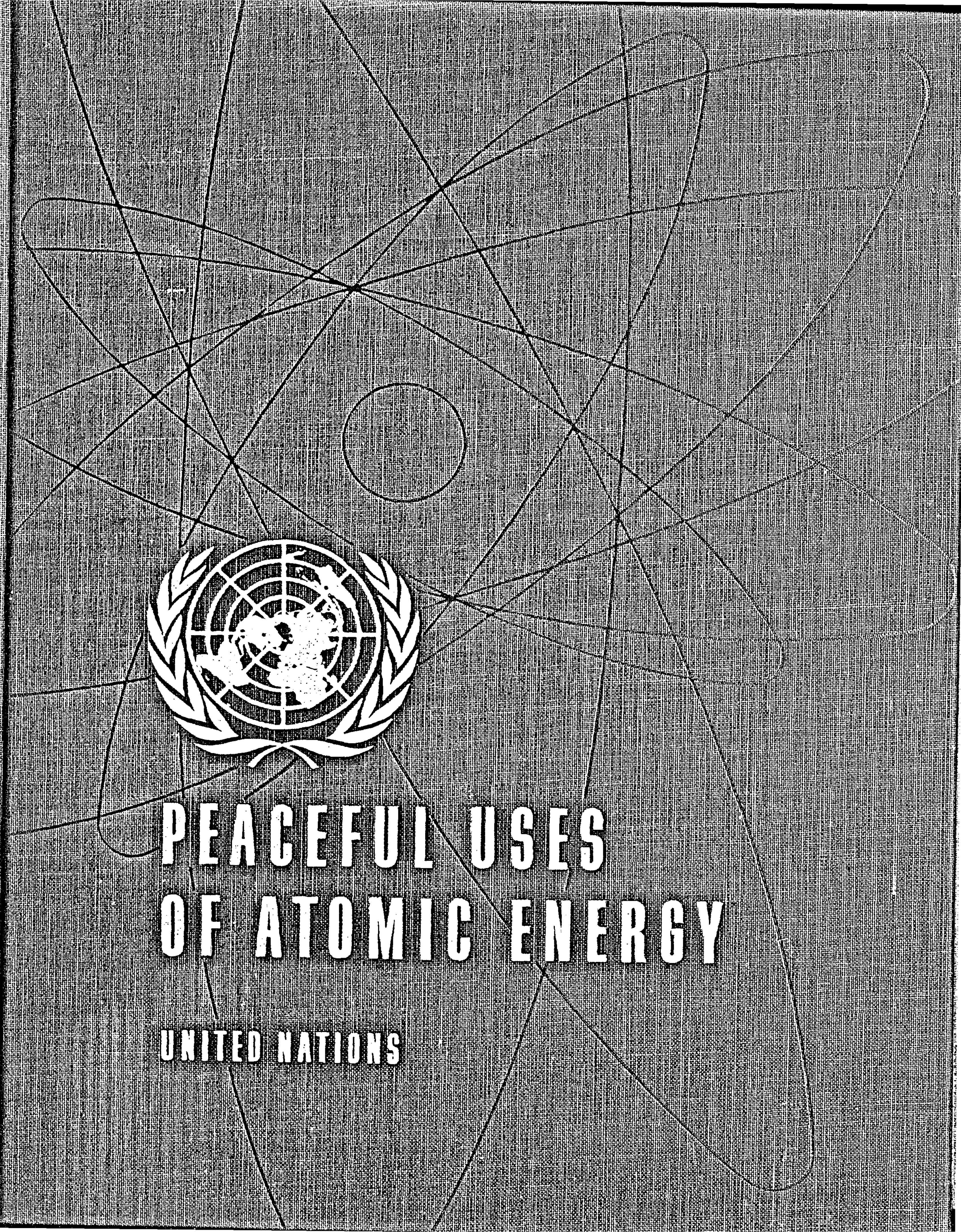

PEACEFUL USES OF ATOMIC ENERGY

UNITED NATIONS

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OF THE
INTERNATIONAL CONFERENCE ON THE PEACEFUL USES OF ATOMIC ENERGY

Volume 14

GENERAL ASPECTS OF THE USE OF RADIOACTIVE ISOTOPES: DOSIMETRY

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on the
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General Aspects of the Use of Radioactive Isotopes: Dosimetry

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PREFACE

The Proceedings of the International Conference on the Peaceful Uses of Atomic Energy are published in a series of 16 volumes, as follows:

Volume Number	*Title*	*Sessions Included*
1	The World's Requirements for Energy; The Role of Nuclear Power	2, 3.2, 4.1, 4.2, 5, 24.2.
2	Physics; Research Reactors	6A, 7A, 8A, 9A, 10A.1.
3	Power Reactors	10A.2, 3.1, 11A, 12A, 13A, 14A.
4	Cross Sections Important to Reactor Design	15A, 16A, 17A, 18A.
5	Physics of Reactor Design	19A, 20A, 21A, 22A, 23A.
6	Geology of Uranium and Thorium	6B, 7B.
7	Nuclear Chemistry and the Effects of Irradiation	8B, 9B, 10B, 11B, 12B, 13B.
8	Production Technology of the Materials Used for Nuclear Energy	14B, 15B, 16B, 17B.
9	Reactor Technology and Chemical Processing	7.3, 18B, 19B, 20B, 21B, 22B, 23B.
10	Radioactive Isotopes and Nuclear Radiations in Medicine	7.2 (Med.), 8C, 9C, 10C.
11	Biological Effects of Radiation	6.1, 11C, 12C, 13C.1.
12	Radioactive Isotopes and Ionizing Radiations in Agriculture, Physiology and Biochemistry	7.2 (Agric.), 13C.2, 14C, 15C, 16C.
13	Legal, Administrative, Health and Safety Aspects of Large-Scale Use of Nuclear Energy	4.3, 6.2, 17C, 18C.
14	General Aspects of the Use of Radioactive Isotopes; Dosimetry	7.1, 19C, 20C.
15	Applications of Radioactive Isotopes and Fission Products in Research and Industry	21C, 22C, 23C.
16	Record of the Conference	1, 24.1, 24.3.

These volumes include all the papers submitted to the Geneva Conference, as edited by the Scientific Secretaries. The efforts of the Scientific Secretaries have been directed primarily towards scientific accuracy. Editing for style has been minimal in the interests of early publication. This may be noted especially in the English translations of certain papers submitted in French, Russian and Spanish. In a few instances, the titles of papers have been edited to reflect more accurately the content of those papers.

The editors principally responsible for the preparation of these volumes were: Robert A. Charpie, Donald J. Dewar, André Finkelstein, John Gaunt, Jacob A. Goedkoop, Elwyn O. Hughes, Leonard F. Lamerton, Aleksandar Milojević, Clifford Mosbacher, César A. Sastre, and Brian E. Urqnhart.

The verbatim records of the Conference are included in the pertinent volumes. These verbatim records contain the author's corrections and, where necessary for scientific accuracy, the editing changes of the Scientific Secretaries, who have also been responsible for inserting slides, diagrams and sketches at appropriate points. In the record of each session, slides are numbered in numerical order through all presentations. Where the slide duplicates an illustration in the submitted paper, appropriate reference is made and the illustration does not appear in the record of the session.

Volume 16, "The Record of the Conference," includes the complete programme of the Conference, a numerical index of papers and an author's index, the list of delegates, the records of the opening and closing sessions and the complete texts of the evening lectures.

TABLE OF CONTENTS

Session 7.1: Isotopes in Technology and Industry *Page*

Session 19C: General Uses, Production and Handling of Radioactive Isotopes

Session 20C: Dosimetry

Session 7.1

ISOTOPES IN TECHNOLOGY AND INDUSTRY

LIST OF PAPERS

Importance of Isotopes in Technology and Industry

By P. C. Aebersold,* USA

Radioisotopes are exceedingly abundant and widely useful byproducts of nuclear reactors. They are a peaceful development of atomic energy that is already bringing rich benefits to humanity. Reactor radioisotopes have now been distributed extensively for nine years and their use has grown rapidly. Yet, they are still far from attaining their vast world-wide potential for gaining new knowledge and improving health and welfare.

As has been true for other nuclear developments, the concepts, discoveries and instrumentation which have led to the production and use of radioisotopes have been international. It is not necessary here to review the long and important role of radioactivity in science, medicine and industry, or how it has been the key to all nuclear discovery—from the discovery of radioactivity itself, to the nuclear atom, and eventually to uranium fission. It is interesting to note however that the one element, uranium, has had both a singular and rejuvenated role as a source of radioactivity. Uranium first brought to man's attention in 1896 the phenomenon of radioactivity, in itself a form of nuclear energy (although not then recognized as such); now almost 60 years later it provides by means of the uranium chain reaction a tremendous source of both useful energy and a broad array of new radioactive species. Uranium, source of many useful natural radioisotopes, has thus also become through the reactor the major source of the more numerous and more widely applicable man-made radioisotopes.

Since the discoveries and inventions basic to production and use of radioisotopes have been international and are well known to technical persons, they need not be referred to here. The published works related to radioisotopes, including characteristics, production, measurement, techniques, radiological safety and utilization in all fields of application, constitute an enormous world-wide literature. It would be almost impossible to give proper credit for or even to list major publications pertinent to all developments or topics discussed in this broad survey paper. Literature references will thus not be attempted.

Furthermore, in presenting statistics on distribution or illustrations of usefulness it has been necessary to refer to the program and experience in the USA. This is because of greater familiarity with that program and is not due to lack of recognition of extensive distribution by several countries such as Canada, Great Britain, France and Norway, and use by over 50 nations throughout the world.

Since most uses discussed in this paper are in technology and industry, they will of course have more application for nations with greater technological development. The basic principles of use may however be applied to special problems of many nations. Uses in science, agriculture and medicine will of course have universal application.

No single measure can be given for evaluating the importance of tools of research and application so variedly and extensively used as isotopes. The endeavor to summarize the importance in this paper will be made on the basis of the following:

1. Great inherent power of isotopes in tracer or indicator techniques.
2. Tremendous range in intensity and radiation energy of radioisotope sources.
3. Rapid growth in utilization.
4. Wide applicability of basic principles of use.
5. Specific illustrations of usefulness in various fields.
6. Present and potential value of uses.
7. Significance of isotope techniques and applications in the over-all development of peaceful use of atomic energy.

POWER OF ISOTOPES AS TRACERS

The sensitivity of the isotope tracer technique depends on detection of the isotopic label after dilution with other atoms of the same element. Dilution with atoms of dissimilar elements can usually be eliminated if necessary by chemical separations.

If a stable isotope with a natural abundance of about 1%, such as C^{13}, were concentrated to 100% abundance, it could be diluted only about 104 times with normal carbon and still be detected. This is of course suitable for some experiments, but is a serious limitation for many. A stable isotope with a higher natural abundance would have a smaller limit of possible dilution. Deuterium, with a natural abundance of only 0.015% and obtainable almost isotopically pure, presents a most favorable stable isotope tracer. In this case, using a mass spectrometer, a dilution with normal hydrogen by almost 10^6 times is detectable although with difficulty.

The dilution attainable with a radioactive tracer depends on available activity per gram of the ele-

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* US Atomic Energy Commission.

ment, efficiency of the detector and reliability of detection above the natural radiation background. For example, "carrier-free" or 100% phosphorus-32, has an activity of about 2.9×10^5 curies/gm or 6.4×10^{17} disintegrations/min/gm. With ordinary Geiger counter methods one can readily detect less than 20 counts/min above background or around 10^{-16} grams of P^{32}. If the sample is diluted with stable phosphorus, self-absorption will reduce the amount detectable. However, self-absorption is greatly reduced and counting efficiency increased by new methods where the radiomaterial is placed inside a liquid scintillation medium or gas counter. Using latest low-level counting techniques, in which the natural radiation background is minimized by shielding and anticoincidence methods, it is possible to measure radioisotope samples containing only a few disintegrations/min/gm. For carrier-free P^{32} this would in theory permit a dilution with ordinary phosphorus of about 10^{17}.

Even if this dilution is "theoretical", it is obvious that P^{32} can be readily detected after tremendous dilution. Many other useful isotopes, such as I^{131}, have very high carrier-free activity and can be detected with comparable sensitivity.

In case the element has a radioactive isotope in nature, as for example H^3 and C^{14}, this activity becomes an inescapable part of the natural background for this element. For "living" (biosphere) carbon the activity is about 13 dpm/gm, whereas for carrier-free C^{14} the activity is about 10^{13} dpm/gm. Theoretically, therefore, C^{14} could be traced after dilution in "living" carbon almost a million million times. Tritium has an activity of about 2×10^{16} dpm/gm, and its natural specific activity is extremely minute, consequently it can be traced through an even greater dilution than C^{14}. Such sensitivities of detection are millions of times greater than for any other means.

In addition, the isotopic labeling technique can be unquestionably specific. An isotope of an element is chemically identical with the element, except for a small "isotope effect" in chemical reactions affected by difference in isotope mass. The isotope effect is demonstrated by altered reaction rates, but not by a qualitative difference in mechanism of reaction. Difference in reaction rates is considerable only for isotopes of great mass difference, such as those of hydrogen, but is demonstrable for isotopes of carbon. The effect can be taken into account however when it affects quantitative experiments.

Several criteria must be met for an isotope to serve as a "perfect" tracer. The dose of radiation or amount of chemical added must not affect the reaction studied. Also the isotopic label must be in the proper chemical state, or affixed in the proper molecular location. These criteria however are in general readily satisfied.

Available counting techniques permit almost any tracer experiment to be accomplished with amounts of radioactivity very much less than might affect the system. To avoid adding chemically toxic amounts of the tracer, it is often necessary to have very high specific activity material. Radioisotopes made by transmutation or fission are obtainable almost carrier-free, hence specific activity is generally no problem. Radioisotopes produced by neutron absorption may have to be irradiated in a high flux reactor to meet this criteria. Reactors with fluxes in the range of 10^{13} to 10^{14} neutrons/cm^2/sec produce sufficiently high specific activity in even the most toxic elements.

Techniques of multiple labeling with isotopes provide additional means of achieving specificity in tracer experiments. Different portions of a molecule can be labeled simultaneously with two or more isotopes. Double or even triple labeling of organic compounds is accomplished with such isotopes as H^2, H^3, C^{13}, C^{14}, N^{15}, P^{32} and S^{35}. Such labeling permits simultaneously tracing individual portions of a complex molecule, thereby checking molecular decomposition as well as eliminating the question of exact reproducibility of experiments.

It is usually possible to identify positively a radioactive isotope by its radiation and/or half-life. With proportional or scintillation counters and pulse-height discrimination circuits, it is often possible to use several radiotracers simultaneously in an experiment and count them separately without chemical separation. Further, with such discrimination equipment, it is possible to do accurate neutron activation analysis. The material to be analyzed is irradiated in a reactor, and the induced radioactivities are differentiated. This technique can identify parts per million or even million million of certain elements. The method has limitations however. A high flux of neutrons is required; the unknown element must be suitably activated; and too many radioisotopes with similar radiation characteristics must not be produced simultaneously.

Other special powerful analytical procedures are possible with radioisotopes. Mention should be made of various types of "isotopic dilution" techniques. An isotope-labeled compound of known isotopic ratio may first be added to a system containing the unlabeled compound. Then after thorough mixing, an isotopic analysis of the altered ratio for an unknown portion of the total gives the dilution of the isotope label and thereby a quantitative measurement of the total amount of the compound originally present. This technique is useful when the system is very large or the compound cannot be quantitatively extracted.

A similar isotope technique is used to determine the success of chemical purification. The desired pure compound is properly synthesized with a radioisotope label. This is then mixed homogeneously with the compound being purified, and the specific activity determined. If after repeated purifications the specific activity of the compound remains constant, maximum purification has been achieved with the procedure. Use of this technique, along with chromatographic, electrophoresis and ion exchange separations, reveals constituents below conventional criteria of chemical purity.

Autoradiography should also be noted as a sensitive and detailed detection for radioisotopes. It uses a photographic emulsion to obtain a picture of the distribution of radioactivity, for example, detailed location of radiomaterial in tissues or cells, or in grain structure of metals. With fine-grain nuclear emulsions, resolution can be achieved down to about 5 microns. A new high-resolution wet-process autoradiography uses a very thin layer of sensitized silver bromide capable of giving resolution down to one micron. Although this is far from the resolution obtainable with good microscopes, when combined with the tracer technique it provides information not obtainable in other ways.

Great versatility is possible in the labeling of materials with isotopes. Useful radioisotope tracers can be produced for most elements. Fortunately, for those elements having radioisotopes with half lives too short for tracer work, such as nitrogen and oxygen, one can use concentrated stable isotopes (N^{15} and O^{18}). In a few cases such as fluorine and copper, however, stable isotope tracing is not feasible, and the radioisotope also has a limited half life. Aluminum was in the latter category, but recently a long-lived isotope Al^{26} has been produced by cyclotron bombardment of magnesium and, although obtainable only in low yield, may provide a tracer for this important element. The over-all situation for tracing elements is thus most fortunate. Fortunately also, most of the important radiotracers can be made with a reactor.

Further, methods have been devised for synthesizing a large number of isotope-labeled compounds, either by chemical or biological means. Over 1000 labeled compounds are now readily available. These include biochemicals, pharmaceuticals and industrial chemicals. Techniques have also been devised for labeling a wide variety of organisms such as blood cells, bacteria, viruses, and higher organisms. Fabricated materials, such as piston rings and machine tools, can be activated directly by irradiation in the reactor. The total result is a very large number of elements, compounds, materials and organisms which have been made radioactive for tracer studies.

The radiotracer technique thus has the combined power of tremendous sensitivity, unquestioned specificity and wide versatility. Such a great extension of power of observation supports the prediction that isotopes may become a scientific tool as routinely and widely necessary as a microscope.

WIDE SCOPE OF RADIATION SOURCES

In addition to use as tracers, radioisotopes have another broad and important category of application as sources of radiation. For this use chemical identity is not the major interest but rather the character and output of radiation of the radioisotope. Here also radioisotopes are very versatile. They provide alpha, beta, gamma and in some cases X-ray sources. Also, by secondary means they may provide X-ray and neutron sources. The range of available radiation energies and half lives is broad enough to cover requirements for a large variety of practical uses.

Reactors provide radioisotopes in far greater intensities and at much lower cost than do natural sources or particle accelerators. The tremendous production capacity of the reactor, while very desirable to produce isotopes even for tracer uses, is absolutely essential for wide-scale application of radioisotopes as radiation sources. For example, the activity of gamma-ray emitting isotopes now used in radiography totals thousands of curies, and for teletherapy of cancer the ultimate use will total hundreds of thousands of curies. Large-scale sterilization of foods and drugs would require millions of curies.

