as utilized by the United States of America, or any similar system used by another responsible state, that gives substance, interpretation, and evaluation to a large part of their safeguards and materials management efforts. The better inventory techniques, the more accurate measurement methods, the more valid statistical sampling plans – all these serve to give more credibility to the quantitative values that flow into and through the NMIS. But first, there must be an information system that is receptive to the data and capable of presenting the appropriate safeguards and materials management reports to those whose function it is to control these activities.

One of the principal guidelines that our experience has given us is the importance of recognizing and utilizing the dual capabilities of the NMIS. The system is designed to serve the needs of both safeguards and materials management efforts. The huge masses of data fed into the system and the use of computers to absorb the data and produce the reports dictate that full use be made of each piece of data; based on capturing it once and only once.

To establish a reporting unit in the NMIS we have assigned an alphabetic Reporting Identification Symbol (RIS) to each entity within the AEC's sphere of concern. At present there are approximately 1200 such facilities. These RIS's are an important control feature and are reflected in some manner in all NMIS programs. The symbols are organized so that those facilities subject to both materials management and safeguards concern can be readily distinguished from those which are only of safeguards concern.

The distinction made for RIS's begins to gain importance in considering the materials covered by the NMIS. For, in addition to special nuclear materials and source materials (which are of dual concern to the AEC) we control several other materials, such as neptunium, deuterium, and tritium for management purposes only.

The degree of concern which the NMIS brings to bear is based on several things. Although the United States in general deals in pounds and ounces, the NMIS is based on material quantities expressed in kilograms or grams. We believe that we are most fortunate in this regard. Having related the reporting unit to the materials being reported, the frequency and extent of reporting, inspecting, and monitoring information for each facility can generally be said to be governed by the functions of the facility in question and whichever of the dual concerns of the AEC it is subject to.

One of the major evolutionary processes in the development of the Commission's Nuclear Materials Information System has been the transition from a manual information system to a computer-based system. The source data originating in the individual facilities flows to the AEC Field Offices where the data is input to a central processing system. The central system performs the data input processing operations, maintains the NMIS data base, and produces the safeguards and materials management reports. These operations are accomplished on the modern, high-capacity computer facilities of the Computing Technology Center (CTC), operated by Union Carbide Corporation and located at Oak Ridge, Tennessee.

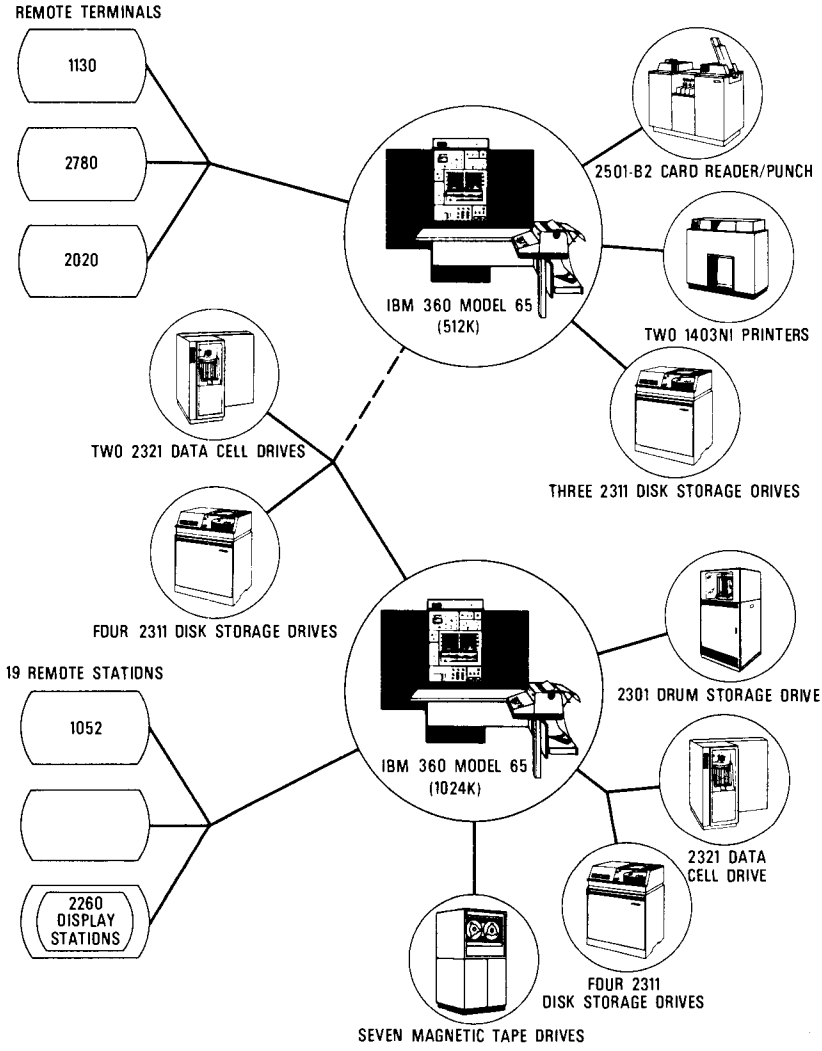

FIG. 1. Primary CTC computer system.

A graphic representation of the primary computer system serving the NMIS is shown in Fig. 1. The system consists of two computers intercoupled in such manner that one processor is the scheduling processor for the total configuration. The system is equipped with a variety of mass data storage devices that provide the capacity for maintenance and active use of the high-volume data base.

One of the characteristics of a centralized data base is the physical displacement from users that are geographically dispersed. The use of telecommunications technology is progressively improving the timeliness of information flows to and from the NMIS data base. A high-speed,

secure data transmission link provides computer-to-computer communications between the Computing Technology Center (CTC) and AEC Headquarters, which are separated by some 800 kilometers. Reports serving the Headquarters organizations are transmitted via the link, and the link is used by Headquarters to input data to the system.

A computer-based data transmission and message switching system is being utilized to transmit source data from some of the AEC Field Offices to the Computing Technology Center. This system utilizes a network of data terminals connected to a message switching center. The terminals are just now becoming operational in the other offices, thus the contribution this system will make to the input operations has not been fully realized. The existence of both the computer link and the data transmission network provides a significant back-up capability to some of the most important lines of information flow.

Many contractors and licensees have in-house information processing systems that are utilized for their facility materials management and control operations. Much of the information required as the facility input to the AEC's safeguards and materials management system is introduced into these local automated systems and we are continuously exploring the potential for improving the data cycles through more direct methods of input to the central system.

INPUT TO THE NUCLEAR MATERIALS INFORMATION SYSTEM

Those of us who work closely with the system are continuously amazed by the relatively few source documents used to provide input to the NMIS. But we are also continuously reminded of how dependent we are on getting good usable data as quickly as possible and the importance of carefully designed edit programs to screen the data when it enters the computerized portion of the system. To assure maximum credibility to the NMIS data the edit routines expose the prospective input to tens of compatibility checks. Some checks cause the data to be rejected while others only send along a warning message regarding it. Resolving the edit rejections is frequently very complex and requires control personnel in the NMIS with a broad understanding of both the computer system and the operations of the facilities.