The only reactor-produced alpha-ray emitter now used widely in industry is polonium-210. It is not only useful as an alpha source, as in self-luminous compounds and static eliminators, but is extensively used with beryllium for neutron sources. Reactor production far surpasses the normal supplies of natural polonium and at much less cost. Po-Be sources containing 50 curies or more per source are readily available. The yield of neutrons averages about 2.3×10^6 n/sec/curie. Many Po-Be neutron sources are used for neutron "logging" of oil wells and other techniques which depend on neutron reflection from hydrogenous material.

Other alpha-ray emitters, such as plutonium-239, can be produced by reactors. Some of these would have certain advantages over polonium, particularly in longer half life, but their applications as sources await further development.

Table I is a list of some important (because of large production or number of uses) reactor-produced pure beta-ray emitters. Many others are available but those listed cover beta-ray energies from 19 kev for

TABLE I. Important Reactor-Produced Pure Beta-Ray Emitting Radioisotopes

Element	*Isotope*	*Half-life*	*Energy of radiation*
Hydrogen	H^3	12.5 yr	0.019 Mev
Carbon	C^{14}	5700 yr	0.155
Phosphorus	P^{32}	14.3 d	1.712
Sulfur	S^{35}	87.1 d	0.166
Calcium	Ca^{45}	164 d	0.25
Strontium	Sr^{90}	28 yr	2.24,* 0.54
Thallium	Tl^{204}	4.0 yr	0.77

* From daughter Y^{90}.

H^3 to 2.24 Mev for the Y^{90} daughter of Sr^{90}. The half lives also have a range from 14.3 days for P^{32} to about 5700 years for C^{14}. The isotopes from H^3 to Ca^{45} are all routinely produced by transmutation and have very high specific activity. They also constitute isotopes of body elements and accordingly are widely used in biology and medicine.

Sr^{90} is extracted from uranium fission wastes and Tl^{204} is produced by reactor irradiation of thallium. These two beta emitters find wide industrial use at present, because of the combination of long life and energetic radiation. For some industrial applications, however, the low-energy beta rays of H^3 are useful. Because H^3 can be produced cheaply, is comparatively safe to handle and has a desirable half life, it should come into extensive industrial use.

spectra; consequently many industrial applications can be expected.

The wide interest in Co^{60} makes appropriate some remarks about its production. It is routinely produced with many times more gamma-ray output per gram than radium. For radiography, Co^{60} with specific activity of 1 to 10 curies/gram (c/gm) is excellent, whereas for teletherapy units material of 50 c/gm or higher is desired. A 1000-curie Co^{60} source has been produced with specific activity of 100 c/gm, about 10% of the stable cobalt having been converted to Co^{60}. Canadian and USA reactors are now scheduled to produce over 100,000 curies per year of high-specific-activity cobalt. It would be feasible to produce over 10 times this activity of Co^{60} per year, especially at lower specific activity.

TABLE II. Important Reactor-Produced Beta and Gamma Ray Emitting Radioisotopes

Element	*Isotope*	*Half-life*	*Energy of radiations*†	
			Beta	*Gamma*
Sodium	Na^{24}	14.9 hr	1.39 Mev	2.76, 1.38 Mev
Iron	Fe^{55}	2.91 yr	none	*K* (6.2-kev X-rays)
Iron	Fe^{59}	46.3 d	0.46, 0.27 1.56	1.10, 1.29, 0.19
Cobalt	Co^{60}	5.2 yr	0.32	1.33, 1.17
Iodine	I^{131}	8.05 d	0.61, 0.34, 0.25, 0.81	0.36, 0.28, 0.64, 0.08
Cesium	Cs^{137}	33 yr	0.518, 1.18	0.662*
Thulium	Tm^{170}	127 d	0.97, 0.88	0.084
Iridium	Ir^{192}	74.5 d	0.67, 0.54, 0.24	0.32, 0.31, 0.30, 0.20
Gold	Au^{198}	2.69 d	0.97	0.411

*From daughter Ba^{137}.

† Radiations are listed in order of abundance.

Table II is a list of currently important gamma-ray emitters which are producible in high intensities and large activities by the reactor. Other gamma-ray emitters, especially those from fission products, can be produced cheaply in quantity and may in time become important gamma sources. Rarely do radioisotopes emit only gamma rays without beta rays; however, for most uses the beta rays are inconsequential or can be filtered out. Fe^{55} is an isotope with no beta rays, which, as the result of *K* electron capture, gives soft manganese X-rays that have special applications.

The gamma-ray energies in Table II range from 0.084 Mev for Tm^{170} to 2.76 Mev for Na^{24}. For biological and medical purposes, including therapy, the short-lived isotopes, such as Na^{24}, I^{131}, and Au^{198}, are extremely useful. Co^{60} and Cs^{137} find much industrial use because of their long life and high-energy radiation. Tm^{170} is useful for its very low-energy gamma ray, but its half life of 127 days calls for frequent source replacement.

It should be noted that beta rays can excite X-rays (bremsstrahlung) and that low-intensity X-ray sources can be obtained from high-intensity beta-ray sources. Lack of long-lived low-energy gamma-emitting isotopes has prompted the development of such beta-ray excited X-ray sources. These sources can have a long life and a wide range of energy

Very great amounts of fission products have already been produced as byproducts of plutonium production, and tremendous amounts will result as reactors are increasingly used for power. Because of this, much attention has been given to development and uses of fission product sources. Of greatest present industrial interest are Cs^{137} for gamma sources and Sr^{90} for beta sources. This is mainly because of their long useful lives. Other fission products also will no doubt become useful in large quantity if they can be obtained economically.

A new multikilocurie fission product processing plant is under construction at the Oak Ridge National Laboratory and should be operating in late 1956. This plant is designed to separate and purify many thousands of curies of important long-lived fission products and fabricate them into large radiation sources. It will have capacity to separate approximately 200,000 curies per year of Cs^{137} as well as thousands of curies of Sr^{90} and other potentially useful fission products. The feed material for this plant will be waste solution remaining after processing reactor fuel assemblies for recovery of uranium and plutonium. The plant will not only permit immediate development of uses for the large quantities of activity produced, but will be a pilot plant for even larger-scale production of megacurie quantities of fission products if such industrial demand develops.

A summary of radiation sources from reactors would not be complete without noting the reactors themselves as sources as well as their coolants and fuel assemblies. Arrangements can be made to use the tremendous gamma-ray flux near a reactor core without serious neutron effects. Reactors with liquid-metal coolants will have large amounts of radiation available in the circulating coolant. Fuel assemblies of high-power reactors become intensely active and while being "cooled" before processing can be used as gamma-ray sources. Fuel elements removed from the Materials Testing Reactor in Idaho have been arranged to provide an irradiation test facility with gamma-ray intensities of about 10,000,000 roentgens per hour.

RAPID GROWTH IN UTILIZATION

The importance of isotopes is perhaps best indicated by the rapid and continued growth in utilization. Radioisotopes were first distributed from reactors in the USA in August 1946. Growth and extent of use during the now 9 years' distribution can be expressed in terms of users, shipments, curies, and published papers.

In the first year of radioisotope distribution only about 100 USA institutions received shipments. In 9 years this number has grown to over 2700. About 1100 are medical institutions and 1200 are industrial firms. Most of these institutions have several groups of users, hence the total number of radioisotope-using groups is around 5000.

Most striking is the recent growth in industrial users. In 1950 only about 100 firms were using radioisotopes. The number has increased 12-fold in just 5 years. This in itself demonstrates the importance of isotopes in technology and industry. Industrial firms would not invest in necessary equipment as well as buy the isotopes (sold at full recovery of costs) unless the values received were worthwhile. Extension of routine industrial uses of isotopes, such as radiography and radioisotope gaging, are taking place rapidly throughout industry. Also with new applications continually being developed, a continued rapid increase in industrial use is expected.

Radioisotope shipments from Oak Ridge National Laboratory alone have grown from only a few hundred the first year of distribution to over 12,000 per year. The number of shipments for 9 years totals almost 72,000. Although Oak Ridge National Laboratory still remains the primary routine supplier, each major USAEC reactor laboratory now produces and distributes some radioisotopes. Reactor laboratories supply only irradiated materials or simple compounds. Production of the very large number of special radioisotope compounds, radioactive pharmaceuticals, special radiation sources, and other preparations of radioisotopes, has been encouraged as a business for private enterprise. Shipments from these private secondary distributors now total more than twice that from the national laboratories. The total number of radioisotope shipments reaching ultimate users can be estimated at around 35,000 per year.

The total activity of radioisotopes distributed per year from all USA reactor sources has increased from around 65 curies in 1947 to over 40,000 curies in 1954. The average activity of individual shipments was originally around 30 mc. Today, excluding shipments of Co^{60}, the average is around 350 mc. This increase in activity per shipment is largely due to industrial uses. Also reactor laboratories now serve as bulk suppliers for secondary distributors. If one includes Co^{60}, the average shipment now exceeds 2.5 curies.

TABLE III. Approximate Number of Shipments and Curies of Principal Isotopes in Nine-Year Period (from Oak Ridge National Laboratory only)

Isotope	*Shipments*	*Curies*
Cobalt-60	1,000	64,300
Cesium-137	600	3,100
Iodine-131	27,600	3,000
Iridium-192	200	2,700
Gold-198	2,600	1,800
Polonium-210	120	1,200
Phosphorus-32	16,100	900
Hydrogen-3	300	700
Strontium-(89)-90	900	350
Carbon-14	2,100	40
Sodium-24	2,600	35
Others	18,800	800
Total	71,900	78,800

Table III shows the approximate number of shipments and curies shipped for principal isotopes distributed in 9 years from Oak Ridge National Laboratory alone. In total curies shipped Co^{60} far exceeds all other isotopes. This is mainly due to requirements for teletherapy units (now over 30 in number). Although Co^{60} has many uses in science and medicine, most of the 1000 shipments of this isotope have been for technological and industrial use. Isotopes such as Cs^{137}, Ir^{192}, Po^{210} and Sr^{90}, which are used mainly in industry, also show extensive use. I^{131} and P^{32} lead in number of shipments because of short life and extensive medical application.

In the future, most isotopes shown will increase steadily in use. Even greater increases can be expected in the total activity used of such isotopes as Co^{60}, Cs^{137}, H^3 and Sr^{90}. Industrial use of fission products from reactor fuel processing would far overshadow in activity the total curies distributed to date.

The distributed activity of C^{14}, 40 curies, does not seem large but in terms of normal cyclotron production, this would require about 200,000 years of cyclotron bombardment.

Another measure of usage is the number of papers published in scientific and technical journals. Publications resulting in 9 years through the use of USA-produced isotopes number around 11,000. The results of a survey of published work during 1951–1954 are shown in Table IV. The number of publications are

TABLE IV. Number of Published Works with USA-Produced Reactor Isotopes in Various Categories during 1951–1954

Diagnostic medicine	169
Therapeutic medicine	289
Clinical research	407
Human physiology	169
General medical research	196
Immunology	96
Metabolite physiology in animals	530
Non-metabolite physiology in animals	96
Injurious agent physiology in animals	55
General animal physiology	408
Animal husbandry	100
Bacteriology	225
Fertilizer uptake by plants	78
Plant physiology	126
Photosynthesis	58
Radiation effects on living organisms	269
Biochemistry	895
Biosynthesis of labeled compounds	345
Chemical synthesis of labeled compounds	146
General chemistry	159
Reaction mechanisms and kinetics	376
Radiochemistry	108
Radiation detection	154
Radiation physics	366
Nuclear properties of isotopes	466
General physics	52
General topics	191
Isotope techniques	308
Applied industrial use	231
Entomology	45
Total	7185

listed in 30 categories of major interest. Over 60% of these publications are in fields directly related to biology and medicine, including agriculture. The remainder fall mostly into the fields of science and technology. Only about 230 of the over 7000 publications are on applied industrial use. The reason publications in the latter category are small, in spite of the increasing number of users, is because many of the uses are routine, or modifications of uses already reported. Also many new industrial uses are not reported until covered by patents. The large amount of published work on basic studies and development of techniques is of course of much benefit to industry.

In addition to domestic distribution, the USA has made isotopes available to other countries since September 1947. In nearly 8 years of such distribution, almost 4000 shipments have been made to over 660 institutions in 46 countries. With the advent of reactors in other countries, closer to many users, most of the international needs for radioisotopes are met more conveniently outside the USA. Nevertheless distribution from the USA to other countries continues to meet special needs. Shipment by air permits users all over the world to obtain most useful isotopes from any reactor laboratory.

It should be emphasized that Great Britain and Canada also have extensive programs of isotope production and distribution. If the above statistics on uses, shipments and publications were accumulated for reactor laboratories of all countries, the figures would be most impressive. They will become even more impressive in the future as world-wide use of these reactor byproducts grows.

VERSATILITY OF PRINCIPLES OF UTILIZATION

The tremendous variety of uses of radioisotopes stems from three very versatile basic principles applicable to radiation: (1) radiation affects materials, (2) materials affect radiation, and (3) radiation traces materials.

The first of these three principles, the use of radiation to affect materials, is the least exploited at present but is expected to play a large role in the future. Although radiation can have many different effects on materials, these effects all result from ionization or excitation of atoms or molecules.

Thus we find the radiation from radioisotopes applied to produce ionization of air in static eliminators, excitation of phosphorescent light sources, destruction of bacteria in foods and drugs, activation of chemical reactions, polymerization of organics, alteration of material strength and conductivity, gene changes, and therapeutic results in patients.

In the second principle of use, radiation is directed at or through a material to gain information about the material. Here the requirements are a radioactive source, the material, and a detector to record the radiation transmitted through or reflected from the material.

All interactions of radiation with materials lower its energy or remove part of it from a beam. Thickness of a material, its density, atomic number and atomic structure, have different effects on different radiations. Therefore much can be learned regarding these properties by observing their effects in absorbing or reflecting radiations.

The major applications of this principle are industrial and clinical radiography, thickness and density gaging, and analysis by radiation penetration. Other important uses include liquid-level gaging and applications where an object is revealed by its effect on a beam of radiation. The principle is also applied to industrial sorting and packaging problems.

In applications based on the third principle, radiation traces material, the radioisotope is incorporated in or carried by the substance of interest. This substance can then be located or traced, or parts of it measured quantitatively.

Radiomaterial can be traced in bulk, as in locating markers, determining fluid flow, following motion of material, detecting leaks and measuring wear and abrasion. Also, radioisotopes as tracer atoms are used to study all kinds of chemical, physical-chemical and biochemical problems. In technology, tracer uses include studies of corrosion, diffusion, detergency, catalysis, and kinetics and mechanisms of reactions.

It is obvious that each of these principles has much versatility in application. They provide a wide spectrum of uses, covering many fields of technology as well as varieties of industrial materials, processes and products.

SPECIFIC ILLUSTRATIONS OF USEFULNESS

A few examples of applications in the three categories mentioned will illustrate their usefulness.

Luminescence, an effect of radiation on special phosphor materials, provides self-luminous material for safety marking in aircraft, mines, and public buildings, for low-level illumination, and for standard light sources. Isotopes used for the purpose include polonium, tritium, strontium-90, and cesium-137. Brightness levels up to 1000 microlamberts are obtained, and a wide choice of colors can be produced. Beta-emitters are preferred for this because there is much less radiation damage to the phosphor than by alpha-emitters.

Voltage production is another application of radiation effects. This extremely small but direct conversion of radioactivity to electricity is attracting much attention. Several types of radioisotope batteries have been developed, operating on different principles. The direct-charging type builds up a potential as negative beta particles from a radioisotope leave one electrode and collect on another. The second and third types depend on induction of a current by the contact potential difference between dissimilar materials, in one case when ions are produced in a gas by the radiation and in the other when a semi-conductor junction in silicon is energized by the radiation. In still another, the heat from radioactive decay is converted to electricity by thermocouples; in another, the light from a radioisotope-activated phosphor is converted by a photovoltaic cell. Such batteries produce minute amounts of current but are very useful, for example, in electronic circuits where long-lived, stable sources of potential are required.

Polymerization of monomers to form plastics is a radiation effect resulting from the breaking of molecular bonds. Many open bonds are formed in the path of each ionizing particle and result in active recombination into new forms and structures. A polymer produced by radiation can be quite different from a product polymerized chemically from the same material. Since no chemical is introduced as a catalyst, the product is relatively free from impurities which can alter its properties. Not only industrial products, such as plastics and synthetic rubbers, but blood-plasma extenders also, can be polymerized to advantage by radiation.

A further effect of radiation on polymers is the initiation of cross-linking between neighboring molecules. Side-chain bonds are rather easily broken by radiation and immediately join with similarly opened bonds in adjacent molecules. A quite rigid, three-dimensional structure is formed, having different strength, heat resistance, and other properties from the original.

While radiation from radioisotopes may not compete with present commercial methods of producing many polymers, its ability to polymerize and cross-link gives some special materials not possible or practical by other means.

Sterilization of foods and drugs is another application of radiation effects. Although at present largely in the research stage, it will undoubtedly call for great quantities of radioisotopes in the future.

Radiation in large enough doses can destroy harmful bacteria and enzymes in a material without significantly raising its temperature. Although the necessary radiation doses cause unwanted changes in taste and color in certain foods, others are quite unchanged and offer great commercial possibilities. Among the potential applications may be mentioned preservation of meat, poultry and fish, killing of trichina in pork and of insects in grain, inhibiting of sprouting and spoilage in stored onions and potatoes, extending shelf life of canned meats and vegetables, and pasteurization of dairy products.

While a good estimate of sterilization cost cannot yet be made, it seems that it will be in the neighborhood of one to seven cents per pound. Future costs depend in large measure on feasibility and economics of obtaining large usable quantities of fission-product isotopes from spent reactor fuels.

Sterilization of drugs and other medical supplies seems to present fewer problems than that of food. Side effects are not as important and radiation processing can assure "guaranteed" sterilization while avoiding disadvantages of high-pressure steam sterilization. Judging from effectiveness of experimental facilities, operating costs would compare favorably with present ones.

The ability of radiation to supply information by reflection from or transmission through material has proved a boon to industry. Most of the routine industrial applications are based on this principle of material affecting radiation.

Radiography, for example, took a great stride forward when strong sources of gamma-ray emitting isotopes such as cobalt-60 and iridium-192 became available. Radiographic testing of numerous materials and products became more convenient and economical. Many small foundries now maintain a routine radiographic inspection of their castings. Inspection teams can move along a new pipeline in the field, radiographing weld after weld in rapid succession.

Radioisotopes are supplying another long-felt need in industry with their ability to give rapid information as to thickness and density of material. The beta-gage, employing beta particles, is the most commonly known radioactive device for this purpose. Beta-gages are now completely established as a routine thickness monitoring instrument, especially in sheet processing industries with products such as steel, aluminum, and other metals, plastics, paper, cloth, and floor covering. The rate of installation of these gages is increasing rapidly.

Thickness gages are not limited to the use of beta-emitters, appropriate gamma and X-ray emitters being employed in gaging larger thicknesses of sheet materials. For example, a gamma-ray density gage has been developed to record continuously the density, and thus the weight, of sugar during a refinery process. A product or process material can be in containers or flowing in closed conduits and still the gamma-ray density gage can "see" it through the walls.

The great variety of materials which industry needs to measure is matched by the variety of radiations among the available radioisotopes. Near one extreme is the use of 67-kev beta radiation from nickel-63 to measure ink-film thicknesses on a lithographic press and, near the other, is the use of 1.3-Mev gamma rays from cobalt-60 to gage sheet steel.

Reflection type gages have been developed which take advantage of the backward scattering of radiation. They require only one side of a material to be accessible, radioisotope source and detector being mounted side by side. Using gamma sources, such instruments allow routine monitoring of internal corrosion in process tanks, logging the types of strata in an oil-well bore, and measurement of soil density. Beta-ray reflection gages permit measuring of thin films on base material of different atomic weight, such as gold plated on copper or plastic over steel. Wear tests on traffic-marking paint used on roads, for example, have been made with such a gage which measures the paint film to within a ten-thousandth of an inch.

A great advantage of radioisotope gages, in addition to their convenience and accuracy, lies in their ability to measure a material without contact. Swiftly moving strips of delicate and perhaps still moist or sticky products can be measured, where formerly production machinery had to be stopped while the strip was measured. In the manufacture of coated abrasives, for example, one company has long been using a series of five beta-gages in one production process. These measure the thickness and amount of backing stock, adhesive layers, and abrasives as they are applied to the product passing swiftly through various stages. The resulting product is superior in uniformity and much saving in raw material is realized.

The use of these gages leads naturally to automatic control of the processing equipment. Such automatic control is now fairly well established in the processing of adhesive tape, steel strip, cigarettes, rubber sheet, and paper and in plastic calendering. Such diversity of application is a good illustration of widespread usefulness.

The third category of radioisotope use in industry, radioactive tracer techniques, does not employ as many total curies of activity as will the production of radiation effects, and are not as numerous as the penetration and reflection techniques. They nevertheless include many of the most interesting applications. Moreover, they can match the others in usefulness and economic value.

Many tracer techniques in industry have reached the routine operational state, usually where an object or material is to be followed or located or to be identified among others of its kind. For example, radioactive material is injected between successive shipments of petroleum products through a cross-country pipeline and identifies the change-over by activating a continuous counter at a later switching terminal. Losses due to uncertainty as to region of mixing have been sharply reduced.

The petroleum industry is an outstanding example in its solving of technological problems with tracer isotopes. Fluid flow is of prime importance from the initial drilling of a well to final delivery of refinery products. Pumping a suspension of radioactive particles into a well and then logging the radioactivity along its length reveals leaks, open formations, and permeable zones. The latter zones are of interest as oil-bearing or oil-thieving areas or as pathways for conduction of water in pressuring an oilfield for secondary recovery.

Radioactive tracers are added to cement, used to block off unwanted zones, to evaluate its position and extent, and are added to pressurizing water to determine its entry into other wells. They are also added to the acid used to treat oil-bearing limestone strata for greater productivity, to tell by means of a counter lowered into the well when the acid is being forced into the proper zone.

The moving-marker technique for cross-country pipeline shipments is applied also in tracing movement of batches of oil through complex piping patterns in the refinery. Also, the rate of flow of solid catalyst particles in petroleum cracking plants is monitored routinely by timing the passage of an occasional radioactive bead.

Underground gas-storage fields utilize a porous formation many hundreds of feet down, sealed above and below by impervious strata and reached by drilled wells. When a leak occurred recently, short-lived radioactive argon-41, emitting 1.3 Mev gamma rays, was introduced through a well into the reservoir. Gas leakage, especially around loose-fitting well casings, could then be easily traced with detectors lowered through the casings.

This is but one example of the power of radioisotopes to detect leaks of all kinds which are a constant source of trouble in industry. A tracer amount of radioisotope can be introduced into a gas or liquid system and then either detected as it emerges from a leak or followed, despite intervening structure, by means of its gamma radiation.

One of the most widely applicable and rewarding uses of tracer radioisotopes is in wear and corrosion studies. Radioactivity introduced into a machine part, cutting tool, or furnace lining is also present in the particles later worn or corroded from the surface. Thus the amount or rate of removal is quickly and accurately measured by the activity appearing in the lubricating oil or furnace product.

In another type of wear test a material applied to a surface is made radioactive. After wear, the amount of radiation shows how much material remains, and thus its resistance to wear. Many protective coatings are evaluated in this way.

Benefits are derived, however, by almost any industry adapting radiotracers to the solution of its problems. As an example in the pharmaceutical industry, rapid analysis for the vitamin B-12 content of large heterogeneous mixtures is performed by adding a small radioactive-labeled sample of the vitamin and using the isotope dilution method. As an example from a widely different technology, the organic chemicals used in water flotation of mineral ores to cause a desired mineral to separate out are evaluated by tracer techniques. With labeled chemicals it is possible to determine the conditions for and extent of adsorption on any particular mineral.

As tracers of molecules, rather than of material in bulk, radioisotopes are widely used in the study and control of chemical processes. Citing the petroleum industry again as an example, a great amount of research is being done on hydrocarbon chemistry. Carbon-14, deuterium, and tritium come naturally as tracers into this work. They have been of inestimable value in elucidating the mechanisms involved in alkylation, polymerization, catalytic cracking, and many other important reactions.

Further development of the versatile industrial uses of isotopes will be limited only by human ingenuity.

PRESENT AND POTENTIAL VALUE OF USES

The total economic value of isotope utilization is very great but difficult to assess exactly. It can, however, be measured by success in (1) saving time of personnel in basic and technological research, (2) increased speed in acquisition of knowledge, (3) saving materials and labor in manufacturing, (4) improving performance and durability of manufactured products, (5) increasing agricultural productivity, (6) decreasing losses from food spoilage, and (7) improving the health of the public.

Although it is not possible to assign quantitative or monetary values to all these benefits, certain examples and generalizations will help give an over-all appraisal.

Because the supply and time of scientific and technical personnel are limited, saving of their time is more important than the salaries involved. But even more important is earlier application of results. The sooner problems are solved, the sooner benefits can be realized.

A good example is the great saving in time of personnel in studies of all kinds of wear and corrosion. Older methods of observation were limited to pronounced changes and were slow. With tracer techniques, very small amounts of wear or corrosion are measured quickly and accurately. The rate of obtaining results is speeded up by a factor of 10 or more. One group studying improvement of lubrication, which involves counteracting both wear and corrosion, obtained results with tracer techniques in 4 years at a cost of $30,000 which would have required 60 years by previous methods and cost $1,000,000.

The major point of this illustration is not the saving in cost of the research, although this is considerable, but the fact that the results were obtained so many years sooner. This research developed a lubricant that reduced wear and corrosion to one-third of previous values. Ultimate savings in wear from introducing the improved lubricant many years earlier will far exceed the savings in cost of research.

Radioisotope techniques have already been applied to a host of wear studies such as wear of automotive and diesel engine parts, electrical contacts, cutting tools, furnace linings, and rubber tires. Corrosion studies using isotopes include testing the resistance of materials to corrosive agents and the effectiveness of various protective coatings such as metal plating, plastics and paints. Since wear and corrosion of all kinds cost the world many billions of dollars per year, the increased rate of finding ways to reduce this loss can result in enormous savings.

Increased speed in gaining new knowledge depends not only on saving time of research personnel but also in using new powerful research tools like radioisotope tracers which can obtain results more directly than older methods. Much of the vast amount of work accomplished with radioisotopes in all fields has built up a body of knowledge much of which would not have been found with other means. This new knowledge immediately helps gain further knowledge and also is soon reflected in practical uses.

Hundreds of radioisotope applications in industry are directed at economy in manufacturing and improving product quality. Radiography alone is used by around 400 USA firms. Radiographic inspection not only prevents damage or waste from product failure but permits economy in materials of construction. Inspection of welds on a large tank or a long pipe line may save many thousands of dollars. Savings from all radiographic uses may amount to millions of dollars per year.

Radioisotope thickness gages are used in hundreds of plants to control production of all kinds of sheet material. Failure to keep thicknesses within tolerance can be very costly in requiring reworking or rejecting a batch of material. Economy in material is also effected by avoiding unnecessary thicknesses. Manufacturers often report savings of many thousands of dollars per month through the use of these gages. Here, again, the total yearly savings for all uses amounts to millions of dollars.

The previously mentioned radioisotope techniques in the oil field can help keep wells, costing a million dollars or more, producing longer and more abundantly. The single idea of timing catalyst flow with radioactive particles saved one oil refinery $100,000 by preventing the shutdown of a huge cracking plant. These and other such economies effected in the petroleum industry total still more millions of dollars per year.

Space does not permit illustrations of the economic value of isotopes in other applications and industries.

but many examples can be found in the development and production of chemicals, plastics, rubber, metals, textiles, detergents, and fabricated articles. In USA industry alone the total savings from all applications of radioisotopes have been estimated at 100 million dollars annually, with the possibility that it may reach 10 times this value in less than 10 years.

These monetary savings, although estimated for the industrialist, can represent even greater economic benefits at the consumer level. Not only may a product be obtained at a lower cost, but, if it performs better or lasts longer, it saves the user further time and money.

Agriculture, comprising a major part of the world's economy, has realized many benefits from radioisotopes. Modern agriculture is in many ways a large-volume production industry. Serving agricultural needs are large industries providing fertilizers, insecticides, herbicides, farm machinery, etc. Radioisotopes are being used with success in solving technological problems in these related industries.

Also related to agriculture are the industries of food handling, processing, and storing. Although radioisotopes have not yet played a major role in this area, extensive studies have shown that radiation can improve the storage qualities of foods without significant change in nutritional or other values. Many believe that this research will lead to new preparation and processing methods as well as distribution and marketing practices. If the potentialities of radiation pasteurization and processing are fulfilled, this use of reactor byproducts alone could result in an annual saving of many millions of dollars.

In agriculture itself, hundreds of radioisotope tracer uses are being made in important studies of plants and animals. Many studies are conducted in the field with a wide variety of crops and are concerned with the uptake and utilization of fertilizers and mineral nutrients, both by roots and foliage. Numerous tracer studies are also being made on the action of herbicides, insecticides, and fungicides, on plant diseases and on the basic process of photosynthesis. In view of the hundreds of millions of dollars in fertilizers used annually, tracer studies can provide very great savings by finding more efficient methods of supplying nutrients to crops. Also, because of tremendous annual losses to weeds and insects, tracer studies to find ways of reducing this loss can result in great economic gain.

In the field of medicine, and the related pharmaceutical industry, radioisotopes are also making important contributions. Perhaps most important is the great body of knowledge being gained of the biochemistry and physiology of humans and animals, in both health and disease. Much valuable information is also being gained about the metabolism and mode of action of a large number of drugs. In addition to studies of drug action, the pharmaceutical industry is using isotope techniques to improve analytical procedures and to study bacteria and viruses and ways to combat them.

A large number of practical applications are being made of radioisotopes in medical diagnosis and treatment. As noted, over 1100 medical institutions in the USA are using these materials routinely for diagnosis and treatment. The number of patients who have benefited from radioisotopes as a diagnostic tool is now around 500,000 and about 50,000 have received radioisotope therapy. These medical uses have grown rapidly and will continue to grow. Radioisotopes are thus already important in public health, but new knowledge being gained will contribute much to further improvements in world health.

SIGNIFICANCE IN OVER-ALL ATOMIC ENERGY DEVELOPMENT

Radioisotopes constitute the most immediately realizable peaceful benefits for a nation undertaking a program of atomic energy development. Their use is also doubly rewarding. In addition to having many direct benefits, radioisotope utilization can have another very important result, the development of a large resource of professional and technical persons familiar with radiations and radioactivity. Since radiation and radioactivity are uniquely important occupational factors of the atomic energy industry, an increasing number of persons trained with radioisotopes both precedes and permits an expanding atomic program.

The fostering of radioisotope uses in all fields will establish "seeds" from which interest and programs in atomic energy will grow. This will also create uses for the increasingly abundant amounts of radiation and radioisotopes which many nations will soon have as byproducts of research and power reactors.

Although world-wide need for propulsive and industrial power is the main interest of most countries in developing atomic energy programs, the byproduct radioisotopes should not be overlooked as of equal importance in the long run to the over-all health and welfare of nations.

Recent Developments of Radioisotopes Used in Industry

By Henry Seligman,* UK

The use of radioisotopes in industry is already well established, only the more recent developments and technical improvements of isotope applications will be discussed.

There are two quite distinct categories of applications, those where the isotopes are built into certain types of equipment, usually for the purpose of process control or inspection, and the other group where ingenious special applications have been evolved for certain definite purposes. It would be beyond the scope of this paper to try to cover the whole applications in this field, therefore only certain recent developments will be mentioned.

Non-contact thickness gauges using beta and gamma emitters for absorption or scattering measurements are already widely used. A number of new developments have extended the field for which these instruments can be employed. Until now it was for example, impossible to measure relatively small areas with the help of such equipment. Therefore where the thickness of materials of curved surfaces have to be determined an improved setup had to be created. Such an experimental instrument has been built and is using a collimated tube of small diameter, filled with the gas krypton-85 as source, which is surrounded by a scintillation counter in ring form. With this instrument an area of only ¾ cm^2 can be inspected.

Another new development is the improvement of gamma scattering gauges for the purpose of detecting corrosion in pipes from the outside. The improvement consists of counting only the scattered and therefore much less energetic gamma rays which are sorted from the original gammas by using a one channel energy selector. Therefore it is not necessary to shield the source and by having source and counter as a very small unit near the pipe to be probed a much better definition can be achieved.

This development promises to be of the greatest significance in measuring corrosion of pipes as the periodical shut-down of a plant for such an inspection is now unnecessary. It is too early to assess the commercial value of this application but an increase of several per cent of the total production of such a plant seems a probability.

Great Britain was one of the countries which saw an early industrialization and therefore we in England have the oldest pipes in our ground. No wonder therefore, that we are specialists in leak detection and a great part of our work has been concentrated on this application.

We have three methods of leak detection, all of them using the same isotope, sodium-24 in the form of $NaHCO_3$. The tolerance level being 8 μc per litre is relatively high so that it is a good isotope to use for this purpose. For one method, one dissolves sodium carbonate in water and bleeds the active solution into a pipeline. The pipe is then cleaned out and the radioactive residues surrounding the pipeline at the leaking places can be measured. However, probing with counters at many places is necessary which makes this a laborious method. The second method measures the rate of flow of the radioactive solution and one can so find the section in which the leak occurs. The third method which has been developed at Harwell is by far the simplest one and mainly used to check oil pipelines. The pipeline which is under pressure with radioactive solution is first followed by a washing solution and then by a "go devil" which carries a halogen quenched Geiger counter, connected to a small wire recorder. To check the time when the "go devil" passes certain places along the pipeline, small cobalt sources are put on the outside as markers. By playing back the pocket recorder into a recording ratemeter the leaks can be detected. Pipelines of up to 20 km length have been investigated already in this way, and this method is easy, time saving and can obviously be extended for investigation of much longer lines.

Many problems in industry depend on solving the simple problem of interface detection. Nowadays interface detection is a common problem in pipelines and one of the new features here is to find more suitable isotopes and techniques for that purpose. The use of scandium-46 owing to its reasonable half-life and strong gamma emission is the most promising one, and a patent has been taken on a safe single shot injection method. Another approach which has been successful is founded on the measurement of changing density by absorption methods. Another method is based on measuring the moderation and scattering of fast neutrons which is almost entirely a function of the H content of the fuel.

Important progress has been made during the last year by developing a method with which it is possible to distinguish between different layers or liquids like oil and brine through steel and concrete, a tricky problem which presents itself in bore holes. This has

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been solved ingeniously by measuring the prompt gammas emitted on absorption of slow neutrons. The energies of the prompt gamma rays are characteristic of the neutron absorbing nucleus. We used a polonium-beryllium neutron source and a gamma scintillation counter, in connection with a one channel kicksorter. Field experiments have shown already that one can get a clear distinction between oil and brine through a steel pipe with a wall thickness of 1 cm which is set in concrete even when the borehole is filled with oil. With this method we will be able to detect a rise of the brine level which often puts a bore hole out of commission. By using this simple device bore holes will be saved from being fouled and the financial reward by employing these instruments may well prove bigger than the construction of several atomic reactors.

Low energy gamma emitters are absorbed mainly by the photo-electric process. This fact has been used for an experiment which was done in cooperation with the National Coal Board in England where we sorted coal from shale. The same method might be used for the important preconcentration of ores. We used thulium-170 as a source which has a gamma emission of 85 kev. Coal scatters more of these gammas than shale. The alternative method for the separation of coal and shale makes use of the different density of the substances. To build such a separation plant costs of the order of ½-million pounds. Apart from this some coal is wasted in the following washing process. This is another of the many cases where isotopes help to make a better product by saving money and labour at the same time.

Isotopes have been used for checking the efficiency of fine filters with radioactive particles. Bacteria fed on phosphorus, as well as radioactive gold precipitated on plastic-balls have been used successfully for this purpose.

Gamma radiography is today a well established inspection process. The cheapness and practicability allows the smallest firm to use this method for process control. It is astonishing to see how many smaller firms which had no non-destructive testing equipment are now making use of cobalt-60, iridium-192, caesium-137 and thulium-170 as sources for radiography. There is no doubt that during the last year or two the percentage of metal parts leaving factories and work shops free of faults has gone up considerably due to the use of radioisotopes for non-destructive testing.

Beta-emitting static eliminators help in Britain to speed up production processes. As the static charge is removed the fire risk is considerably lowered and the machines can run faster. In some cases figures of 20–50% increase in production has been claimed after static eliminators have been installed.