Very few of our facilities have serious problems in meeting our current deadlines for sending in their data. It is when the data is rejected by an edit program and must be corrected by organizations separated from our headquarters and each other by hundreds or even thousands of kilometers that we lose vital time. For, if a shipper codes his shipment as 1000 units of normal uranium and the receiver codes it as 1000 units of plutonium we must determine which is right and then correct the data that was in error. Situations such as this necessitate our control personnel making dozens of contacts each day. They then must place the correct information into the data base before the report period can be closed and the output reports prepared.

The most frequent type of data and possibly the backbone of the NMIS is that covering a transaction. In this category we have our Nuclear Material Transfer Report, Form AEC-741. In addition, we rely on an SS Material Transfer Receipt, Form AEC-284; loss reports covering

normal operational losses, burn-up of uranium, decay, accidental losses, and material unaccounted for; journal entries; and reports from foreign entities.

Transaction data will reflect the identification symbol of the shipper and receiver; the transaction number; the dates of shipment and receipt; the material type and form; container and batch numbers; and for both the shipper and receiver: their independently arrived at element and isotope weights, per cent enrichment, and related limits of error. Management needs also call for the nature of the transaction; ownership data; transfer authority; financial transfer date; project numbers; and for whose financial account the transfer is made from and to.

A second type of NMIS input source document is the Material Status Report, Form AEC-742. While the 742 form still temporarily serves this source document function, it is a very good example of the evolutionary aspects of the system. The report is currently prepared by each RIS semiannually and has been in use for over twenty years. However, since we are making every effort to be more responsive to changes and more timely in our reporting, six months is clearly an unacceptable frequency of data input.

In analysing the type of data being provided it was determined that by going to a monthly or 'event' level of reporting frequency for less than half a dozen data elements the full AEC-742 report would no longer have to be submitted to the AEC for input to the NMIS. However, the 'balancing' concept built into the report was recognized to be such a useful safeguards and materials management tool that the NMIS now generates the same report data and presents the information as an output report.

Composition of Ending Inventory (COEI) data represents an extremely important segment of input to the NMIS. The RIS's submit this information to us in varying degrees of frequency and detail. At present our management concerns, as distinct from safeguards, call for either monthly or quarterly submissions in substantial detail. And while safeguards needs are presently keyed to less detail and predominantly semiannual submission, we envision both of these criteria becoming more informative and timely in the future.

Our COEI data follows both a usage and a physical form style of presentation to the NMIS. Inventories are reported as being in non-production research and development; as feed materials; as in process; as intermediate products; as inactive material; or as scrap material. Each of these major categories are detailed to reflect the related physical forms that the materials can take.

The project requirements forecast and inventory data entering the NMIS is essentially of management concern, but does have overtones of safeguards interest. Those facilities engaged in non-production research and development activities must provide this information for any special nuclear materials that are involved. Each RIS submits a forecast of its material needs for several years in advance. This input reflects inventory levels, new material needs, and returns of material by each material type. This information is further broken down by each of several hundred research and development programs currently active within the AEC.

As with any forecast or materials budget, they are subject to change as program interests shift, new programs are identified, or quantitative material values become better known. All of these events are fed into the NMIS at periodic intervals.

Once nuclear material is assigned to a research project any subsequent changes to its status must be reported to the system at least monthly. So, just as each RIS must report events related to it as a whole, it must report similar data for each of the dozens of projects it may be engaged in.

With the system's heavy reliance on computers it follows that standard codes are used whenever practicable. Through the use of such codes it is possible for our larger facilities to program their own systems to provide reports in forms which interface with the NMIS to a high degree. There are three control codes which are particularly significant in this regard.

The first is the unique three-letter Reporting Identification Symbol code given each entity in the NMIS. It assures positive facility identification for any input. A standard two character numerical code is used to provide material type identification. The third major control code is the research and development project number. While the project numbers are used only for management programs, the other two codes serve a dual purpose.

When we relate the interaction of 1200 RIS's, over 500 project numbers, and 12 major material types and then recall the many different data elements captured by the NMIS input as it concerns itself with transaction reports, material status reports, inventory reports and forecast and project reports one can readily see that the NMIS has a tremendous data base.

REPORTS PREPARED BY THE NMIS

From the data sources described and the data base that has evolved, the NMIS is routinely issuing about seventy different reports of safeguards and materials management concern. These reports are on frequency schedules varying from weekly to semiannually. Distribution itself encompasses over forty major organizational units of the AEC in addition to the Office of Safeguards and Materials Management which administers the system. We have long been providing the IAEA with transaction and inventory data for selected facilities and are looking forward to broadening this interface in the future.

When we consider the many RIS's and the diversification of the nuclear industry in the United States it will come as no surprise that our 'Transaction' family of reports represents a substantial portion of the NMIS reporting structure. These reports are issued monthly and reflect all transfers of material, whether the transfer involves physical movement, or in-place programmatic or ownership change. Both the shipper's and receiver's weights are shown, as well as their respective limits of error, and if there is one, the shipper-receiver difference. Thousands of transfers are reported to the appropriate AEC field offices responsible for administering the safeguards and management functions.

Other types of transaction reports cover the production of material, procurement of material, and loss mechanisms such as normal operating losses, approved inventory write-offs, and material unaccounted for. In addition, reports of transactions which have not been completed and of apparently missing transactions reports are distributed to operating organizations throughout the Commission so that they may take the necessary corrective action.

Composition of Ending Inventory statements from the individual RIS's are translated into dual use reports on a monthly and quarterly basis. Each of the twelve material types is the subject of separate handling. Summary reports may show only total amounts; others will literally show hundreds of lines of detail to describe the different physical forms, program usages, and isotopic enrichments. Our enriched uranium reports reflect nineteen ranges of ^{235}U enrichment and our plutonium reports show seven ranges of isotopic content.

A COEI is presented for individual RIS's; for clusters of RIS's; for research and development holdings; for assay ranges; and for scrap inventory holdings. Other reports serve to consolidate holdings by field offices; and finally we have those that consolidate on an AEC-wide basis.

The many variations of our inventory reports are given wide distribution. Some of the receivers use them for management planning and scheduling. Others are primarily concerned with the safeguards story they tell. For example, the quantity of scrap may represent a recovery problem for materials managers and indicate production inefficiencies. The same scrap inventory may be viewed with concern by safeguards staff because of the difficulties of obtaining accurate measurements of the scrap and the awareness of its potential for masking diversions elsewhere in the process.

Material Balance Reports serve a very useful purpose in assuring the inclusion of all transactions and events in evaluating the adequacy of controls. The balancing equation has the beginning inventory plus new production and receipts being equal to the ending inventory plus shipments and loss mechanisms. When this balance is reached, those responsible for materials management and safeguards know that all of the values which must be verified and evaluated have been included in the report.

These reports are normally prepared semiannually. A separate report is prepared for each of the twelve material types held by each of the 1200 RIS's, which in turn are consolidated into twelve field office reports representing the sphere of concern of each office. And finally, the twelve reports are consolidated into a single report for the entire United States. It is not unrealistic to say that the effort to reduce 1200 individual plutonium reports to twelve reports and then to one report is uniquely suited to an automated system. This same operation is repeated for each of the other eleven material types. This same type of consolidating effort is applied to arrive at our composition of ending inventory reports. But while the task would be truly huge if undertaken by hand, the NMIS computers do it all in less than six minutes.