The special applications cover an enormous field and only a few can be discussed here. In the metal industry it is a problem to measure the solidification boundary of continuous cast metal. This has been measured in the case of aluminium by two methods. Firstly, by adding a small amount of radioactive gold to the casting, the solidification boundary could be found by investigating the bars with auto-radiographic methods. A second method which gave the same good result was to add traces of non-active gold and make activation analysis of the bar later.

After last year's Comet crash one avenue to be explored was to find out if the disaster may have come about by the spillage of fuel. Kerosene which is used as fuel was labelled in one case with palladium-109-acetyl-acetonate and in another with palladium-109-diethyl-dithiocarbamate. Flight experiments were conducted in order to find if spillage occurred and if so whether the kerosene got near parts of the plane where an explosion could occur. These experiments were successful in so far as they excluded this possibility from the list of possible reasons for the tragedy.

A special investigation of general interest is the marking of Thames mud in order to follow its movements. Experiments made in conjunction with the Hydraulic Research Station and the Port of London Authority were started last summer and are continuing right now. The method used is to grind scandium glass to the right particle size. After that it is irradiated and mixed with a small amount of mud, which has been scooped up from the bottom of the Thames. It is then released on the bottom of the Thames and the movement followed up by underwater Geiger counters. At last year's experiments the radioactive mud could be followed for two weeks without difficulty and it is hoped to solve some of the siltation problems of the Thames Estuary with this method. As siltation problems in general are very difficult problems to solve, this method may prove to be a universal one, to be useful for any harbour. The reorganisation of the dredging procedure depends on the outcome of these experiments.

Leaving apart the increasingly important fission product applications it becomes clearer everyday that the applications of radioisotopes do not constitute merely a useful contribution to industry but are becoming of the utmost economic importance. Radioisotopes play already an important part in the nation's economy; in the long run they may turn out to be one of the most important assets which man can derive from this atomic age.

Production and Use of Radioisotopes in Japan

By K. Suzue,* Japan

The production of radioisotopes in Japan is now limited to that by the cyclotron on a small scale. The cyclotron now in operation in Japan is at the Scientific Research Institute, Ltd., Tokyo, and is producing mainly short-lived radioisotopes, such as sodium-24, potassium-42, and copper-64, which it is impossible for us to import from abroad in useful form. The details of the cyclotrons in Japan will be found in Table 1.

Therefore, almost all radioisotopes used in Japan were those imported from the USA, England, or Canada. The affairs related to radioisotopes in the Japanese Government are handled by the Scientific and Technical Administration Committee (STAC) of the Prime Minister's Office. The STAC examines the applications for use of radioisotopes and authorizes eligible users to import isotopes into the country. The importation of radioisotopes started in 1950 firstly from the USA. At the beginning of the importation they were used for basic researches by only fifty-nine groups in fifteen institutions. Afterwards, with the spread of the knowledge regarding the availabilities of radioisotopes and with the increase of persons trained to handle and use them safely, the number of users has gradually increased. Thus, at the end of last year, 431 groups in 100 institutions were found to be adopting isotopes for scientific research, medical application and industry.

The millicurie amounts of radioisotopes imported into this country during the time from the beginning of the importation to the end of March, 1955 are shown in Table 2. The number of institutions in each ministry which uses radioisotopes is in Table 3, and the classification of the number of users and cases of use according to the field of use is in Table 4.

TECHNOLOGY AND INDUSTRY

Of the above-mentioned groups, approximately 7 per cent falls under the field of industrial research and industry. As tracers, industrial utilization of radioisotopes has been to date confined to laboratory-scale investigations, but active researches have been extensively carried out in different laboratories. Tracers have been used in metallurgy, for example, in studying the diffusion of one solid metal into another and the self-diffusion between two samples of the same metal. On the problems of steel manufacturing, tracer atoms have been also adopted for the study of carburization and desulfurization. Industrial researches on the use of radioisotopes as tracers are found in the field of ceramics, especially in the study of glass structure and fire-brick formation. The use of radioisotopes has been extended to the study of friction, for example, in the case of engine wear and lubricating oil.

Industry in Japan has already adopted in several ways the radiation from radioisotopes on a plant-scale. In the first place, the nondestructive test of welds and castings by radiocobalt, radiocesium and so on is in routine work in the leading ship-building and boiler-making factories all over the country. Radioactive thickness gauges and liquid level gauges are also in practical use in automatic and quality control in the process of production in several companies. Two companies are producing T.R. tubes to be used for radar by applying radiocobalt to their electrodes to eliminate the delay and disorder of electric discharge. In addition, the indication of locations of oil

TABLE 1. List of Charged Particle Accelerators in Japan

Location	*Magnet weight (tons)*	*Pole piece diameter (cm)*	*Particle, energy (Mev)*	*Status*
Scientific Research Institute, Ltd. (Tokyo)	24	66	d—4	In operation
Faculty of Science, Osaka University (Osaka)	53	111.8	d—12	Completed
Institute for Chemical Research, Kyoto University (Kyoto)	80	105	d—16	Under reconstruction
Institute of Science and Technology, Tokyo University (Tokyo)	7	40	d—2 p—4	Under construction
Nuclear Research Institute, Tokyo University (Tokyo) (Synchro-cyclotron)	300	160	d—21 p—70	Under construction

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* Chief, Office of the Scientific and Technical Administration Committee, Prime Minister's Office.

TABLE 2. Amounts of Radioisotopes Imported, in Millicurie Contents Ordered. (Contents in Parentheses Are Not Yet Imported.)

Isotopes	*Chemical form or specific activity*	*1950*		*1951*	*1952*		*1953*		*1954*	
		Former term	*Latter term*	*Whole year*	*Former term*	*Latter term*	*Former term*	*Latter term*	*Former term*	*Latter term*
				1. Separated isotopes						
Hydrogen-3										(20 mc)*
Carbon-14		8 mc*	3 mc*	23.5 mc*	32.5 mc*	17 mc*	28.5 mc* 4 mc‡	20 mc*	25.5 mc†	69.2 mc†
Sodium-22				2 mc*						(4mc)†
Phosphorus-32		72 mc*	177 mc*	785 mc*	1806 mc*	1652 mc*	2412 mc*	3328 mc*	4810 mc*	2869.5 mc† (2449.5 mc)
Sulfur 35 (Sulfate in weak HCl)		1 mc*	8 mc*	40 mc*	158 mc*	81 mc*	97 mc* 1 mc‡	119 mc*	215 mc*	99.5 mc† (96.5 mc)
Sulfur-35 (Barium sulfide in Ba(OH)$_2$ solution)		4 mc*	2 mc*	35 mc*	19 mc*	5 mc*	14 mc*	38 mc*	21 mc*	14 mc† (17 mc)
Sulfur-35 (Elemental sulfur in benzene solution)				1 mc*	5 mc*	9 mc*	9 mc*	24 mc*	5 mc*	48 mc†
Chlorine-36		36 μc*	6 μc*	33 μc*	79 μc*	52 μc*	39 μc*	54.5 μc*	57.5 μc†	164 μc†
Calcium-45	0.2–0.4 mc/gm	6 mc*	4 mc*	58.5 mc*	208.5 mc*	126 mc*	188 mc*	134 mc*	138 mc*	(173 mc)*
Calcium-45	5–60 mc/gm				36 mc*	22 mc*	37 mc*	49 mc*	80 mc*	(139 mc)*
Calcium-45	1000 mc/gm			1 mc*					7.2 mc*	(2 mc)*
Scandium-46				5 mc*				2 mc*		(2 mc)*
Chromium-51						5 mc*	2 mc*	5 mc*		
Iron-55				10 mc*	11 mc*	13.5 mc*	21.5 mc*	12 mc*	20 mc*	(21.5 mc)*
Iron-59		30 mc*	5 mc*	10 mc*	14 mc*	8 mc*	20 mc*	21 mc*	24.2 mc*	(6.5 mc)*
Iron-55, 59										4 mc*
Cobalt-60		120 mc*	19 mc*	292 mc*	185 mc*	205 mc*	67 mc*	154 mc*	221 mc*	(122 mc)*
Nickel-63					0.5 mc*	3.5 mc*	0.5 mc*			
Zinc-65			3 mc*	34 mc*	104 mc*	14 mc*	22 mc*	17 mc*	358 mc*	(126 mc)*
Germanium-71										(1 mc)†
Arsenic-73										(5 mc)*
Selenium-75										(1 mc)*

TABLE 2. Amounts of Radioisotopes Imported, in Millicurie Contents Ordered. (Contents in Parentheses Are Not Yet Imported.) *(Continued)*

Isotopes	*Chemical form or specific activity*	*1950*		*1951*	*1952*		*1953*		*1954*	
		Former term	*Latter term*	*Whole year*	*Former term*	*Latter term*	*Former term*	*Latter term*	*Former term*	*Latter term*
Strontium-89				7 mc*	9 mc*	42 mc*	42 mc*	34 mc*	71 mc*	(133 mc)*
Strontium-90					18 mc*	130 mc*	93 mc*	236 mc*	253 mc*	(822 mc)*
Yttrium-91										(91 mc)†
Zirconium-95								20 mc*		10 mc† (7)
Ruthenium-106								1 mc*		(3 mc)*
Silver-110								13 mc*	17 mc*	(16 mc)*
Tin-113										(1 mc)*
Indium-114									4 mc*	
Cadmium-115					1 mc*	5 mc*	5 mc*	8 mc*		
Antimony-124									6 mc*	6 mc† (1)
Iodine-131				236 mc*	585 mc*	671 mc*	1249 mc*	1452 mc*	3029 mc*	1744 mc§ (1477 mc)
Cesium-134					1 mc*	23 mc*		4 mc*		(58 mc)*
Cesium-137					16 mc*	105 mc*	52 mc*	131 mc*	290 mc†	29 mc† (10 mc)
Barium-140						6 mc*		6 mc*		2 mc*
Cerium-141					2 mc*				20 mc*	13.5 mc* (14.5 mc)
Cerium-144								16 mc*	10 mc†	19 mc†
Promethium-147						0.5 mc*				
Thulium-170										(220 mc)*
Tantalium-182										(5 mc)*
Tungsten-185							2 mc*	6 mc*	3 mc*	(4 mc)*
Iridium-192										(20 mc)*
Mercury-203				2 mc*	6 mc*		1 mc*	6 mc*	3 mc*	(8.1 mc)*
Thallium-204						5 mc*	2 mc*	7 mc*	3 mc*	(7 mc)*

TABLE 2. Amounts of Radioisotopes Imported, in Millicurie Contents Ordered. (Contents in Parentheses Are Not Yet Imported.) *(Continued)*

Isotopes	*1951*	*1952*		*1953*		*1954*	
	Whole year	*Former term*	*Latter term*	*Former term*	*Latter term*	*Former term*	*Latter term*
2. Irradiated units							
Phosphorus-32					1 unit*		
Sulfur-35					1 unit*		
Chlorine-36		1 unit*				1 unit*	
Calcium-45					1 unit*		
Chromium-51							1 unit†
Cobalt-60		1 unit*					
Nickel-63						1 unit*	
Zinc-65							1 unit†
Arsenic-77	1 unit*						
Zirconium-95							1 unit†
Technetium-99							1 unit†
Silver-110			1 unit*				
Indium-114							1 unit†
Antimony-125		1 unit*					
Cesium-134						1 unit*	
Europium-153							
Tantalum-182							1 unit†
3. Isotope-labeled compounds							
C^{14} labeled barium carbonate							
C^{14} labeled benzoic acid		2 mc*					
C^{14} labeled benzene		0.5 mc*					
C^{14} labeled dl-alanine	0.1 mc*						0.5 mc†
C^{14} labeled dl-phenylalanine			0.5 mc*				
C^{14} labeled dl-tyrosine							0.2 mc†
C^{14} labeled ethanol					0.15 mc†		
C^{14} labeled D-glucose		1 mc*	2 mc†		0.1 mc†		
C^{14} labeled glycerol-1			0.1 mc†		0.1 mc†	0.3 mc†	0.1 mc†
C^{14} labeled glycine-1			0.1 mc†		0.1 mc†		
C^{14} labeled glycine-2			1 mc†	0.1 mc*	0.3 mc†		1 mc†
C^{14} labeled methyl iodide							0.1 mc†
C^{14} labeled phenyl iodide							0.1 mc†
C^{14} labeled pyruvamide					0.15 mc†		
C^{14} labeled sodium acetate-1					0.1 mc†		0.1 mc†
C^{14} labeled sodium acetate-2							0.5 mc†
C^{14} labeled sodium cyanide						0.5 mc†	
C^{14} labeled sucrose							4 mc*
C^{14} labeled stearic acid-1						0.2 mc†	
C^{14} labeled stearic acid-2						0.1 mc†	0.1 mc†
C^{14} labeled urea				0.5 mc*			1 mc†
S^{35} labeled ferrous sulfide							10 mc†
S^{35} labeled methionine			1 mc†				
S^{35} labeled thiourea			10 mc†			1.5 mc†	
I^{131} labeled diiodo-fluorescein			10 mc†				
4. Others							
Cobalt-60 small source¶	1700 mc† 1342* mc‡	3926 mc†	800 mc* 1700 mc† 690 mc‡	2033 mc* 300 mc† 500 mc‡	3857 mc* 560 mc‡ 450 mc†	2000 mc* 1890 mc† 500 mc‡	500 mc* 800 mc† (2046 mc)
Cobalt-60 multicurie source	170 c*			200 c*	243 c*		
Strontium-90 (plaque)						20 mc†	
Cesium-137 (metallic source)						250 mc†	1 C†
Iridium-192 (metallic source)			6C†			8 C†	5 C†
P^{32} sterile solution	2 mc*						
I^{131} sterile solution	1 mc*						
Fission products						50 mc*	
Tritium-zirconium target						5 pieces*	4 pieces†
5. Reference sources							
C^{14} polystyrene sheet	1 sheet*	1 sheet*		1 sheet*	4 sheets*	1 sheet*	1 sheet* 5 sheets†
Carbon-14							2 sets* 2 sets†
Phosphorus-32					1 set*		
Cobalt-60				2 amp.*	1 set*		2 sets*
Iodine-131					1 set*		
Beta-ray simulated source							3 sets†

* Imported from USA.
† Imported from England.
‡ Imported from Canada.
§ Imported from the Netherlands.
¶ In 1951, 90 mc was imported from USA.

TABLE 3. List of Numbers of Institutions Using Radioisotopes

Ministries concerned		*1950*		*1951*	*1952*		*1953*		*1954*		*Total of actual number of institutions*
		Former term	*Latter term*	*Whole year*	*Former term*	*Latter term*	*Former term*	*Latter term*	*Former term*	*Latter term*	
Ministry of Education	National	8	10	17	22	21	22	26	25	27	32
	Public	2	1	6	5	8	10	12	14	11	14
	Private	1	2	7	9	8	9	11	11	13	20
Ministry of Welfare	National	2	4	4	6	8	3	5	5	10	12
	Public								1		1
	Private						1	2	8	4	15
Ministry of Agriculture & Forestry	National		1	2	2	2	3	5	3	7	9
	Public	1		1	2	1					2
	Private			1		1					1
Ministry of International Trade & Industry	National		1	1	2	1	2	1	4	4	5
	Public						2		2		3
	Private	1	2	4	6	12	2	12	8	13	33
Ministry of Transportation	National					1	1	1	3	3	6
	Public										
	Private										
Ministry of Postal Service	National				2	2	4	4	5	5	5
	Public										
	Private										
Ministry of Construction	National			1	1		1		1	1	2
	Public										
	Private										
Defence Agency	National									2	2
	Public										
	Private										
Total		15	21	47	57	66	60	79	90	100	161

stratum, the disinfection and sterilization of foods and drugs, and the acceleration of polymerization reaction in highly polymerized compounds are experimentally utilized in industries. The atomic battery has been also taken up in research projects in two companies.

In Japan there are several cases in which radioisotopes were applied to civil engineering or public works. One case was the investigation of the movement of rolling stones carried out in the Tone River in order to obtain necessary data for sand control works. In this investigation, stones of different sizes were tagged with radiocobalt and radioactive stones were placed on the river bed. Next, how long these stones would be moved in various stages of a rising or flood of the river was detected and the relative function of the sizes of stones to the velocity of running water was investigated with considerably good results. Another was the investigation of floating sand carried out on the Tomakomai Coast in Hokkaido. On this coast, the harbor constructions are extremely obstructed because the sand there is violently moved owing to wind and waves. In order to investigate the state of movement of this sand, radioactive glass grains having the same size and specific gravity as sand grains were made by using radiozinc. The glass grains were scattered on the sea bottom and the movement and distribution of floating sand were investigated.

There is an example of the application of radiocobalt to antique works of art, which goes beyond the proper area of industry, but is very interesting. In Japan, there are many Buddhist statues supposed to have been made during the time from the Asuka period to the Nara period. These statues were radiographed by radiocobalt to make clear the inner structure and the method of the making. By this test were determined the time and place of the making and the maker of the statue, which were hitherto only roughly judged from the costume and facial expression of the statue. Moreover, this test plays an important part in securing the preservation of these antique works,

TABLE 4. List of Numbers of Users (Representative of Groups) and Cases of Use of Radioisotopes

Year	*1950*				*1951*		*1952*				*1953*				*1954*			
Classification by field	*Former term*		*Latter term*		*Whole year*		*Former term*		*Latter term*		*Former term*		*Latter term*		*Former term*		*Latter term*	
	User	*Case*	*User*	*Case*	*User*	*Case*	*User*	*Case*	*User*	*Case*	*User*	*Case*	*User*	*Case*	*User*	*Case*	*User*	*Case*
Physics	4	6	7	7	13	16	19	29	9	15	16	28	22	26	15	16	20	47
Chemistry	2	3	7	7	33	35	20	30	16	27	16	22	31	53	15	30	35	62
Industrial research or industry							19	31	20	33	15	23	8	17	31	39	37	52
Biology							18	26	13	14	11	12	15	17	14	16	13	13
Plant physiology	7	8	13	16	34	44	18	21	11	14	30	37	26	30	29	34	43	65
Animal husbandry							7	8	6	6	9	11	7	7	11	14	21	62
Medicine	26	42	40	50	90	135	150	215	131	190	190	301	201	316	231	357	256	449
Others					2	2	3	3	3	3	6	6	5	5	9	11	6	7
Total	39	59	67	80	172	232	254	363	211	300	293	440	315	471	355	517	431	757

because the inner scar and crack of the statues are easily detected thereby.