Forecast reports are instrumental in the management and control of inventory levels in the specific projects and programs within the AEC's sphere of concern. The detailed forecast information provides a projection of specific inventory quantities and the estimated schedule of material withdrawals and returns. Such information is a basic tool for both short-term and long-term budget and production planning. In the forecast reports, information is detailed and summarized in a variety of ways to present the specific organizations with the information needed for administration of the programs. Variations in reports include the clustering of data by program division, usage category and reporting location. Uranium quantities are reported by isotopic enrichment.

The forecast information permits the schedule of material withdrawals to be balanced against the schedule of returns across all programs.

Production schedules can then be set to produce the quantities of material that cannot be supplied from the quantities being returned from the projects. The forecast is prepared annually and then adjusted at the midpoint of the year to reflect revisions from previous forecasts and add a new forecast for an additional six-month period.

Allotment transaction reports are prepared each month and distributed to the program divisions, the field offices having programmatic responsibility and the field offices having jurisdiction over the facilities physically involved in the movement or processing of the materials. The transaction reports reflect activities that are directly related to the forecast of material requirements and usage. Material withdrawals, returns, losses, and transfers between projects are identified.

Project inventory reports are produced quarterly, based on inventory data reported by the facilities. These reports, like the forecast reports, are created in the various forms and summarized levels needed to serve the many organizations involved in the administration of the programs and the control of the nuclear materials used by these programs.

Project inventory reports also are available from the system records on a 'book' inventory basis at the close of each monthly cycle of the transaction data system. Currently, the reports are generated and issued quarterly. These reports are examples of the information available from interactive processing utilization of the different components of the centralized data base. Project inventory data is updated by transaction information to arrive at the current book value of the project inventories.

In addition to the 'families' of reports which have been described, the NMIS prepares a number of others that are essential contributions to satisfying our dual concerns. One that is of extreme importance is our loss mechanism report that is prepared semiannually. This report exposes our losses and material unaccounted for values to several series of statistical evaluations involving standard deviations, linear regressions, multiple regressions and other sensitivity measurements. Besides being in tabular form, the more significant evaluations can be displayed graphically for quick review.

The NMIS maintains two Reporting Identification Symbol Directories; one covering Commission contractors, and the other covering commercial licensee facilities. In addition to identifying each facility and its related RIS, the directories provide mailing addresses, shipping addresses, and other items of administrative importance.

In a primarily materials management vein, the system maintains an index of all of the Commission's Research and Development project numbers and titles. As with the RIS, these project numbers can be used to control the reporting format of our transaction reports, inventory reports, and many others. The nuclear materials held in a project inventory can be readily related by the project number to the appropriate project manager. This feature makes it possible for safeguards staff to monitor the inventories and quickly contact a project manager if imbalances are noted.

Normally, the development of working papers by hand consumes a large part of an inspector's time, both at home and when in the field. The NMIS utilizes a program to prepare a basic set of some of the necessary working papers for any RIS or combination of RIS's. The schedules show all the transactions for the inspection period. The receipts and shipments are grouped by transfer parties and each series reflects monthly totals.

In addition to showing both shipper's and receiver's weights, respective limits of error are given as well as any shipper-receiver differences in weights. By comparing this information to the facility's records the inspector can identify differences and make the necessary reconciliations.

As mentioned earlier, the system has the capability to generate a 'book' material balance report for any facility at any time. This is very helpful in monitoring individual facilities inasmuch as the report can be scanned month by month to note any significant operating changes or inventory level changes that might warrant scheduling an inspection at once rather than waiting for some routine date in the future.

Since our AEC-wide material balance reports and ending inventory reports are due fifteen days after the end of a report period, this ability to compute the 'book' material balance and 'book' ending inventory for a facility is essential. With such a large number of facilities required to report, some are always late. These are identified by the computer which then proceeds to prepare the corresponding 'book' reports and uses those values in developing the consolidated reports rather than waiting until the facility sends its values in.

Each shipper and receiver of nuclear material is expected to make independent measurements of the amount of material involved and show the measurement limits of error on the copies of the transfer document sent to the NMIS. This data then serves as the basis for statistically analysing any shipper-receiver difference that results from the transaction. This capability is one of safeguards strongest tools for control, yet frequently it is objected to by materials managers who for financial reasons wish to compromise on a single set of values for payment purposes. It should be clear that in such instances the safeguards concerns should prevail.

NEW PROGRAMS

We in the Office of Safeguards and Materials Management who are concerned with the administration of the Nuclear Materials Information System have some very definite ideas regarding new programs for the system. We also have more general ideas for some other programs, and then, still further out in our planning, lie a number of conceptual ideas. At present, we are working on, or scheduled to work on, twenty-four development modules for the system. Some of these are ready to be placed on an operational basis now, others have not yet reached the program development stage.

Frequently we find the future plans being dictated by the requirements of the many organizational units of the AEC that look to the NMIS for staff support. In these instances we must re-examine our development priorities to bring the new modules in at the proper points in time.

A never-ending effort involves the conflicting goals of improving the accuracy of the data and increasing the timeliness of the data. There is no question but that substantial progress can yet be made in both areas within the existing technology. And while we work with the tools available today we will be continuously alert for future tools and techniques that will allow us to approach real time in our information system. A very high

percentage of the input is on a weekly cycle now and with the improved communications that are being developed there is no reason why we cannot obtain daily input within the next three years.

Modifying and supplementing existing programs is going on constantly. Occasionally it involves input data and in those instances it is usually necessary to modify facility reporting requirements and input forms, with all the attendant delays of redesigning forms and instructing the facilities in their proper use and then reprogramming the many interlocking programs to accept and reflect the new data elements. Most frequently, however, it is our report presentation that is changed. A change may be a minor rearrangement of the information on an existing report or it may call for the development of completely new reports within the program module. Some specific modules that are scheduled for substantial buildup are the inspection working paper programs and the project control programs. By setting threshold levels in many of our existing programs we will be able to provide the inspectors with an array of operational and historical information for each facility. Project control will be expanded beyond the research and development activities to include all production activities and all twelve nuclear materials.

One of the program development modules which has been brought from a relatively low priority level to the first priority level is that designed to generate financial reports based on quantitative data. As a result of close examination and some adjustments, the types of financial data needed by AEC management have been matched with a number of existing NMIS programs. By making a comparatively few changes to the NMIS and adding a data bank containing nuclear material unit cost information it appears that related financial data and reports can be obtained within the NMIS at practically no additional cost. This application of the single data input concept will benefit the AEC by avoiding the need of establishing a duplicate information system based on financial costs alone.