MEDICINE, BIOLOGY AND AGRICULTURE

Approximately 60 per cent of isotopes used in Japan in 1954 has been in the field of medical research, diagnostic or therapeutic application and about 18 per cent in the areas of biology, plant physiology and animal husbandry.

In the field of medical research, radiophosphorus has been most widely used to study the metabolisms of phosphorus compounds in various tissues and further to investigate the relation of these metabolisms to the function of various organs. The phosphorus metabolism in different bacilli and the measurement of the volume of circulating blood have been also studied in various ways. Radioiron and radiocalcium have been used for the study of bone tissues and hematopoietic function. Radiochlorine has been applied for the measurement of liquid volume and permeability between the two tissues. In diagnosis, radioiodine has been used for the examination of thyroid function and radiophosphorus and radioiodine for the determination of the location and extent of a certain brain tumor. In therapy, radiophosphorus has been used to treat blood disorders, such as chronic leukemia and polycythemia vera. The beta rays of radiophosphorus and radiostrontium have been applied to surface lesions. Radiocobalt needles applied to accessible tumors have been used in small private hospitals. Recently the exposure container for mass-teletherapy has been improved so as to treat deep-seated tumors and 50–200 curie units of radiocobalt have been adopted in about thirty hospitals in Japan.

In biology, radioisotopes have played an effective role, for example, in the pursuit of decomposition mechanism of materials in a living body and intermediate metabolism in biosynthesis. In this area have been noteworthy the study of photosynthesis, the phosphorus and metallic metabolism in microorganisms, the production of vitamin B-12 in microorganisms, and the fixation reaction of carbonic acid gas by microorganisms. As a study peculiar to Japan, the mechanism of calcium deposition in the shell and the pearl of the pearl oyster has been investigated in a few institutions. In plant physiology, field studies using different isotopes are being carried out on such various crops as rice plant, barley, tobacco, etc. Especially, several studies are being conducted in connection with the growth of rice plants. Rice is a staple article of diet in Japan, so that it is thoroughly studied from every point of view. In sericulture too all-round studies have been pursued using various isotopes on subjects ranging from soil to mulberry trees and finally to silkworms. In the area of animal husbandry researches have been conducted mainly about the metabolism, especially metallic metabolism, in domestic cattle and fowls.

Statistical Data on the Production and Use of Artificial Radioisotopes in France

By C. Fisher,* France

The Atomic Energy Commission, in France, is the only producer of artificial radioisotopes for use by the public. The first of these were delivered to the users as early as 1949. Until now, they consisted exclusively of the elements created in nuclear reactors. The elements produced in cyclotrons and the electromagnetically separated stable nuclides are not yet being produced on a routine basis.

A description will be given of the following aspects of the utilization of artificial radioisotopes in France: artificial radioisotope consumption in France; French production and imports; and education and application studies.

CONSUMPTION OF ARTIFICIAL RADIOISOTOPES IN FRANCE

The over-all number of deliveries of the radioisotopes in use in France increased as follows: 1949, 147; 1950, 1199; 1951, 2128; 1952, 2107; 1953, 2758; 1954, 3315; 1955 (3 months), 1114 for a total of 12,768 deliveries.

Over the year 1954, the French users of radioisotopes were broken down between the following categories:

Nature of work	*Number of users*	*Number of deliveries*
Medical and therapeutic research	27	1281
Scientific research (physics, chemistry)	17	711
Animal biology	17	142
Plant biology	5	17
Industrial applications	51	255
C.E.A.		829
Miscellaneous	21	80
Total	138	3315

The Atomic Energy Commission alone used 25% of the radioisotopes. Among the other users, it will be noted that medicine, which takes up 39% of the available radioisotopes, accounts for only 20% of the users, while the industrialists use but 8% of the radioisotopes, even though they represent 37% of the users. In fact, only properly qualified hospitals may receive radioisotopes, which limits the number of medical users, although there is no restriction on the number of deliveries.

It would have been of interest to know, from year to year, whether the various categories of users were increasing or decreasing. Unfortunately, these data could be gathered only for the radioisotopes prepared by the C.E.A. Although they are limited, they mirror the general trend. The number of the users served has been: 6 in 1949; 33 in 1950; 45 in 1951; 49 in 1952; 59 in 1953; 104 in 1954; and 82 in 1955 (3 months).

The uses for scientific research, animal and plant biology have changed but little. There are 600 deliveries a year on an average, for scientific research, and 100 for biology, exclusive of the research conducted at the C.E.A.

The increase in the number of the medical applications, which trebled between 1953 and 1955, is not significant. It is due to the replacement of former imports by new production: in fact, the demand from the physicians has not increased over the last two years.

On the other hand, the number of industrial utilizations has been as follows: 8 in 1950, for a total of 5 users; 71 in 1951, for a total of 11 users; 97 in 1952, for a total of 10 users; 131 in 1953, for a total of 13 users; 174 in 1954, for a total of 23 users; and 80 in 1955, (3 months) for a total of 14 users.

During the year 1954, the industrial applications made in France broke down as follows:

Nature of the work	*Number of users*
Mechanical constructions	22
Metallurgy	11
Measuring instrument making	7
Chemical industry	4
Mining, oil	2
Textiles	1
Miscellaneous	4

The major area, for the utilization of the radioisotopes, is the Paris region, which accounts for two-thirds of the users. In the provinces, one finds one or several users in 23 different centers. It is of interest to note that:

Of the 15 French provincial university centers, 12 received radioisotopes for medical or industrial applications in 1954. These were: Algiers, Bordeaux, Clermont-Ferrand, Grenoble, Lille, Lyon, Marseille, Montpellier, Nancy, Rennes, Strasbourg and Toulouse.

Gamma-ray inspection was the most significant industrial use. Most of the other centers of utilization

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Original language: French.

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have important industrial centers: Le Havre, Saint Nazaire, and Saint-Etienne.

Finally, radioisotopes were supplied to countries of the French Union, particularly to Sénégal and La Réunion.

PRODUCTION AND IMPORTS

Radioisotope production depends on the neutron fluxes and space available for irradiation. The Châtillon and Saclay reactors are of the heavy water type. In other words, they offer the advantage of supplying a relatively intense flux, but the drawback of accommodating only a relatively small number of samples, which can be placed in the reflector.

The fluxes available in the vicinity of the core, in the C.E.A. reactors, were the following: May 1949 to July 1953—Zoë reactor—5×10^9 n/cm^2/sec; from July 1953 on—Modified Châtillon reactor—2×10^{11} n/cm^2/sec; from September 1954 on—Saclay reactor—2×10^{12} n/cm^2/sec.

The flux available at the Saclay reactor is satisfactory for most needs and its potentialities, as regards the preparations it can receive, are limited, mostly by the space available for irradiation.

Even before the Saclay reactor was placed in operation, the French users were able to obtain the radioisotopes which they needed, and which the C.E.A. could not deliver to them, from foreign countries. As a matter of fact, as each foreign country showed itself ready to export a part of its production, France endeavored to make it easier to import such elements. Business relations were developed successively with the United States, Great Britain, Canada and the Netherlands.

The following table shows the number of imports into France since 1949:

Year	*Imports*			
	Great Britain	*United States*	*Netherlands*	*Canada*
1949	21			
1950	259	5		
1951	674	4		
1952	944	21		
1953	1248	14	2	
1954	1168	19	9	
1955 (3 months)	299	7	6	2
Total	4613	70	17	2

The important radioisotopes are, mainly: cyclotron prepared radioisotopes (Netherlands), stable isotopes (United States, Great Britain), tagged molecules (Great Britain), long-lived radioisotopes (Great Britain), and cobalt sources for teletherapy (Canada).

The number of imports has been decreasing since 1954. Conversely, the production potentialities of the C.E.A. have been increasing from that date on. The radioelements for which demand was greatest, and the near total of which, until then, was imported, could henceforth be made in France. Their selling price, as well as the prices obtaining for French production as a whole, were lined up with the English prices.

The number of radioisotope deliveries made by C.E.A. was:

Year	*Deliveries*
1949	126
1950	935
1951	1450
1952	1142
1953	1494
1954	2119
1955 (3 months)	800
Total	8066

The number of deliveries has been increasing in a nearly constant fashion. This increase is due, in particular, to the new production program.

Thus it is that the number of P^{32} deliveries coming from C.E.A. underwent the following changes: 1950, 21 deliveries; 1951, 93; 1952, 86; 1953, 128; 1954, 397; and 1955 (3 months), 158. Since 1954, 6.7 curies of P^{32} were imported, and the C.E.A. supplied, from the elements separated in its own laboratories, 7.9 curies in 1954 and 5 curies for the first quarter of 1955.

As regards colloidal Au^{198}, the preparation required a specific activity of several curies per gm of gold and could be undertaken only with the help of the Saclay reactor. A part of the demand was met by the C.E.A., beginning in January 1955 (40 deliveries in 3 months with an activity of 7.6 curies). Since March, it has been meeting the entire demand.

In the field of C^{14}-tagged organic molecules, C.E.A. concentrated its main effort on the synthesis of the compounds which could not be obtained from Great Britain. Thus, in two years, out of 77 C^{14}-tagged molecule deliveries, only 13 were made by the C.E.A. These, however, were fairly complex molecules: DL-Valine, isonicotinic hydrazide, heteroauxine, glycyl-valine, etc.

EDUCATIONAL PROGRAM AND STUDY OF THE APPLICATIONS

Even as the artificial radioisotopes were being prepared, it became patent that this new means of investigation would be used to better purpose if their use could be promoted more actively. This has mostly taken the form of the organization of specialized education and the development of a department for the study of industrial applications.

Education of the radioisotope users was organized beginning in 1950 by the National Atomic Energy Commission, the National Center for Scientific Research and the National Institute of Hygiene. Its purpose, through courses and practical work, is to train research men of diverse backgrounds in the use of the artificial radioisotopes.

There are two sessions each year, in May and October, attended by some 25 to 30 students with whom two groups are made up (biology-medicine, physics-chemistry-industrial applications).

Since 1950, 283 persons have attended the courses, 150 of them in the biology-medicine group, and 153 in the industrial applications group. Among these, there are 41 foreigners of 21 different nationalities.

Broken down by occupations or professions, the research men were distributed as follows: researchers†, 121; physicians, 66; engineers, 46; military men, 47; and miscellaneous, 3.

Also, beginning in 1950, the Atomic Energy Commission undertook, for various industrial organizations and at request, some specialized studies on the applications of the radioelements.

This demand multiplied rapidly. If we do not allow for the numerous counsels and consultations given by the technicians of the C.E.A., their number reached 4 in 1950, 8 in 1951, 10 in 1952, 12 in 1953, 36 in 1954, and 42 in 1955 (3 months). All of these requests were not such as to end up in an application, and 48 were left pending out of a total of 112.

The studies effectively carried out bore on: nondestructive control,‡ 28; static electricity discharge, 12; action of radiation on matter, 7; agriculture, 4; hydrology, 3; analyses, 3; and miscellaneous, 7.

The industries affected are extremely varied and include, in particular, the chemical industry, textiles, paper, automobiles and metallurgy.

Finally, in order to complete the training of university or industry educated personnel, the C.E.A. welcomes outside students to its laboratories for study periods. This form of cooperation is much encouraged when dealing with the study of a specific industrial application.

CONCLUSIONS

The development of the utilization of radioisotopes in France has been rather rapid, and it has not as yet reached its maximum (see Fig. 1).

Some techniques which are now already considered conventional in medicine and industry will receive ever-widening applications. The radioisotopes needed for this development are now available.

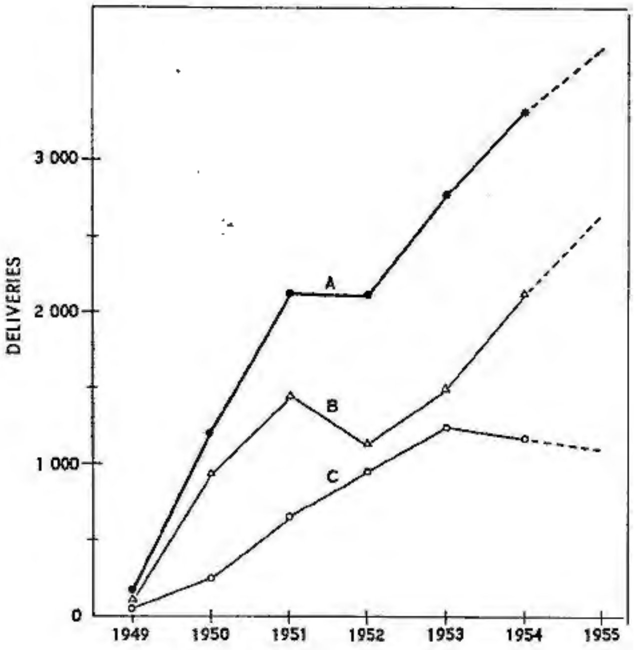

Figure 1. Distribution of the artificial radioisotopes in France: (A) total; (B) prepared by the C.E.A.; (C) imported

The above applications, however, bring into play only a small fraction of the radioelements which can be produced, and the activities used are very small in relation to the enormous possibilities afforded by the fission products. When very high intensity and small volume, i.e., concentrated, sources will be available at reasonably low prices, it is likely that investigation and application fields which are now inaccessible will at once be explored, both by the physicians and industrialists.

Although the manufacture of such sources can not take place in France at this writing, this is the direction in which the efforts of the Atomic Energy Commission must be oriented.

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† Universities and government laboratories.

‡ Measurements of thickness, wear, homogeneousness, etc.

The Industrial Use of Radioisotopes in France

By M. J. Guéron,* France

As soon as a nuclear industry is developed, it becomes possible to use the radioisotopes produced in a great many fields. In particular, a wide variety of industrial applications soon come to the fore. It belongs to the authorities concerned with the development of atomic energy to promote the use of these secondary, but significant, applications.

The Atomic Energy Commission, to this effect, took three sets of measures: (1) it opened up specialized courses; (2) it encouraged the equipment manufacturers; and (3) it created a consulting service for the users.

The courses began in 1950. They are organized, with the help of the National Center of Scientific Research, under the responsibility of Mr. Grinberg, and last for a full five weeks. Lectures are in the morning, practical laboratory work in the afternoon. After a general introductory course, common to all, the students are made up into three groups: medicine, biology, physics and chemistry. There are two sessions a year, attended by approximately 25 persons each. One-fifth to a third of the students are foreign researchers who work in France, or come to France, to follow the courses.

The C.E.A. first had to make its own detecting and counting devices. It turned over a substantial number of these for outside laboratories. For two years, various manufacturers have been making C.E.A. models on a production basis. The C.E.A. confines itself to the making of prototypes and short series.

On the other hand, several firms offer detectors made to their own designs and specifications, or import foreign instruments.

A group of engineers from the Atomic Energy Commission has been devoting its efforts, since 1950, to the industrial applications of the radioisotopes. The group endeavors to create such applications, and to extend its help to the new-comers to the field. The preferred method is to have an employee of the organization which raises a problem work with the group for a while. This serves to train specialists. Some fifteen visiting engineers, on such temporary stays, have spent periods varying from a week to several months in our laboratories.†

The request for applications of the radioisotopes received by the C.E.A. were but a few until 1953, namely about ten a year. There were three times as many in 1954, and a continuous increase was noted in 1955 (over 40 in 5 months). More than half of these consultations led to the joint execution of a research project by the C.E.A. and the applicant.

Most of the work (45%) bears on non-destructive testing, followed by static electricity problems (20%). The remainder breaks down just about equally between the use of intense radiation, agronomic problems, hydrological matters, and chemical analyses.

Obviously, the above does not faithfully illustrate the procedure followed in the use of the radioelements by French industry. Gamma-ray analysis and activation or isotopic dilution analyses are widely performed without any C.E.A. participation, while the latter, on the other hand, is not the only possible consultant used, since the equipment manufacturers play an ever growing part in that capacity.

Two comprehensive articles[1, 2] gave, in 1951 and 1953, a description of the industrial applications then being carried out or under study in France. We shall not go back to subjects already covered, but will remind readers of a few significant cases:

1. Detection of leaks in cables kept under pressure: the methyl bromide method is in routine use for a part, at least, of the French telephone network.[3]

2. Hydrological studies on the site of the Serre Ponçon power dam: in a soil made up of fine alluvial clay, radioactive tracers revealed a relatively short range transfer which a massive injection of dye had not been able to objectivize. This served to define the type of preliminary study needed for such experiments in the field.[4]

3. Detection, *in situ*, of neutron moderators: the dosage of hydrogen, mostly in the form of water, in the soil, was described. The method used indium resonance in order to make the results independent from the chemical nature of the ground (see page 74A of reference 2). The main drawback of the method is that it is necessary to activate the indium detector for a fairly long time, and then to measure its activity after it has been removed from the site of its activation. This does not allow for much more than one measurement.

By giving up the advantage inherent in the use of epithermal neutrons, a gain in sensitivity can be achieved, and some engineers of the Public Works Department developed a device, for the dosage of humidity, which uses a small BF_3 counter so mounted as to make *in situ* counting possible.[5]

Original language: French.

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† These are regulated by contracts which provide for payment to the C.E.A. of a daily indemnity, and include clauses bearing on industrial property rights, should patentable processes be developed.

Along the same lines, the comparison of the gamma and neutron activities noted on drilling logs (in which the capture gamma released by the moderated neutrons are detected) provides an excellent method for revealing coal seams (page 73A of reference 2).[6]

For the last two years, industrial applications have been mostly in four fields: activation analysis, the use of bremsstrahlung, non-destructive testing, and field studies. Activation analysis is covered in another paper from France,[7] as is the use of bremsstrahlung.[8] We refer the reader to those texts.

Among the non-destructive testing methods, a note should be made of the measure of the concentration of the air trapped in water mains. The study, which was carried out at the request of the Electricité de France, adds up, on principle, to the conventional measurement of a density by radiation.[9] We have been able to determine the amount of air trapped in a duct 60 cm in diameter with a relative accuracy of 5% approximately, with a volumetric concentration of air close to 10%. A conventional source of Co^{60} (300 mc), of the type used in radiology, is adequate for the purpose, and counter detection is suitable, as long as precautions are taken to obtain a moderate opening bundle of gamma rays. Results can be recorded and the use of a scintillation detector could bring about an improvement in accuracy.

The continuous measurement of the density of highly radioactive solutions also had to be covered. It has to be done during one of the stages of the extraction of the fission products. This can be done, on the fraction circulating through certain pipes, by the absorption of the radiation of a Co^{60} source detected by means of a crystal. A discriminator is used to eliminate the influence of radioactivity (which is much more intense) of the solution. We still have to look into the behavior of the detector under heavy irradiation and for long periods of time.

The field studies have been many and varied. In tagging of insecticide, the activated insecticide, which contains Na^{24}, could be determined by the counting of sheets on the spot (with a portable scintillator) or in the laboratory.

A first experiment was also carried out, for the purpose of determining whether it would be possible to follow the displacements of the sand, on the Mediterranean beaches, with the help of radioactive tracers. A short description of the test was already published,[10] and a more detailed description of it will soon come out.

Tagging was with Cr^{51}-containing glass, 3 millicuries per kilogram. This glass (having a density and grain sizes similar to those of the sand on the beach) is easily carried, since the Cr^{51} radiation is not very penetrating. The sensitivity of detection, however, is satisfactory: a portable scintillometer equipped with a 2.5 cm thick crystal, 4 cm in diameter, makes it possible to reveal 0.3 microcurie/kg: a fixed setup is 3 to 5 times more sensitive. Under those conditions, it is enough for a 1 kg sand sample to contain 200 grains of glass (ranging in diameter from 0.2 to 0.8 mm) in order to be suitable for detection. The dilution, with respect to the glass used, then is about 50,000. This factor can be increased, by raising the specific activity of the glass. In our case, it has been possible to follow the displacements of the tagged area for four days, under the action of a slight swell and moderate wind, before a strong two day gale came and swept everything off. The oceanographers feel that the method is interesting. It would doubtless be more difficult to apply it on the beaches of an ocean having tides. It would be of interest to compare this test with that which was conducted shortly before on the silts of the Thames estuary,[11] where the fineness of the sediment led to the use of a glass of much finer grain. The considerable dilution which took place before the detection of the tagged solid imposed, on the other hand, the use of a specific activity, and of a total activity, far greater than ours. The whole preparation of the glass was thereby modified.

The future of these applications is conditioned, above all, by the construction of portable light, sensitive, sturdy and sealed detectors. It is to be hoped that, with transistors, it will be possible to make those, giving "open air radioactivity" a new impetus.

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The Use of Radioisotopes in Austria

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1. ORGANIZATION OF THE DISTRIBUTION

As a result of negotiations between the Institute for Radium Research of the Austrian Academy of Sciences and British authorities a distribution center for radioisotopes from the Atomic Energy Research Establishment in Harwell was established at this Institute for every kind of application in Austria. The Rockefeller Institution granted a subvention of £ 500 for facilitating the beginning of the work in this field. At first the authorities of the British Element of the Allied Commission took care of the transportation. The Austrian authorities supported the new Institution in every respect: the Ministry of Finances granted exemption from duties for isotopes used for medical or scientific purposes, the National Bank carried out payment in foreign currencies in a particularly appropriate way. The centralized regulation of all pertaining questions has many advantages in administrative and financial respects, in particular all sources come under control as regards measures against radiation health hazards.

In 1953, the necessary formalities were settled by the Chancellery and the Austrian Embassy in Washington for the import of radioisotopes from the U.S. Atomic Energy Commission. It was decided that all applications had to be submitted to the Institut für Radiumforschung for approval and that the import had to be controlled. Yet for technical reasons, so far practically all shipments have come from Harwell. In 1954, the Philips Company contacted the Institute about the import of isotopes from the Norwegian-Dutch pile and the Amsterdam cyclotron. So far there has been no need for an import from these institutions.

To prevent health hazards, preparations are only delivered to persons observing the necessary safety measures. Sources must not be passed on without the consent of the Radium-Institut.

For therapeutic use radioisotopes have been distributed only to clinics and public hospitals under the condition that a radiologist is consulted. The Labour Control Office sees to the observation of safety measures in industry. The Radium-Institut is arranging training courses in radioisotope technique and can be consulted at any time on special problems. The physicist in charge of the distribution has gathered additional experiences by participating in conferences and by visiting corresponding laboratories in England, Sweden and Switzerland. There was also opportunity for collaborators of the Institute to see French, Yugoslav and American institutions. Experiences are also exchanged with the German Radioisotope Distribution Center in Göttingen, and arrangements have been made with this office regarding the control of the import of isotopes from Germany. The British Council sponsored the training of Austrian medical doctors in the use of isotopes.

The total activity distributed in Austria since 1949 amounts to 250,000 mc in the form of more than 2000 single preparations handed over to some 60 institutions in 16 different places. Table 1 shows the shares which research, medicine and industry had in the use of the imported isotopes, as well as the demand increasing year by year.

2. MEDICAL APPLICATIONS

About 30 clinics and hospitals have used iodine, phosphorus, gold and cobalt with a total activity of

TABLE 1. Increase of the Use of Radioactive Isotopes in Austria

Date	*Medicine*		*Industry*		*Research*		*Total amount*	
	*N**	*mc*†	*N*	*mc*	*N*	*mc*	*N*	*mc*
1949	4	6	–	–	2	1	4	7
1950	46	218	–	–	2	1	46	219
1951	150	2249	1	15	10	18	161	2282
1952	173	3070	8	23394	12	90	189	26554
1953	541	7065	19	97341	11	30	558	104436
1954	795	20079	13	66274	21	64	821	86417
1955	1235	5992	3	22200	2	4	240	28196
Total	1944	38679	44	209224	60	208	2019	248111

* N = number of preparations. † mc = activity in millicuries.

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35,000 mc for diagnostical therapeutical and research purposes, particularly in clinical diagnostics.

Diagnostics

Using radioiodine the functioning of the thyroid gland and the position of strumas and metastases of carcinomas of the thyroid have been tested. Up to now 12 Austrian clinics and hospitals have applied the method of radioiodine examinations. In the Second Medical Clinic of the University of Vienna, in which by far the most comprehensive experiences were gathered, no less than 3000 tests of uptake and 800 plasma tests have been carried out for examining the functioning of the thyroid gland; 70 tumours have been located.

Therapeutics

Iodine is used for the treatment of thyreotoxicoses, of heart diseases and of carcinomas of the thyroid. Altogether about 35 cases of carcinomas have been treated with very high dosages.[C18, 23]

Cobalt is applied in treating tumours in a number of hospitals marked with "Co" on the list.

Phosphorus has been used in 80 cases of polycythaemia vera and in a few cases of lymphosarcoma and leucaemia.

Gold has been administered in an intravenous form on certain cases of (chronic-myeloic) leucaemia[C19] and liver metastases while with certain metastases and maliquant effusions intraperitoneal or intraplernal application has been used.[C32]

Thorium-X proved to be successful in the treatment of Morbas (about 80 cases).[C2]

Medical Research

The uptake of iodine by the human and animal organism and the iodine metabolism were examined under very differing conditions. A series of investigations deals with the importance of the chemical form of iodine for the uptake.

The influence of various treatments, e.g., the administration of potassium perchlorate, chronic lack of iodine, strumectomy[C7] on the functioning of the thyroid as well as the connection with different organs (adrenal gland, nervus sympathicus) was investigated.[C22]

The resorption of iodine by the cornea,[C8, 24, 25, 26] skin[C27, 29] and lungs[C21, 27, 29, 30] was traced; studies of particular importance for therapeutic applications.[30, 3] Moreover iodine served as indicator in the following investigations: problems of the blood circulation, peripheral tissue clearance, measurement of the plasma volume, and permeability of isolated arteries.[C6]

Phosphorus was used for measurements of the blood volume and for studies of the influence of radiophosphorus therapy on the coagulation of blood.

Gold served for a new method for measurement of the blood circulation through the liver.[C17, 20] The influence of gold therapy on the coagulation of blood[C15] was investigated, and the possibilities of an infiltration therapy with gold in the treatment of certain tissue diseases were explored.

Sodium was used for radiocardiographic studies.[C13]

3. SCIENTIFIC APPLICATIONS

Analytical Chemistry

Analytical procedures for germanium were tested with radioactive germanium. Microanalytical methods for iodine were worked out.[A1, 2, 3]

Biochemistry

Several groups in Austria are using radioisotopes in this field. The following problems are being thoroughly investigated:

Metabolism of tissue cultures: the amounts of different substances (fats, proteins, amino acids) that the cultures use up as well as the amounts they produce are studied with C^{14}. It is intended to compare normal and cancerous tissue.[B5]

Tobacco mosaic virus: the conditions of growth were studied carefully.[B3, 4, 16]

Biosynthesis of sugar marked with C^{14}: the methods have been so much improved that the sugar (required for the tissue cultures) is now prepared with specific activity. Purity and yield have been much improved.[B1, 2]

Metabolism: experiments were undertaken regarding cancerogenetic action of this substance.[B15]

Biogenesis of lignin has been studied.[B7, 8, 9, 12, 13, 14]

Fermentation processes: A new ferment from yeast was discovered.[B6, 10, 11]

Forensic Medicine

The distribution of white phosphorus was studied.

Histology and Human Physiology

The resorption of iodine was investigated in various details.

Pharmacology

The distribution of Schadran in the organism was measured.[E9] Problems of Chlorophyllins with magnesium, zinc, and cobalt were treated, and therapeutic applications considered.[E1, 2, 4, 5, 6] Investigations were carried out in cases of injuries caused by oleum Chenopodii.[E7]

Plant Physiology

The uptake of phosphate in foliar fertilization was examined.[F2]

Metal Physics and Metallurgical Chemistry

The plasticity of metal crystals was studied in connection with problems of radiation damage.[G7, 8] Diffusion processes in metals were studied.[G1, 2, 4]

4. INDUSTRIAL AND TECHNICAL APPLICATIONS

γ-Radiography

Eight industrial institutions have used about 40 sources of cobalt and iridium with more than 200,-

000 mc total activity for material testing purposes. The sources have proved particularly valuable in the testing of large pipelines of waterpower stations in the Hogh mountains.

Ground Density

The method for measuring the density of the ground by γ back-scattering was developed further. Whereas a ratio of about 15–20 is given in the literature for background to scattered radiation, a ratio of 70 could be reached. The improved method was used successfully for measurements in the city of Vienna.

An apparatus for snow-measurements using wireless transmission of G-M-counter-pulses has been constructed and tested (Siemens-Halske).

A radiographic method for the examination of surfaces was developed.$^{G5, 6}$

5. AGRICULTURAL APPLICATIONS

The difference in iodine-uptake by cattle living in warm stables in the winter and animals living in the open air was measured.^{F3} Problems of foliar-fertilization were treated, and extensive studies of the distribution of fertilizers and of trace elements in the soil are planned.

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Use of Some Radiation Applications at the Laboratório Nacional de Engenharía Civil of Lisbon

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Among the problems of interest to civil engineers, the study of several of these can be considerably improved by the application of methods making use of radioactive isotopes.

Amongst others, the Laboratório Nacional de Engenharía Civil concentrates its efforts mostly, on the one hand, on measuring of the humidity content and specific weight of the soils; and, on the other hand, on the study of the displacements of beach sands along the coasts.

In so far as the first problem is concerned, it would be idle to take up again a subject that has already been so extensively dealt with.[1]

Conversely, we believe that the preliminary results we obtained in designing a method for the study of the displacements of the sands by means of the radioactive silver (Ag^{110}) are of some interest.[2]

This work aims at determining the amount of sand on a beach originating from another, this at the ratio of 1 to 10^{6}, i.e., in such a way that one part of the sand studied may be detected in 10^{6} parts of the local sand sampled.

Since Ag^{110} is endowed with particularly favorable properties it can be added to the sand under study. They become so strongly bound that, under the action of the sea water as studied in a large swell basin, some 70% of the silver kept adhering to the sand. On the other hand, the method used does not produce any substantial modification, either in the shape of the grains or in their specific weight. This is very important in order to justify a claim that the transport of the tagged sand gives a faithful picture of the phenomenon under study.

The sand is tagged in the following way.[3] For 1 kg of sand we prepare a 100 cm^3 solution comprised of 20 cm^3 of a solution of 50% of ordinary silver nitrate, 20 cm^3 of concentrated ammonia hydroxide, and 20 cm^3 of a 25% solution of potassium or sodium tartrate. The 100 cm^3 volume is made up with water. The radioactive isotope, Ag^{110}, in the form of silver nitrate, is mixed with common silver nitrate. We have used $10\mu c$ for 1 kg of sand.

Following dispersion, the tagged sand has to be found in samples of regular sand by detecting Ag^{110} activity. For this purpose, a calibration curve is first drawn with a measuring device having a constant geometry, made up of a Geiger-Muller tube around which there is a sleeve which can hold up to 1 kg of sand. With this device, we can measure an activity of 10^{-3} c of Ag^{110} distributed in the mass of 1 kg of sand.

Such a sensitivity thus corresponds to a dispersion of 10^{4} with respect to the activity of the tagged sand. However, we can increase the sensitivity up to

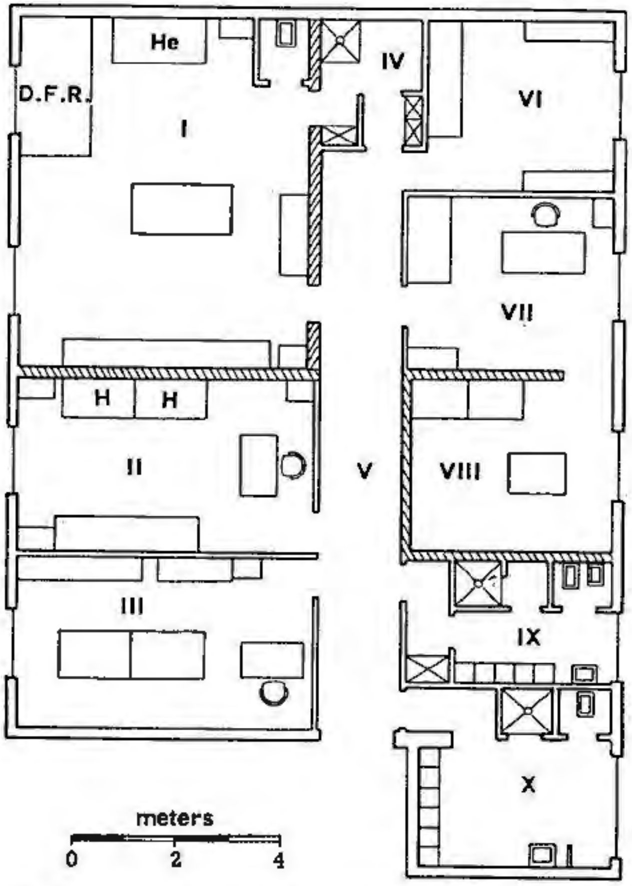

Figure 1. General plan of the laboratory. Room I: Hot room; D.F.R.: Storage of radioactive substances; He: Hood, equipped with forced ventilation and filters, concrete walls 20 cm thick, special doors of equivalent thickness, and sewers leading to a dilution chamber. Room II: Semi-hot room; H: Hoods. Room III: Room for routine type work. Room IV: Bathroom, shower; access to the filter closet, located on the roof. Room VI: Equipment, supplies and dark room. Room VII: Library, file room, meeting room. Room VIII: Calibration room, concrete walls, 20 cm thick, special door. Rooms IX and X: Locker rooms and wash rooms (men and women)

Original language: French.

a dispersion of 10^6 by a chemical treatment of the sand samples which affords a reduction to about one-hundredth of the volume of inert matter taken from the sampled sand, the reduced volume still containing all the silver that existed in the original sample.

This extraction is done in the following way: After it has been washed with water, the sand is successively treated with acetic acid (which removes the calcium), hot nitric acid, and ammonia. All the silver can be found in the dry residues of the nitric and ammoniacal extracts, so that these constitute a reduction of the mass of the sand so treated to about 1% of what it initially was.

This technique allows us to consider labeling 10 tons of sand with barely 100 mc of Ag^{110}, thus under good conditions of safety. Some difficulties, which did not arise out of the method under study, made it impossible for us to apply it in an actual test, but we believe, nevertheless, that it will not be useless to mention the results achieved in the laboratory, particularly when proper regard is given to the extreme dispersion allowed by this method.

This paper provides an example of an application of radioactive isotopes in which they are used in the form of a compound which participates in various chemical and mechanical processes.

A study of methods based on means of this type can be carried out in complete safety only in suitably equipped facilities. Accordingly, we have built a special laboratory for the handling of radioactive isotopes, of which Fig. 1 is an over-all plan.

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The Rationale of an Organized Supply of Tracer Compounds

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1. INTRODUCTION

For tracer work, radioisotopes are generally required in a specific chemical form to suit the experiment, and there is consequently a growing demand for a wide range of labelled compounds, both organic and inorganic. Many hundreds of preparative methods have been described in the literature, but in this paper we are concerned with the more general technical and economic factors which in our experience affect an organised supply of these materials.

In producing a labelled compound there are essentially three stages: neutron irradiation of a target material in a pile, separation of the required isotope in its primary chemical form from the irradiated material, and conversion of the primary chemical form into the desired compound. These three processes are usually distinct. The chemical form of the isotope at the second stage depends on the nuclear reaction chosen and the extraction process, and this is only rarely the chemical form required by the user. One therefore has to proceed by chemical or biological synthesis. Attempts have been made to prepare labelled compounds by direct irradiation of appropriate target materials, but the small yields, low specific activity, lack of control over position of labelling and difficulties with subsequent purification, render these methods of little practical importance so far.[1-3]

As most of the more common radioisotopes can now be obtained in separated form from various national atomic energy organizations, the individual research worker is seldom concerned with the first two stages. For a while a number of laboratories using the tracer technique extensively undertook the synthesis of labelled compounds for themselves, but such work is now generally regarded as being unrewarding, and it is customary to purchase labelled compounds from laboratories specializing in their preparation. Nevertheless, no organized supply can hope to provide all the compounds that may be asked for, and it is clear that the individual research worker must still undertake to prepare certain special materials for himself, just as he would in the case of other special reagents not available commercially.

Clearly there is much in common between the manufacture of ordinary reagent chemicals and the production of isotopically labelled compounds, and the general advantages accruing from large scale production apply equally in both cases; costs are reduced, high standards of quality can be maintained, and developmental work can be supported by experience and physical resources. Nevertheless radioactive reagents have a number of distinctive features affecting their manufacture.