While many of the details remain to be identified and the best methods searched out, we recognize that the pending application of the IAEA's Non-Proliferation Treaty safeguards in the United States will necessitate the development of very careful interface programs. Every effort will be made to assure the flow of data from our domestic facilities to the IAEA in consonance with their needs. At the same time, the integrity of the data flowing into the NMIS must remain intact. Since the IAEA's control level is the material balance area and the NMIS is based on the facility level we will be expanding our 1200 reporting entities, with a corresponding increase in data flow to the system. With the flexibility and capacity of our computer system, we do not envision any real problems in processing the data and preparing reports. Rather, we expect that most of the problems will arise in trying to bring the material balance area data to the quality levels required by our edit programs without causing unacceptable delays in closing the data base for the period.

Both the computer industry and communications industry are devoting tremendous efforts toward research and development activities to improve their utility to everyone. We intend to take every advantage of their progress to further the responsiveness of our information system. The AEC is sponsoring considerable research and development in the betterment of nuclear materials measurement tools and techniques. As these many development programs become suited for operational application, we

expect to come closer to a national real time information system that will tie in our individual facilities to the NMIS by direct communication links.

When the capability is available to us we can look toward daily inventory positions being reported and analysed by the NMIS and passed along to nuclear materials managers and safeguards staffs both in our headquarters and in the field. And when it is necessary to transfer nuclear materials beyond the relative safety of processing plants we will monitor the movements through the use of relay points tied into a control program that will follow each shipment to its destination.

We feel confident that our experiences in developing and operating a nuclear materials information system are not unlike those of many others, but to the extent that we can share our experiences in greater detail on a case-by-case basis, or as they may apply to specific problems, we would welcome the opportunity.

A REVIEW OF RESEARCH AND DEVELOPMENT IN JAPAN ON SAFEGUARDS FOR NUCLEAR MATERIALS IN FABRICATION PLANTS

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Abstract–Résumé–Аннотация–Resumen

A REVIEW OF RESEARCH AND DEVELOPMENT IN JAPAN ON SAFEGUARDS FOR NUCLEAR MATERIALS IN FABRICATION PLANTS.

The Japanese government has in the last three years made great efforts to establish a reliable nuclear material accounting system for safeguarding nuclear materials in fabrication plants. Plant operators have been working on the following items.

For highly enriched plate-type fuel, a quick and reliable control system using a minicomputer has been developed. The system is designed so that real time data handling, large volume data storage and easy output of many different data can be achieved. The system is expected to increase the speed and accuracy of accounting control, and to improve process control, manufacturing techniques and safeguards inspection.

A material control system for low-enriched UO_2 fuel has also been developed. In batch-type operation, a system has been designed to calculate plant inventory at any given time with a certain accuracy and confidence level by giving loss and scrap rates in each process operation. The system consists of SMODNUP which gives characteristics of plant and SBIDNUP which calculates plant inventory. In flow-type operation, a system has been designed to calculate plant inventory by giving loss rate in each material balance area and monitoring the throughput there. The system includes SBIDNUP-II which gives a warning and suggests operations for maintaining a given confidence level.

To test the reliability of the accounting control system, it is very desirable for weighing data to be obtained automatically with sealed measuring equipment but such a procedure should not greatly complicate the fabrication process. To meet these requirements, prototype automatic weighing and recording equipment has been developed and tested for UO_2 pellets during the stacking, the last step in the fabrication process before introduction into the cladding.

ETUDES ET REALISATIONS AU JAPON CONCERNANT LE CONTROLE DES MATIERES NUCLEAIRES DANS LES USINES DE FABRICATION.

Le Gouvernement japonais déploie depuis trois ans beaucoup d' efforts pour mettre au point un système de comptabilité matières sûr pour assurer la garantie des matières nucléaires dans les usines de fabrication. Les exploitants de ces usines travaillent principalement sur les deux questions suivantes.

Pour le combustible en plaques fortement enrichi, ils ont mis au point une méthode rapide et sûre comportant l' emploi d' un petit ordinateur. Elle est conçue de telle sorte que l' on peut procéder au traitement des données en temps réel, mettre en mémoire un grand nombre de variables et calculer facilement de nombreux paramètres. On pense qu' elle permettra d' augmenter la rapidité et l' exactitude de la comptabilité matières et d' améliorer le contrôle, les méthodes de fabrication et les modalités de l' inspection aux fins de garanties.

Ils ont mis au point également un système de contrôle des matières pour le combustible à l' UO_2 faiblement enrichi. Pour cela, ils ont élaboré une méthode permettant de calculer à tout moment le stock d' une usine fonctionnant en discontinu, avec une certaine précision et un certain degré de confiance, indiquant les taux de perte et de résidu de chaque opération. Elle comprend deux parties: le «SMODNUP» qui donne les caractéristiques d' une installation et le «SBIDNUP» qui calcule son stock. Pour l' exploitation

en continu, ils ont mis au point une méthode permettant de calculer le stock de l'usine à partir du taux de perte pour chaque unité de bilan matière, par contrôle du débit de l'unité. Cette méthode comprend le «SBIDNUP II» qui joue un rôle d'avertisseur et suggère les mesures à prendre pour maintenir le degré de confiance.

Pour vérifier la fiabilité de ce système de contrôle comptable, il est extrêmement souhaitable que les pesées soient faites automatiquement aux points stratégiques au moyen d'appareils de mesure scellés; mais cette méthode ne doit pas compliquer exagérément le processus de fabrication. A cette fin, on a mis au point et essayé un prototype de balance enregistreuse automatique pour la pesée des pastilles d'UO_2 avant gainage.

ОБЗОР ПО ИССЛЕДОВАНИЯМ И РАЗРАБОТКАМ, ВЫПОЛНЕННЫМ В ЯПОНИИ ПО КОНТРОЛЮ ЗА ЯДЕРНЫМИ МАТЕРИАЛАМИ НА ЗАВОДЕ ПО ИЗГОТОВЛЕНИЮ ТЕПЛОВЫДЕЛЯЮЩИХ ЭЛЕМЕНТОВ.

В течение последних трех лет японское правительство затрачивает много усилий на создание надежной системы учета ядерных материалов для контроля за ядерными материалами на заводе по изготовлению тепловыделяющих элементов. На этом заводе были разработаны следующие системы.

Для пластинчатых тепловыделяющих элементов на основе высокообогащенного топлива разработана быстродействующая и надежная система тщательного контроля за ядерным материалом с использованием миниатюрной электронно-вычислительной машины. За реальное время обработки система позволяет хранить и легко выдавать множество различных данных. Ожидается, что система повысит скорость и точность учета и улучшит контроль за технологическим процессом, методы производства и соблюдение гарантий.

Была также разработана система контроля за материалом для тепловыделяющих элементов на основе двуокиси урана низкого обогащения. В случае периодического проведения процесса система позволяла учитывать продукт в любой момент времени с определенной точностью и степенью надежности, учитывая потери и отходы на любой стадии процесса. Система состоит из двух частей: СМОДНАП, которая выдает характеристики по всему заводу, и СБИДНАП, которая ведет учет продукта. В случае непрерывного процесса, система учитывала продукт путем контроля за потерями на каждом этапе, на котором подводится материальный баланс, и путем измерения исходных количеств. Система включает СБИДНАП-II, которая дает предостережение и рекомендации по технологическим условиям, необходимым для поддержания необходимого уровня надежности.