Unstable materials are familiar enough in chemical commerce, but the extreme range of half-lives characteristic of radioisotopes and the fact that their rates of decay are immutable, are novel features of radioactive chemicals; so also is the concept of specific activity, which is of course similarly variable. For isotopes whose half-lives are less than a few days, an organized supply of their compounds is hardly practicable, and for those measured in weeks or months an acceptable level of efficiency of production is difficult to attain, particularly when the demand is spasmodic as it is at present. Clearly the waste of effort entailed in repeated preparations of short-lived compounds can be reduced by making them at the highest possible specific activity, so that the compound remains usable over the period of several half-lives, but the onset of decomposition through internal irradiation sets a limit to this procedure.[4-8]

The relative abundance of the radioisotope in a labelled compound is very rarely as high as 25%, and is commonly much lower, often indeed almost negligibly small. Therefore chemical quantity and radiochemical quantity are independently variable within very wide limits. In the product a high specific activity is generally desirable, and because the chemical size of the batch cannot be reduced below a minimum set by ordinary manipulative difficulties, the radioactive scale of the preparation has to be as large as possible; and this in turn is limited by the marked available for the product, the intrinsic cost of the isotope, and possibly also by radiological hazards. Such considerations all favour a centralized source of supply of labelled compounds, and preferably one which is closely associated with the primary source of radioisotopes themselves.

The radiological hazards associated with many labelled compounds also distinguish them from ordinary chemical reagents. The fact that a radioactive poison cannot be denatured, the significance of imponderable amounts of the more hazardous isotopes, the possibility of effects at a distance from the source, and the insidious nature of radiation damage generally, all pose problems in control which are quite different from those familiar in the case of chemical toxicity. Such considerations apply not only to manufacture, but also to distribution, which is generally controlled by legislation.

Finally, isotopic labelling introduces criteria of purity which are novel as compared with ordinary chemical standards. The sensitive methods of detection make extreme precautions necessary to maintain not only isotopic purity but also radiochemical purity (freedom from unwanted labelled molecules) which is not necessarily related to the chemical purity. The numerons possibilities of position isomerism create another requirement of radiochemical purity, and also restrict the choice of synthetic methods and greatly increase the number of distinct compounds. Thus if we introduce one isotopic carbon atom we have three species of pyruvic acid and six of glucose.

The extent to which these generalities affect the manufacture of a particular class of labelled compounds varies tremendously according to the character of the isotope concerned, and so we may now consider them individually.

2. CARBON-14 COMPOUNDS

The unique interest of carbon-14 need hardly be emphasized; it is the most widely used tracer isotope. The target material used in its production is now commonly a metallic nitride,[11] and to obtain a high specific activity the carbon-12 content must be very low, a difficult requirement to meet in producing the relatively large quantities of target material needed. Long irradiation periods and high neutron fluxes are necessary for an adequate output, and consequently carbon-14 is one of the more expensive isotopes. The current standard prices are £12/mc in England and $36/mc in the United States.

Isotopic abundances of up to 25% are now fairly freely available. At this level 1 mc corresponds to about 0.8 mg of elementary carbon or 0.06 mg molecule (mM) of any compound labelled in one carbon atom. Labelled compounds are not often prepared at this level, but 5–10 mc/mM is now not at all unusual. Experience in many laboratories shows that 2–20 mM (depending on the character of the reactions) is the smallest quantity which it is usually convenient to employ in preparative organic chemistry, even with specialized techniques,[12, 13] and this implies batches of 10–200 mc if specific activity is not to be further reduced by carrier dilution. In terms of weight this means (roughly speaking) batches of 0.1–1.0 gram.

Such high specific activities are not necessary for all types of tracer work. If they can be reduced 10–100 fold or more, preparations come within the scope of conventional practical organic chemistry and large radiochemical batches are no longer obligatory. Many syntheses of this kind are carried out by individual workers, although there is still a widespread reluctance to undertake such work, which may be attributed to excessive caution over the hazard of using C^{14} and the risk of loss of valuable material.

It is obviously a duty of any organized supply to provide the high specific activities which are beyond the reach of the individual worker. For compounds in regular production they may even be cheaper, as the small-scale preparations often give distinctly better yields than those obtained by standard methods. For occasional special syntheses in small quantity the reverse is usually true, as the development and establishment of small-scale methods is time-consuming, and the resulting increase in cost is not usually offset by the improved yield in a single preparation.

Carbon-14 is conveniently extracted at present only in the form of carbon dioxide, and the atom to be labelled must therefore be derived from this chemical form. The chemical aspects of this kind of syntheses cannot be dealt with here; they have been reviewed by several authors.[9, 12-15] Both chemical and biological methods are used. The long half-life permits of preparation of large batches for stock, and the soft beta radiation requires no screening or remote handling. The ingestion hazard of C^{14} is now regarded more liberally than formerly,[16, 17] and control of contamination is upon the whole less difficult than with most other radioactive materials.

Chemical synthesis, when possible, is generally preferred to biological synthesis. Its advantages are control over the products and specificity of labelling, and relative ease of purification of the compounds produced. Disadvantages are the tedious multistage syntheses often required and the losses and complications introduced by stereoisomerism and optical isomerism. The slow progress made in preparing ring-labelled benzene compounds reflects the first disadvantage, for it is a long way from carbon dioxide to (say) ring-labelled paraphenylenediamine or phenylalanine. The second disadvantage handicaps amino-acid synthesis at high specific activities, as resolution of DL-mixtures on a small scale is difficult and wasteful; even more does it handicap the synthesis of labelled sugars. Methods which are in fact partial syntheses such as those for cholesterol-4-C^{14},[18, 19], glucose-1-C^{14},[20], arginine-omega-C^{14},[21] and methionine-methyl-C^{14},[22], obviate or reduce this particular difficulty but such methods inevitably impose their own restrictions on position labelling.

Biological preparations with C^{14} present a more complex topic. Up to the present time simple methods of total biosynthesis have been most used, and some of these are very satisfactory. The photosynthesis of uniformly-labelled glucose, fructose and sucrose in detached leaves is one of the most efficient biological preparations.[23, 24] The use of *Chlorella*[25] or *Thiobacillus thiooxidans*[26] labelled with C^{14} as a source of protein and L-amino-acids, although having a fairly good total efficiency, suffers from the fixed composition of the protein, so that some of the amino-acids remain very scarce. All these methods utilize the isotope efficiently in its cheapest form, carbon dioxide, and the microbiological cultures have the advantage that the isotope may be recovered from the residues without fall in specific activity. For minor components such methods are less attractive on account of the large investment in isotope which is necessary, and even nucleic acids and nucleic acid components must at present be considered as being on the borderline of economic production in this way. Intact higher plants grown for relatively long periods in a carbon-14 dioxide atmosphere have been used to prodoce minor plant components such as digitoxin[27] but the yields are exceedingly small and the specific activity limited by the radiation which the plant will withstand over a long period. Intact animals are especially wasteful as a source of labelled compounds; a yield of 0.25% of glucose-3, 4-C^{14} from bicarbonate is not unrepresentative.[28] The preparation of C^{14}-labelled plasma proteins by McFarlane and his co-workers[10] is one of the most successful of its kind, but such materials are inevitably expensive to produce and must, it seems, remain so. The concept of an "isotope farm" has not in fact proved practical.

What may be termed "controlled biosynthesis" has been little exploited in this context, but has rich possibilities. Examples recently described are the enzymatic amination of synthetic alpha-ketoglutaric acid to give L-glutamic acid-1, 2-C^{14},[29] and the conversion of glucose-1-C^{14} to cellulose-C^{14} by the isolated cotton plant boll.[30] Such methods can make the best use of both chemical and biological syntheses and often give control of the position of labelling, and it is in this direction that biosynthesis may be expected to develop in the future.

Some biosynthetic methods can provide specific activities much higher than can be reached by laboratory synthesis. At the Radiochemical Centre *Chlorella*-C^{14} has been grown successfully at 4 mc/m-atom, and photosynthetic sugars up to 6 mc/m-atom. General and uniform labelling is obtainable, which is usually practically impossible by synthesis. Often, however, biosynthetic compounds are neither specifically nor uniformly labelled, and such labelling is not as a rule much in request by tracer workers, although for some uses it is no handicap. Another disadvantage of biological methods is that the extraction and purification procedures necessary are often tedious, and may present particular difficulties with large quantities of radioactive material.

The value of carbon-14 as a tracer and its convenient properties have made it possible to prepare and distribute C^{14} compounds in far greater variety than has proved practicable for any other isotope; a number of different laboratories now supply between them some hundreds of labelled compounds and the range available continues to increase rapidly. Demand and supply have not unnaturally concentrated upon intermediates of general and fundamental biochemical interest, such as sugars, amino-acids, fats, and purines (and their precursors and metabolites) rather than on compounds having highly specific biological effects, such as vitamins, carcinogens, and synthetic drugs. Carbon-14 has long ceased to be a novelty, and has become a basic tool in the systematic study of metabolic processes.

3. TRITIUM COMPOUNDS

Tritium is a cheap isotope (£20/curie in Great Britain, $100/curie in the United States), very high specific activities are available, and the half-life (12 years) is conveniently long. In the quantities used in tracer work it cannot be considered a very hazardous isotope.[31, 32] It has found uses as a hydrogen tracer and as an auxiliary tracer for carbon. As a hydrogen tracer the large isotope effects are a distinct disadvantage.[33, 34] Tritium has aroused more interest as an auxiliary tracer for carbon, sometimes because of its cheapness but often when the use of carbon-14 is not practicable because of insufficiently high specific activity or impossibility of labelling.

Tritium labelling is usually achieved by exchange of "active" hydrogens, catalytic hydrogenation of unsaturated compounds, or catalyzed hydrogen exchange. The procedures are at once simpler and less amenable to systematic development than are carbon-14 syntheses. Caution must sometimes be exercised; Glascock and Reinius[35] have shown that tritium added to the double bond of elaidic acid in the presence of a noble metal catalyst is by no means exclusively located at carbons 9 and 10. Compounds labelled by tritium-hydrogen exchange have to be degraded to determine the position of the isotope; the labelling is sometimes complex.[36, 37] Biological synthesis is little used with tritium. The facility of hydrogen-water equilibration means that the isotope utilization is usually very inefficient, and although the isotope is cheap the large radiochemical batches which would be required are an embarrassment; while the radiation dose received by the organism used (from its labelled water content) will set a limit to the specific activity of labelled organic compounds obtained in this way.

Tritium is still not very widely used. Many workers are undoubtedly deterred by the prospect of operating the gas-counting methods which have been most generally employed; it remains to be seen whether scintillation counting of tritium will encourage its use as a tracer. The lack of an adequate supply of tritium-labelled compounds is also a handicap. If a sufficient demand existed such a supply could be organized; it could hardly at present be economically self-supporting, and the slow development of interest in tritium does not justify great expectations of its future in general tracer work, although for specific applications (in routine analysis or diagnostic procedures, for example) it should prove of value. The present character of tritium applications, on the fringe as it were of carbon-14 tracer work, makes the demand for compounds extremely varied but small, and the choice of items for a central supply would be perplexing. The extremely high specific activities which are a special advantage of tritium labelling will certainly lead to extensive radiation decomposition, and this will make it difficult to hold stocks at such levels.

4. IODINE-131 COMPOUNDS

Iodine-131 is in abundant supply from irradiated tellurium or from fission products. Its extraction in high purity and at high specific activity (10^4 mc/mg) as iodide is simple and relatively cheap. Its short half-life, however, precludes stock holding of iodine labelled compounds and severely limits the time allowed for syntheses; this, combined with the additional difficulty of radiation protection and remote handling, determine the types of chemical manipulation which can be carried out with iodine-131.

The production of iodinated human serum albumin, for example, is complicated, not only by the chemistry, the low radiochemical yield and the protection required, but also by the need to operate throughout in aseptic conditions and to allow time for biological testing of the product. This is one established exception to the conditions which have previously been stated and the additional cost both on capital equipment and decay losses can only be justified by a supplier when the demand is regular and large.

Another approach to the problems incurred by the short half-life of iodine-131 is to seek rapid and simple synthetic routes which can be readily and reliably reproduced when the need arises. Such methods as the direct iodination of, or exchange reactions with, inactive intermediates sometimes provide a route to compounds otherwise difficult of access. An example of this is the preparatioo of L-thyroxine I^{131}, which although a complex substance, can be unambiguously labelled by the iodination of triiodothyronine and thus readily prepared at specific activities as high as 1000 mc/mM. By contrast the standard methods for introducing iodine into organic compounds are unattractive except when the iodination is the last stage in the preparation[38] and are rarely used in I^{131} labelling, which relies more upon simple substitution and addition reactions.

5. PHOSPHORUS-32 COMPOUNDS

The radioisotope is simple and relatively cheap to prepare, either by the direct neutron irradiation of elementary red phosphorus or by the carrier-free separation as phosphate from irradiated sulphur.

The former method provides a convenient starting material for many phosphorus-32 compounds because of the ease with which red phosphorus can be chlorinated to provide such reagents as phosphorus trichloride and phosphorus oxychloride which, in their turn, are necessary for the synthesis of many organo-phosphorus compounds. The direct irradiation of phosphorus, however, provides only a moderate specific activity (1–10 mc/gram) which is inadequate for the synthesis of many organo-phosphorus compounds, e.g., anticholinesterases, where specific activities of 100 mc/gm or greater may be required. Here a situation arises similar to that found in carbon-14 work, namely all syntheses must start from one rather unsuitable material, phosphate. With the half-life of 14 days the need for speed in synthesis is not so pressing as with iodine-131, but again no stockholding is possible and the same approach to maintaining an organized supply is adopted as is the case with iodine-131.

Many phosphorus-32 compounds of biological importance, such as phosphate esters, are accessible by biosynthesis, but the small and diverse demand together with the short half-life make it hardly possible to organize a supply of such compounds at the present time.

The energetic beta-radiation adds somewhat to the difficulty of preparation of large batches of phosphorus-32 compounds.

6. SULPHUR-35 COMPOUNDS

Sulphur-35 is usually extracted as sulphate at high specific activity from neutron-irradiated potassium chloride, and is a comparatively cheap isotope. The half-life (87 days) allows of extended syntheses and also, to a limited extent, stockholding of labelled compounds, but few sulphur-35 compounds are in sufficient demand to justify holding of stocks. Most interest is again shown in compounds of biological importance such as methionine and cystine, and such compounds can be kept in stock by preparations three or four times a year, although even for these decay losses are considerable and preparations must still be considered as rather speculative. Other compounds have to be prepared as required. The difficulties and costliness of this procedure are partly mitigated by maintaining stocks of useful sulphur-35 intermediates such as sulphate, sulphur dioxide, elementary sulphur and hydrogen sulphide, or by devising methods by which these may be rapidly prepared from the target material. As with carbon-14, a scale of 2–20 mM is convenient for chemical syntheses, but the high specific activity of the isotope and its comparative cheapness cause radiation decomposition to become a more important limiting factor, even when the storage life of the compound is less than a year. Purification of the stock material at intervals as required (by use of carrier or otherwise) is a partial solution to this difficulty.

Biosynthesis has been used for sulphur-35 compounds, notably methionine and cystine. At low specific activity, and especially for the individual preparative worker, it has advantages, notably provision of the L-forms of optically active compounds. For an organised supply it seems unlikely to compete with synthesis, and its scope is of course limited.

As with carbon-14, the soft beta-radiation of sulphur-35 does not give rise to radiation exposure in handling even large radio-chemical batches.

7. CONCLUSION

There are still many problems to which the tracer method cannot be applied because the necessary labelled compounds are not available. Some call for a more thorough exploration of synthetic methods to permit of their production at a reasonable cost, but the large and diversified range of minor biological components, most of them beyond reach of synthesis, for which labelling is demanded present more difficult problems. Some of these components, especially plant products such as alkaloids and cardiac glycosides, have been prepared by total biosynthesis, in very poor yields, but their partial biosynthesis from specific precursors may eventually prove possible with greater efficiency. For others, such as specific proteins (particularly animal proteins), insulin, heparin, haemin and the like, there is not even this prospect of improving yields, and many such compounds, despite their interest, must be considered inaccessible to carbon or hydrogen isotope labelling at useful specific activities. It is for this reason that "tagging" with I^{131}, which is now rarely used when there is any better alternative, continues to be employed for protein tracer work.

It is interesting to note that complex mixtures of natural origin such as mineral oils and coal tar fractions are not susceptible of exact labelling unless by radiation synthesis, which is possible only at very low specific activity.

The development of labelled compound supply towards a wide variety of biochemical intermediates reflects their use, which is still predominantly in biological research work. There are, however, signs of a widening of interest, and indeed the inherent possibilities of tracer compounds in analysis, process control and development, and diagnosis are far greater than one might conclude from the rather meagre use so far made of them in these fields. Carbon-14 must inevitably retain its pre-eminent value as a biological tracer, but development of tracer work in fields other than biology and biochemistry will lead to greater interest in compounds of cheaper and shorter-lived isotopes.

In conclusion it may be noted that the supply of labelled compounds available to a research worker must have considerable influence upon the direction and rate of progress of his research. The technical advantages of a central laboratory specializing in the preparation of isotopically labelled materials, working on the largest possible scale, and associated with the primary producer of the radioisotopes, have been mentioned in our introduction. But if such an organization is used, it is important that the users shall have an effective influence upon its programme of production. If a national laboratory for this purpose is financed directly from public funds, some form of users' committee can be formed to consider policy; but with the continuing evolution of research it is difficult to keep such a committee fully representative. Therefore in our view the most satisfactory type of organization is a self-supporting trading activity in which the demand must determine the supply.

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An Electromagnetic Isotope Separator and Its Application in Laboratories for Nuclear Research

By J. Koch and K. O. Nielsen,* Denmark

Preliminary experiments on electromagnetic separation of stable isotopes were started at the Institute for Theoretical Physics in 1939 with the immediate purpose of preparing pure isotopic targets for the investigation of artificial nuclear transformations. In the years which followed a small separator[1] was built (resolving power $M/\Delta M \sim 50$), which made use of a low voltage capillary arc as an ion source and an electrostatic lens system for the formation of ion beams of the energy 40–80 kev. The beam analysis was carried out by means of a circular magnetic field of small dimensions. These experiments clearly demonstrated the useful emission characteristics of low voltage ion sources, which make it possible to form parallel beams of atomic ions of several hundred microamperes; in particular, favourable experimental conditions were obtained by using high acceleration voltages, which tend to reduce the effects of space charge as well as to decrease the angular spread among the ion paths.

On the basis of these early experiences a larger and improved isotope separator was designed in 1940, essentially incorporating a heavier magnet. The new apparatus, which was put into operation in 1943,[2] turned out to have a resolving power large enough to separate the isotopes of even the heaviest elements at a rate of separation which was sufficient for carrying out a number of experiments with separated isotopes in our laboratory.