Для надежной работы вышеописанной системы весьма желательно, чтобы процедура взвешивания проводилась с помощью автоматической аппаратуры, соответствующим образом загерметизированной, без существенных помех для технологического процесса. Для удовлетворения этих требований был разработан и испытан прототип автоматического оборудования для взвешивания и регистрации таблеток из двуокиси урана, направляемых на операцию сборки – последний этап в процессе изготовления тепловыделяющих элементов, перед тем, как они помещаются в оболочку.

INVESTIGACION Y DESARROLLO EN EL JAPON SOBRE LAS SALVAGUARDIAS DE MATERIALES NUCLEARES EN PLANTAS DE FABRICACION.

Durante los últimos tres años, el Gobierno japonés ha tratado por todos los medios de crear un sistema exacto de contabilidad, para la salvaguardia de los materiales nucleares en las plantas de fabricación. Los explotadores de las instalaciones han venido trabajando sobre los puntos que se indican a continuación.

Por lo que concierne a los combustibles muy enriquecidos del tipo de placas se ha elaborado un método de control rápido y eficaz, en el que se utilizan minicomputadoras. El sistema se ha concebido de modo que se pueda realizar el tratamiento en tiempo real, manteniendo una memoria de gran capacidad y facilitando la salida de muchos datos diferentes. Con este sistema se espera poder aumentar la velocidad y exactitud del control contable, mejorar el control del proceso, las técnicas de fabricación y la inspección de salvaguardias.

Se ha elaborado asimismo un sistema de control de materiales, para los combustibles de UO_2 poco enriquecido. En funcionamiento discontinuo, se ha ideado un método para calcular con cierta precisión y fiabilidad el inventario de la planta en cualquier momento, dando los coeficientes de pérdida y de producción de desechos en cada una de las operaciones del proceso. Este método comprende un «SMODNUP», que da las características de la planta, y un «SBIDNUP», que calcula el inventario de la misma. En funcionamiento continuo, se ha concebido un método para calcular el inventario de la planta, dando el coeficiente de pérdida en cada zona de balance de materiales y controlando el caudal en este punto. El método comprende un SBIDNUP-II que da una señal de alarma y sugiere las operaciones a efectuar para conseguir una fiabilidad determinada.

Para ensayar la fiabilidad del sistema de control contable, es muy conveniente que los datos de las pesadas se obtengan automáticamente con equipos de medición precintados y que el procedimiento no complique

excesivamente el proceso de fabricación. Para ello se ha desarrollado y ensayado un nuevo equipo prototipo de pesada y registro para las pastillas de UO_2 durante el apilamiento, última etapa en el proceso de fabricación antes de introducir el combustible en las vainas.

1. INTRODUCTION

It is becoming more and more important for fuel plant operators to use an effective and reliable safeguards system for nuclear materials, in view of the growing emphasis on applying an international safeguards system including international inspection procedures.

The Japanese Government, aware of the situation as early as 1967, began analysing and developing a nuclear material accounting system for fuel cycles. Fuel manufacturers are concerned about this problem since the strict international inspection of nuclear materials may disrupt their commercial plant operations and may lead to the leakage of important commercial and technical information. To avoid any disparity between their standards and the standards set up by the international safeguards system for fuel plants, fuel plant operators are concentrating on establishing an effective and reliable nuclear material accounting system.

It is natural for fuel plant operators to wish to simplify the inspection techniques of the safeguards system as far as possible.

2. A NUCLEAR MATERIAL ACCOUNTING SYSTEM

2.1. A material control system for highly enriched plate-type fuel

A rapid and reliable data acquisition/distribution system for nuclear materials, which utilizes a minicomputer, has been developed for plants manufacturing highly enriched uranium-aluminium alloy plate-type fuels. The system is designed to be especially precise in the accounting of highly enriched uranium.

The function of the nuclear material accounting system is to acquire, evaluate and distribute information on the source data, such as location and form, of all the nuclear materials in the plant. The basic unit in the system is the subsidiary material accounting area (sub-area) in which the inputs and outputs of the nuclear material will be estimated by a suitable technique. The sub-area must be established to fit the requirements of an effective material accounting system and to take into account the physical location of equipment. Another consideration in defining sub-areas is the possibility of identifying the nuclear material entering, leaving or remaining in the sub-area. The entire plant can be considered as a single material balance area which combines all the sub-areas in the plant. It is possible in this way to account for the quantity of nuclear material at each stage of the sub-area by determining the material discrepancies in each sub-area.

In the nuclear material accounting system, nuclear material will be transferred in batches between the sub-areas. The processing of the nuclear material corresponds to the passage of the material through one sub-area. The plant is represented by a network of interconnected sub-areas which are considered mutually exclusive. The flow of the nuclear material in the plant is equal to the sum of the transfers between sub-areas.

The sub-areas will be determined in a way which corresponds to the manufacturing processes or to the storage of the materials. The inputs and outputs of nuclear material to and from the specified sub-areas are uniquely identified by the lot number in the sub-area. The weight of the nuclear material at the time of transfer between sub-areas cannot be determined at the exact time of transfer so the best estimated value must be used.

When sufficient data are available for maintaining a proper nuclear material accounting system, the material balance concept is applied to trace the flow of nuclear material with the lot number in the sub-area. This results in the estimation of the process loss and MUF (material unaccounted for) in the sub-area.

In this way, the system keeps track of the location, form, and quantity of all nuclear material in the plant.

The design of the nuclear material accounting system is based on the following criteria:

(a) All material must be located in certain sub-areas
(b) All material should be assigned a lot number and belong to one and only one sub-area at any specified time
(c) The sub-area must be truly representative of the current flow of the material in process
(d) The material balance should be maintained at each transfer of the nuclear material through every sub-area or several sub-areas. The sub-areas are mutually exclusive.

In this system, the process loss plus the MUF is equal to the product of the input and α (alpha), defined as the ratio of the difference between input and output to input. The value of α is determined empirically. The state of the operation is estimated on the basis of the difference between the empirically determined α and the actual α with respect to the specified lot number in the sub-area. The process loss and the MUF are controlled together because the accounting system is accurately maintained in the plant at all times and an inventory is conducted after every complete cycle in the manufacture of nuclear fuels.

The data acquisition system is designed with the following points in mind:

1. Economic considerations
2. Real time data processing and data storage
3. Large volume data storage
4. Decrease of error of input data
5. Data station for a large computer
6. Output record for specified requirements.

To satisfy these requirements, an M4-Databee system is used with a minicomputer (NEAC-M4) as the central processor unit, a magnetic drum (MD 400) as the memory, "Databee" as the terminal input source, and an input-output typewriter (150 B) as the input-output control device.

In this system, the operator can supply accounting data of each transfer and change in the nuclear material to the central processor unit with "Databee". This is designed to decrease the volume of input data by using coded cards for fixed data, tokens for semi-fixed data, and slide keys for variables.

The input data consists of the name of the campaign, the original location and the receiver's location of transfer, form, the number of lots, the quantity of the compounds and the U-contents. The name of the cam-

paign corresponds to the enrichment (and the origin) of nuclear materials. The material balance is kept in the weights of both U and ^{235}U.

The input information is checked with the record of nuclear criticality and capacity stored in the memory bank of the magnetic drum, and the result is shown by the lamp of "Databee". The input data are punched on the paper tape and typewritten on the paper. The data and other information put in the system by the operator are stored in the magnetic drum. In addition to the above input method, there is a sub-program which evaluates the U-contents in uranium-aluminium alloys by measuring the specific gravity of the alloys.

This program stored in the magnetic drum consists of nine sub-programs and is used as necessary. Its functions are as follows:

1. Reading input data from the terminal input equipment, filing, and storing them in the memory bank of the magnetic drum
2. Making an inventory list relating to the location, form, etc. of nuclear materials
3. Evaluating the amount of throughput in a given process in a specified lot number
4. Making the material balance table in a specified material balance area
5. Evaluating the loss, MUF, products, etc. in a specified process
6. Providing necessary information for formal reports
7. Adjusting and correcting the accounting data
8. Critical control in the plant.

The program is flexible enough to be widely used in various processes. For example, with minor changes it can be used in the manufacturing process of power reactor fuels as well as plate-type fuels. The program enables the accounting data to be recovered rapidly and precisely, and in addition is very useful, for example, for improving manufacturing techniques, process control of materials, and safeguards inspection.

2.2. A control system for low-enriched UO_2 fuel

For low-enriched UO_2 fuel plants, the safeguards problem is very important because the annual throughput is increasing rapidly as the demand for power reactor fuels increases. According to theoretical considerations, the loss amount during processing can be estimated on the basis of various assumptions and by various monitoring procedures in the material flow. However, in practice, it is almost impossible to measure the actual loss amount since "Loss" is mainly due to the material which cannot be located and identified during processing. The term 'MUF' has been introduced to define the difference between the book inventory and the physical inventory. The MUF approach used in this accountability system for a fabrication plant for low-enriched UO_2 fuel is generally considered to be an indispensable part of any safeguards system.

UO_2 fuel plants are classified into two types according to the mode of operation. One is operated according to the "batch-type" operation and the other according to the "flow-type" operation. A "batch-type" operation means that a nuclear material batch, such as a bottle of UO_2 powder, is maintained as a unit from the starting point through all the processes until it reaches the "pool" position where the identified batch will disappear

and a new batch will be created as the output from the "pool". As long as the material belongs to a specified batch, the material amount will be controlled and booked according to the specified batch itself, and the identification of each batch and the border line between two batches will be maintained exactly during the operation.

The system developed for the "batch-type" operation process consists of two different computer programs, SMODNUP (System of Modeling for Nuclear Plant) and SBIDNUP (System of Booking for Inventory and Distribution in Nuclear Plant). SMODNUP has two different functions. One is to establish a calculation procedure for the confidence levels of the accounting system (mainly the confidence levels for the book inventory) under specific plant conditions and operation modes for one specific project operation.

The plant conditions include such items as material flow sequence, equipment capacities in the process line such as throughput for the unit time operation, location and size of "pool" (usually similar to storage), and the same items in the recovery process line also.

The operation modes are characterized by the various states of the material in the process line, such as interim products, recoverable scrap, unrecoverable scrap, accountable loss, and by various operational rules in each process, such as range of batch size, identification of each batch, quantitative restrictions on rejected material with statistical value, and weighing procedures.

The other function of SMODNUP is to establish an accounting system by which the booking of the nuclear material in a plant can be performed and the book inventory and distribution in a plant can be calculated at a specific time with a certain confidence level and accuracy. The SBIDNUP program can calculate the book inventory and its distribution in the plant at a specified time with daily data from the plant by using the procedure set up by SMODNUP as mentioned above.

The accounting system for the batch-type-operated fuel plant can also be used for analysing the nuclear material accounting system in commercial operations because if there is any deviation from the normal operation procedure, as indicated by the calculations, an alarm will be given, thereby alerting the operator that a special survey of the process or accounting procedure should be conducted. In this way the safeguards accounting system can be combined with the control of the commercial operation of the plant. This is a basic consideration in the maintenance of an effective safeguards system and in the simplification of inspection techniques.

SBIDNUP, in calculating the book inventory and its distribution in the plant, will report the estimated book values and the physically measured values at key measurement points in the plant. The key measurement points can be selected as the starting point of the process in the plant, such as the powder receiving point, and the last stage of the process, such as the shipment of the fuel assembly out of the plant. These key measurement points shall be specified before the plant operation starts on a project. Other measurement points may be required in order to perform temporary physical measurements in accordance with the mode of operation in the plant. These temporary measurements shall be performed at a time when some deviation from the normal operation, such as any delay or malfunction in the process, occurs. Another measurement may be necessary at the inlet or outlet point of the "pool". Whether or not the measurement is necessary will depend on the quality and quantity of the material.

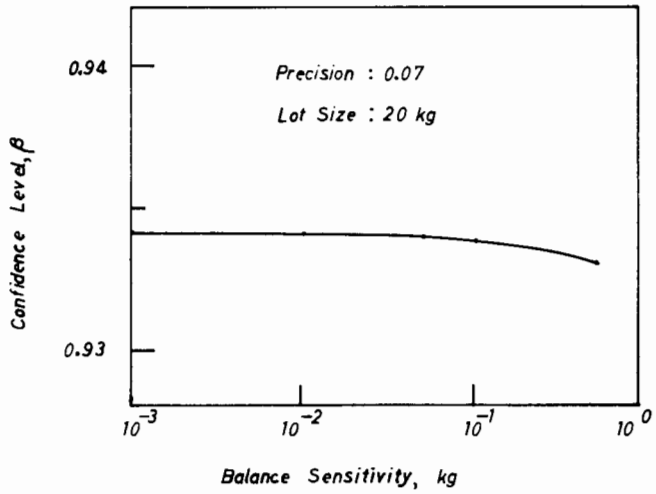

FIG.1. Effect of balance sensitivity at "input point" on confidence level at weighing point.

The confidence levels and the precision of the accounting system are mainly determined by plant characteristics and operational modes. For instance, at the weighing point at the end of the process line (such as the input to the pellet storage or the fuel assembly shipment point) a physical weighed value will be obtained. The accounting system will determine whether the difference between the calculated and the measured value is within the preset precision or not. The confidence levels of the accounting system will be reviewed at specified times. As long as the confidence levels and the precision are satisfactory, the accounting system is maintained and the nuclear material control in the plant is satisfactory both in terms of plant operation and safeguards. If these are found to be unsatisfactory, SBIDNUP will signal that a review of the process of the specified batch and the plant operation conditions should be carried out. This procedure is very effective when the plant operation is satisfactory, and the plant operator is assured that there has been no diversion of nuclear material for that period. There is some compromise between the application of the safeguards system and the commercial operation of the plant in this system.

Figure 1, an analysis by the SMODNUP program of balance sensitivity, shows that balance sensitivity at one weighing point will not affect the confidence levels of the accounting system to a significant degree. The confidence levels are affected mainly by the variance of loss and scrap rates, specified or assumed. This means that the loss and scrap rates and their variance should be analysed very carefully for each project operation if the system is to be maintained in operation. A slight increase in balance sensitivity would not improve the accounting system confidence levels under these conditions.

For a UO_2 fuel plant with "flow-type" operation, different considerations are required in the accounting system. A "flow-type"-operated plant does not have "process" and "pool", but has some sub-material balance areas corresponding to individual process stages or series of

process stages. In each sub-material balance area, input, output, and rejected material such as scrap, loss and sample, are terms in the material balance equation. The accounting system makes use of a group of sub-material balance areas which may form one material balance area (MBA). The storage area and the process area can each be considered particular MBAs. It is not necessary to identify all materials in the MBA with a specific input batch.

The SBIDNUP-II program has been developed as an accounting system in "flow-type"-operated plants. The program will calculate the inventory both in each sub-material balance area and also the inventory in each group of sub-material areas (MBAs) and its distribution in the plant with the preset loss and scrap rates. The report consists of actual weighing values and calculated values. A check on the confidence level will be performed in each sub-material balance area. The confidence level of the accounting system in each sub-material balance area is previously determined and the system is designed to keep these areas in the preset range of the confidence level and precision. If the calculated values are worse than the preset values, a warning notice is printed out that a check of plant operations should be conducted. The notice will identify the sub-material balance area concerned instead of the batch number as in SBIDNUP. The printed alarm notice advises the plant operator to begin the "reset operation" for the specified sub-material balance area, i.e. to stop the operation in order to take a physical inventory to determine the value of MUF, and to clean up the area. The SBIDNUP-II program is more comprehensive than SBIDNUP because it provides the plant characteristics and calculates the inventory.

In both cases, the reliability of the accounting system in the plant during the project operation is continuously reviewed and checked, by controlling the confidence levels and precision of the system; this includes the control, essential in any nuclear material accounting system, of the MUF values in the plant.

Efforts to clarify the characteristics of MUF in regard to structure, dependency on other conditions, variance in the time sequence, causes and their relationship, still continue. Plant operators hope that in time more efficient ways will be found to implement the safeguards system and inspection techniques.

Although SMODNUP and SBIDNUP have been developed as nuclear accounting systems for the batch-type-operated fuel plant and SBIDNUP-II for the flow-type-operated plant, these systems are based on purely mathematical considerations. Further trials using the systems in actual plants must be carefully carried out and evaluated.

3. PROTOTYPE OF AN AUTOMATIC BALANCE FOR UO_2 PELLETS

It is desirable in a material accounting control system for weighing data at key measurement points in a fabrication plant to be obtained automatically and also for the procedure not to interfere with the operation of the plant.

To fulfil these requirements, a prototype of new weighing and recording apparatus has been developed and tested for UO_2 pellets during the stacking process which is the last step in the fabrication of UO_2 pellets. It includes both a balance and an instrument which is able to make a record stating the quantity of UO_2 treated in the process.

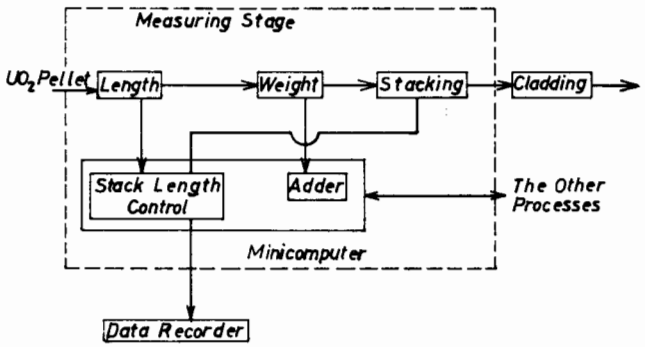

FIG. 2. Block diagram of arrangement of the equipment.

The pellet handling processes concerned must be automated to eliminate human error, thus ensuring the precision of the original data. Manual operations must be avoided.

This equipment consists of a mechanism for transferring the pellets, instruments for measuring the length and weight of the pellets, a computer, controllers and a data recorder. The block diagram of the arrangement of the apparatus is given in Fig. 2.

A pellet is conveyed from the hopper to the length-measuring stage every 5 seconds, and its length is measured automatically by the transducer head (differential transformer) connected with the A-D converter. This information is then sent to the minicomputer. With the help of the computer, the stack length is calculated and controlled, and the pellet is transferred to the next stage where it is weighed. The computer controls the adjustment of the stack length by ordering specific pellets provided from the sub-hoppers. In the weighing step, several pellets are weighed together by the automatic balance. The weight information is then sent to the minicomputer, edited and typed out when required. The steps in which the length and weight are determined are under the control of the computer throughout the production process. Because the system must satisfy contradictory conditions, that is wide range and high precision, the measuring instruments are calibrated with master standards before every stacking operation. Their values are memorized by the computer cores and used to correct the data for each program. Every effort is made to ensure that correct data concerning both equipment and program is collected.

To ensure the correctness and high reliability of the data, the area shown within broken lines in Fig. 2 should be completely automated. So several pre-tests have been carried out. After the equipment is assembled, a long-term test operation is carried out under various difficult conditions until standards of endurance, stability and controllability are fully satisfied.

To use these automatic balances in providing an effective safeguards system, it is preferable for them not be applied at arbitrary points in the manufacturing line but to be installed at each key measurement point in the specified material balance area. This apparatus is only the first step in the above system.

The design philosophy and techniques acquired through the development of the prototype will be helpful in the development of an overall safeguards system.

4. CONCLUSION

Three different fuel plant operators in Japan are making great efforts to develop an effective safeguards system and a convenient inspection procedure which avoids any unnecessary disturbance to commercial plant operations. The clarification and understanding of the problems concerning MUF control are considered very important by plant operators, who are very anxious to continue their efforts to develop and perfect a nuclear material accounting system for fabrication plants.

DISCUSSION ON AGENDA ITEM 5.2

Safeguards techniques and instrumentation

DISCUSSION ON ALL THE PAPERS IN THIS AGENDA ITEM:

P/842	*USA*	*Presented by G.R. Keepin*
P/809	*FRG*	*Presented by C. Weitkamp*
P/286	*Belgium*	*Presented by Ch. Beets*
P/415	*FRG*	*Presented by D. Gupta*
P/773	*IAEA*	*Presented by W. Gmelin*
P/101	*USA*	
P/265	*Japan*	

F. MORGAN: The papers presented by Mr. Keepin (P/842) and Mr. Weitkamp (P/809) deal with nuclear materials management and describe methods employed principally by the operator. Safeguards inspection, on the other hand, is a verification process. How is such verification to be carried out – by observing calibrations, by sending samples away for analysis or by cross-checking with the help of portable instruments?

G.R. KEEPIN: Much of the emphasis of the United States program for developing non-destructive assay instrumentation is on materials management, quality and process control and safety in various types of nuclear facility. Verification by an international inspectorate (in this case the International Atomic Energy Agency) is seen as a cross-checking procedure superposed on an already existing on-line measurement and material balance system; the inspector would check on the calibration of instruments with a set of his own standards and by sending samples to a central analytical laboratory for independent assay. Such a laboratory could be controlled either by the international inspection organization or by a national government.

At all events, it is clearly in the economic interest of nuclear facility operators to have an efficient materials control system, which in turn should facilitate the implementation of safeguards and render them more effective.

C. WEITKAMP: There is little to add to Mr. Keepin's answer. All the procedures mentioned by Mr. Morgan – including the use of portable instruments – have their place in safeguards, and the fact that we have not discussed them in greater detail in our paper does not mean that we discount them.

M. DAVIS: The authors of Papers P/842, P/809 and P/286 speak with confidence of the instrumental methods which they have developed for

materials accounting and safeguards. Could they please give us their views regarding those problems which have not yet been solved?

C. WEITKAMP: I think I can best answer this question by saying that every measuring problem can be solved if one is satisfied with the accuracies obtained or if one lets the instrument makers decide how much money is necessary in order to achieve the desired improvements. Perhaps Mr. Keepin, who presented paper P/842, would care to expand on my reply.

G.R. KEEPIN: The poorly defined geometries which characterize typical scrap and waste configurations present what is probably the most difficult and challenging assay problem today, for they are the most susceptible to matrix and geometric perturbations, the most difficult to formulate standards for and – in many United States facilities – the greatest source of MUF. Accordingly, much of the United States research and development effort in the non-destructive assay field is directed towards the increasingly important problem of scrap and waste assay. At the same time, current concern about the environment has lent emphasis to the need for the tighter control and more accurate measurement of fissionable waste.

Another area of current research and development effort which might be mentioned is the quantitative measurement of fuel sub-assemblies and complete fuel elements.

A long-range goal of research and development in the field of non-destructive assay is to provide an economic means of increasingly accurate rapid measurement which is competitive with or superior to the more conventional methods of analysing fissionable materials.

Ch. BEETS: In Belgium, we are co-operating with the International Atomic Energy Agency in trying to solve a number of outstanding problems. Our efforts are concentrated on the fuel cycle of the BR 2 reactor (a materials testing reactor) and on the fabrication of PWR fuel (slightly enriched UO_2). So far, only passive assays have been carried out.

D. GUPTA: In the range of accuracies mentioned in Paper P/842, are the systematic error components also included?

G.R. KEEPIN: It is difficult to answer this question briefly. Let me say, however, that the range of accuracies achieved by non-destructive assay and cited in our paper includes not only the random errors (precision) but also an estimate of the systematic error component (bias), the latter being as a rule comparatively small. All non-destructive assay measurements are made relative to a standard, so that all quoted accuracies are based on comparison with the standard used. The standard itself is often extremely difficult to characterize accurately, due to problems inherent in the chemical analysis, the mass spectrometric measurements or whatever method is used to establish the fissionable material content of the standard. Thus, when we cite an accuracy of 1% or better for small, well-characterized samples, we mean that the unknown can be determined to within 1% – relative to the standard used.

In the case of poorly defined samples, the errors can amount to several per cent (up to 20%, depending on the composition and geometrical configuration of the sample being assayed).

Apart from problems of standards calibration, there are, of course, the errors inherent in the assay instrument itself (instabilities, drift, background, counting statistics and so forth); these may have both random and systematic components. Such error contributions are exclusive to a given type of instrument, the standard used and the assay problem under

consideration, so that it is very hard to generalize. Nevertheless, an attempt has been made to present the best estimate of the random-plus-systematic errors for each type of non-destructive assay instrument discussed in the paper.

F. MORGAN: Isotopic correlations such as those noted in Paper P/415 have been studied extensively over the years and there can be little doubt that, for complete cores of a standard reactor, the correlations are fairly good – perhaps to within a few per cent. There are, however, pronounced flux variation and temperature effects which have to be taken into account, particularly in dealing with the partial reprocessing of a reactor core. Plutonium-240/239 can change by 10-20% as the temperature increases from 580 to 660°K; similar effects occur with plutonium-241/239 and plutonium-242/239.

A more important point is that the paper introduces a generalization which may be misunderstood: that isotopic correlations 'enable the safeguards authority to dispense with the verification of tank volume and density measurements completely'.

A fundamental question is involved here. In a reprocessing plant, there are two well-established and primary methods of estimating the material input: multiplication of the observed concentration and the volume; multiplication of Pu/U and the weight of the uranium input. The paper really deals only with the second of these, and the above-mentioned generalization has little to do with isotopic correlation. The two methods are of almost equal validity, but may give different answers for total plutonium. A discrepancy requires a valid explanation, for neither one answer nor the other can be rejected out of hand; a weighted average might be appropriate. They have to be used to calibrate absolutely the method suggested, which can therefore be inherently of no better accuracy. Accordingly, I consider that isotopic correlations may be of assistance in resolving discrepancies, but cannot be regarded as prime data.

D. GUPTA: It is true that the ratios mentioned by Mr. Morgan may change with the changing operating parameters of a reactor. It is also true that the accuracy of the isotopic correlation method cannot be better than that obtained by mass-spectrometric and volumetric measurements if these methods are used to calibrate the original correlation curves.

However, the purpose of using correlation techniques was not to improve the accuracies of material balances but to reduce the safeguards effort at the same level of material balance accuracy – and this appears possible.

There are other methods of calibrating the original correlation curves. One method, based on an approximate knowledge of conversion and burn-up factors, was elaborated in detail by Häfele and Nentwich at the IAEA Symposium on Progress in Safeguards Techniques held in Karlsruhe in 1970 (Ref. [21] of our paper). When this method is used, volumetric and density measurements of the dissolved solution can in fact be dispensed with for safeguards purposes.

Lastly, as is pointed out in the paper, the reduction in safeguards effort achieved by the use of isotopic correlations is low – less than 3% of the total safeguards effort.

C. WEITKAMP: Was the NMIS system described in Paper P/101 set up mainly for safeguards?

T.J. HAYCOCK, Jr.: The NMIS system was established initially for safeguards, but we soon found that, while safeguards people could use

information compiled for other purposes, others could also use information compiled for safeguards. For example, we supply data to the International Atomic Energy Agency and to many divisions of the USAEC (Production, Waste and Scrap Management, Reactor Development, Research, Forecasting and Analysis, Finance). However, the greatest need for timely and accurate data is in safeguards.

The inventory and other information is not only supplied to users but also analysed and reviewed by computers. For example, all shipper-receiver differences and inventory variations are analysed, the results being made available in appropriate reports.

W. HÄFELE: Could Mr. Gmelin (P/773) tell us the definition of LRIE?

W. GMELIN: LRIE has been defined as the minimum level of inspection effort at a facility necessary to establish the international credibility of Agency safeguards. According to our approach, the specific situation at each facility has to be considered in order to determine the minimum number of activities needed for Agency verification, and especially to enable the Agency to evaluate statements on MUF and its related limits of uncertainty. LRIE is consequently the inspection effort necessary to carry out the minimum number of activities.

D. GUPTA: Did you estimate the uncertainties in the establishment of a material balance for the two extreme cases (namely MRIE and LRIE)?

W. GMELIN: The uncertainties mentioned in the paper reflect the present capability of measurement systems. In our paper, we considered that these uncertainties, together with the measurement values, would be verified according to the procedures described. For example, the input accountability at a reprocessing plant could be verified by the combination of isotope correlation techniques with a 95/95 random sampling scheme as described in the paper.