Actual separation experiments were at first carried out in order to prepare pure isotopic targets of some of the light stable elements for the investigations of the group working with the 2.5-Mev van de Graaff generator of the Institute. In the course of this cooperation it was shown that ions of the neon isotopes might be effectively collected in the outer surface layers of a metal into which they penetrate due to their high kinetic energy.[3] Continued experiments seemed to show that this technique could be applied to the collection of any gas with various collector materials.[4] In particular experiments were undertaken with the separated isotopes of the noble gases. In addition to the production of gas-targets for the van de Graaff, in which case only small samples were required, larger amounts of separated noble gas isotopes were prepared for optical hyperfine structure investigations. Experiments with separated radioactive isotopes of the noble gases produced in fission, were soon taken up and became the starting point for a number of investigations to clear up the radiation characteristics of the radioactive isotopes of Kr and Xe. A survey of the work on noble gas isotopes carried out at the Institute up to 1951 was presented at the Washington Conference of the N.B.S. on "Mass Spectroscopy" and is published in the proceedings of this meeting.[5]

In connection with recent considerable extensions of the Institute for Theoretical Physics, which comprised several buildings for experimental research, a large laboratory was put at the disposal for work with the electromagnetic isotope separator, and the installation was transferred to its new site in 1951. At the same time several improvements were introduced, serving the dual purpose of facilitating the operation of the machine and producing very pure samples of a wide variety of stable and radioactive isotopes. Much attention was paid to the question of developing new types of ion sources which might be used for gases and vapours as well as for solids; high efficiency of the source, small amounts of charge material, and freedom from residual gases were further requirements. In what follows a short account is given of the design and operation of the separator and in addition a survey is made of the investigations recently carried out with the isotopic samples.

A general impression of the separator may be obtained from Fig. 1, showing the entire installation from the collector side. The ion source and associated equipment are enclosed in a large corona screen situated on top of the apparatus and mounted on a heavy perspex plate, insulating these parts sufficiently to apply acceleration voltages of 60 kv or even more. The ions of the discharge are emitted through a small circular aperture in the bottom of the ion source and are then accelerated downwards in one single step in the space between the source and a grounded cylindrical electrode below. The slightly divergent beam is rendered parallel by the action of an electrostatic, unipotential lens system, operating with a variable decelerating voltage. The magnetic analysis of the beam is accomplished by means of a homogeneous, 90 degrees sector field, deflecting the ions in circular paths with a mean radius of curvature of $r = 80$ cm. The separated isotopic beams are focused at a distance equal to r from the edge of the pole pieces. At this

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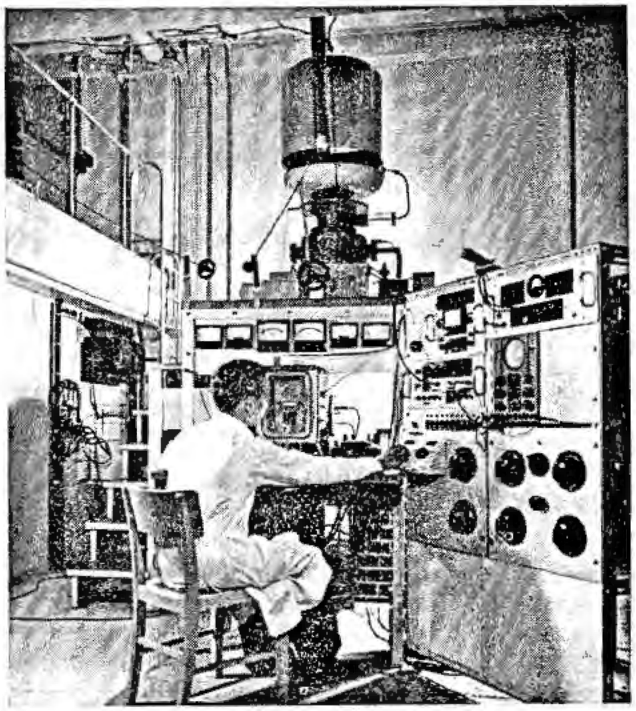

Figure 1. General view of the electromagnetic isotope separator

focus various accessories are fixed or may be turned into the beam for visual inspection of the mass spectra, for the measurement of the beam currents and of the resolving power; the particular metal plates for the collection of the separated isotopes are also located here. On the figure the operator is facing the collectors mounted behind a perspex plate, while he is adjusting the various power sources necessary to run the apparatus. The mass spectra may be recorded on the screen of a cathode-ray oscilloscope. The high voltage set and the power supplies for the ion source are located on the balcony to the left in Fig. 1. A more detailed description of the separator will be given elsewhere.[6]

Several new ion source constructions have been tried; in all cases the magnetic type was preferred having the magnetic field directed parallel to the extracted ion beam. The most recent design is shown in Fig. 2, where the different elements of construction are explained. The anode is formed like a cylinder, which at both ends is closed by plane cathodes. The electrons from the filament will thus oscillate along the axis of the cylinder. The source is operating with a hot discharge chamber, which is connected to an externally heated oven for the evaporation of solids. For separation of short-lived isotopes, e.g., from the cyclotron of the Institute, the oven may be removed from the vacuum through a valve for filling with the active materials. Considerable versatility is obtained as regards the choice of mass markers, since gases and evaporated solid materials may be added to the discharge chamber simultaneously. The source operates with milligram amounts of charge material and, since the ratio of the number of atomic ions to the total number of atoms leaving the source is high (5–15%), the isotopic separations may be carried out with great efficiency. Currents of up to 400 μa may be obtained with sources of this type.[7]

Without going into too much detail, a few other features regarding the operation of the separator may be mentioned. The adjustment of the apparatus to maximum resolving power, which may exceed one thousand, is made by scanning the isotopic beams across a slit and recording the mass spectra on the screen of a cathode-ray oscillograph. During the proper time of separation the isotopic beams are fixed on the collector plate by means of a kind of "pin stabilization," and if the beam should be accidentally displaced, e.g., by sparking, the mass spectrum will be swept sideways (in the longitudinal direction of the lines) in order to avoid isotopic contamination of the collected samples. By letting the ion beams pass through an electrostatic cylinder lens, situated in front of the magnetic analyzing field, the height of the mass spectrum lines may be reduced considerably, without any appreciable sacrifice in resolving power; this concentration of the sample on a small area is of particular importance if the sample is intended for use in a beta-spectrometer.

Among the investigations more recently carried out with samples prepared in the isotope separator the following examples may be mentioned. A beta-spectrometer study of the radiation from the 25-day P^{33}, which is produced together with P^{32} by the irradiation of sulphur in piles, was undertaken mainly to determine the maximum beta-energy but also to establish the mass assignment. Next, the optical hyperfine structure of the 9.4-yr Kr^{85} was studied using a gas discharge tube filled with about 15 μg of Kr^{85} and helium gas in excess acting as a carrier;[9] the techniques of preparing such tubes is described elsewhere.[10] The Kr^{85} sample, having an activity of about 6 mc, was prepared in the separator using about 60 mc of fission krypton from Harwell as starting material. The hyperfine structure of the Kr^{85} line at 8508 A is shown in Fig. 3 together with a similar exposure using very pure Kr^{84}. One may observe the five components originating from Kr^{85} (in two orders) and in addition one component due to a Kr^{84} impurity in the separated sample. As a result the spin of Kr^{85} was determined to be $\frac{9}{2}$. A comparison with similar exposures using separated Kr^{83} permitted the measurement of the ratios of the magnetic moments and of the quadrupole moments of the nuclei Kr^{83} and Kr^{85}. In another investigation beta-spectrometer samples were prepared by collecting radium emanation, Em^{222}, in thin aluminium foils. About 3 mc were collected from about 25 mc starting material; the density of the activity on the collector foil was of the order of 20 $\mu c/mm^2$. The isotopes of lead and bismuth were used as mass markers in this investigation. The beta-spectra from the decay of RaB and RaC were studied by means of the separated samples.[11] Quite recently several strong samples of the

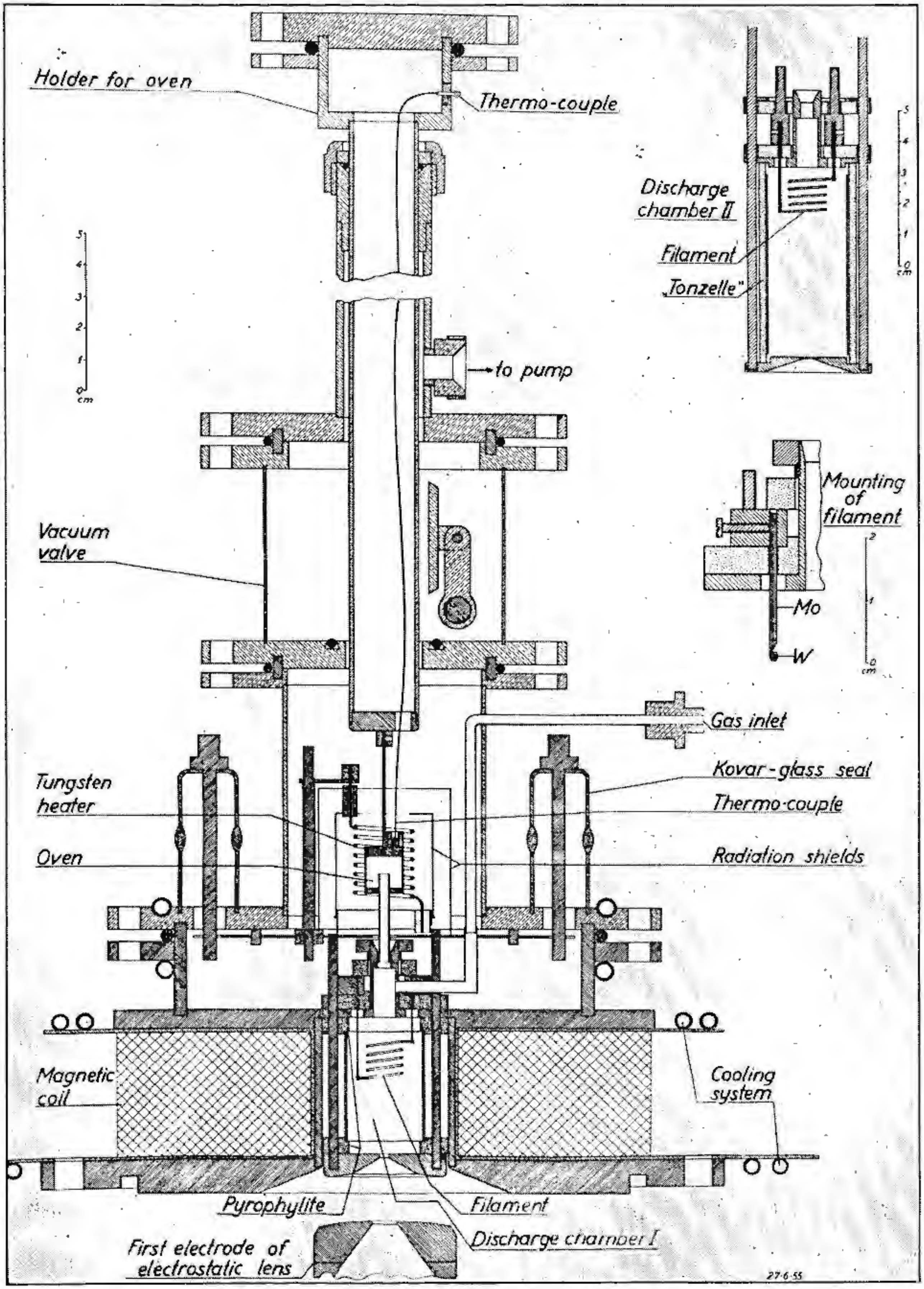

Figure 2. Design of magnetic ion source

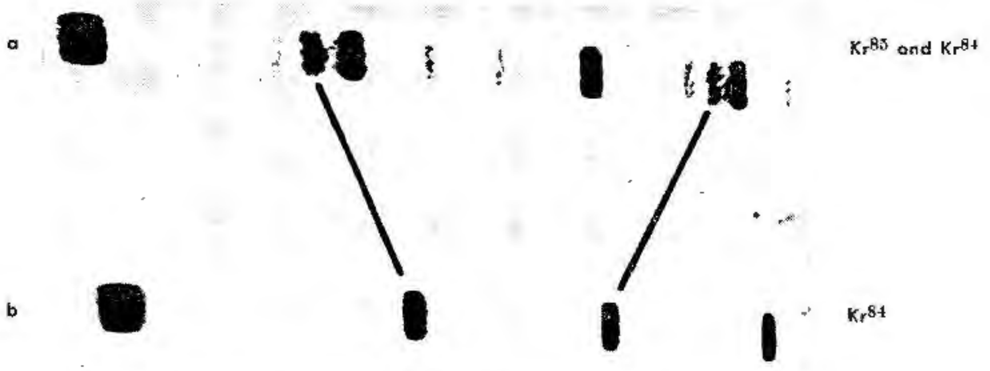

Figure 3. Optical hyperfine-structure of the Kr^{85}-line 8508 Å: (a) exposure with mixture of Kr^{85} and Kr^{84}, (b) exposure with pure Kr^{84}

45-day Hg^{203} were prepared from pile irradiated mercury for a beta-spectrometer study by Dr. Wapstra in Amsterdam. The compound HgO was used as charge material in the ion source. In summarizing it may be said that beta-spectrometer samples prepared in the separator are very useful, not only due to the fact that the isotopic purity is high and the mass assignment unique but also because the isotopes are deposited in a well defined layer, and, if necessary, in thin foils.

Although, as described above, most work has been concentrated on the separation of radioactive isotopes, several samples of stable isotopes were prepared. Targets of Mg^{25} were used for studying the radiation from the 6.6-sec positron emitter Al^{26m} produced in the Mg^{25} (p, γ) reaction.[12] Furthermore, targets of Mg^{24}—collected on various metals—were used to study the penetration of the impinging ions into the collector material as well as the concentration of the embedded atoms,[13] by using the γ-rays from the 224-kev proton capture resonance in Mg^{24} as an indicator, in a way similar to that used earlier at the Institute for determining the stopping power of thin foils.[14] It may be added that information relating to the problems called radiation damage in pile technology may eventually be obtained from such studies with the separator, since the energy of the impinging ions is of the same order of magnitude as that imparted to the atoms inside, e.g., a metal by collision with fast neutrons.

In the separation of isotopes having half-lives not much less than one hour, the active starting material may conveniently be transferred from the place of irradiation and introduced to the separator; similarly the separated sample may easily be removed from the place of collection for insertion in the various beta and gamma spectrometers for further investigation. This procedure was used for example in the studies of the long-lived isotopes of krypton produced in fission.[15, 18, 19] However, in order to extend these investigations to more short-lived isotopes, the active gas was pumped through a long tube leading from the cyclotron to the ion source.[16] The consequent further development of this procedure would be to place the separator in the immediate neighbourhood of the cyclotron. Since these early experiments[16] a beta spectrometer of the lens type was built at the Institute, which may be directly attached to the separator in such a way that the collector foil of the separator serves as a sample for the beta-spectrometer. It is hoped that the use of such technical improvements will result in additional information about the radiation from even the very short-lived isotopes of krypton. In case of other short-lived isotopes one may think of producing the activity right in the separator itself, i.e., by letting an external cyclotron beam hit a target located inside the ion source. The development of this technique, which would permit a more detailed study of the radiation from an additional number of short-lived isotopes is under consideration. Especially in the case of spallation studies, it would be favourable to separate the manifold of activities produced. One is led to envisage the use of other instruments, such as linear accelerators, for producing the radioactive isotopes.

There are at present altogether four electromagnetic separators in operation of the general lay-out described here, but it is known that several others are under construction. The separator at the Nobel Institute for Physics in Stockholm has been involved in a great number of important investigations devoted in particular to study of the radioactive isotopes of Kr and Xe.[18, 19] The separator at the Gustav Werner Institute in Upsala was engaged in the separation of spallation products,[20] while the separator at the Physics Department at Chalmers University of Technology was used for various other separation and development problems.[21] Due to the similarity of the design of these instruments as well as of the kind of investigation in which they are involved, the four research teams have been working in close co-operation for several years. The communication of results—of whatever kind they may be—is hereby greatly facilitated, and common plans for the scientific and technical development work have been set up.

As a concluding remark it may be said that isotope separators of the laboratory type described here have proved to be very useful in nuclear studies.

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Record of Proceedings of Session 7.1

THURSDAY MORNING, 11 AUGUST 1955

Chairman: **General Bernardino C. de Mattos (Brazil)**
Scientific Secretaries: **Messrs. L. F. Lamerton, A. Finkelstein and I. D. Rojanski**

PROGRAMME

P/308 Importance of isotopes in technology and industry .... P. C. Aebersold

P/395 Recent developments of radioisotopes used in industry .... H. Seligman

DISCUSSION

The CHAIRMAN: The production of electric power is undoubtedly the most important application of atomic energy. It will, however, take several years before the grids of the world are supplied with electricity of nuclear origin in any considerable quantities.

But certain by-products of atomic energy are already in current use, and their field of application is going to be rapidly extended in the immediate future. They are the artificial radioisotopes to which the present session is devoted.

Natural radioactivity, which was discovered more than fifty years ago by Becquerel, gave us the first medical applications of radioisotopes. The rarity and high cost of radium, however, seriously limited its use.

Artificial radioisotopes now furnish abundant sources of radiation.

We shall see that these can be classified in two categories: (1) fission products representing, so to speak, the ashes of nuclear combustion, and (2) radioisotopes produced by the irradiation of natural elements in a nuclear reactor.

It must be emphasized that this distinction applies only to the method of production and that there is no basic difference between the two kinds of radioactive isotopes.

The papers which are going to be presented will show us the many different applications of artificial radioisotopes: to scientific research, to industry, medicine, biology and agriculture.

We shall be given statistical data showing how fast the use of artificial radioisotopes is increasing in the various countries.

It should be noted that this field of application of atomic energy is not limited to those countries which now have reactors in operation and that we have here one of the first examples of international co-operation in the peaceful uses of atomic energy.

More than fifty countries have already used radioisotopes in the various fields of application.

As the number of research reactors increases, countries now remote from the great production centres will undoubtedly be able to obtain larger deliveries in the future.

Industrial uses cover the whole range of control techniques, from radiography to height or thickness measurement, to long-range detection. Research laboratories are opening up an even wider field of applications of which we will only be able to give a few examples here.

The use of artificial radioisotopes in medicine, biology and agriculture covers a vast field in which progress is so rapid that a complete survey can be made only by experts possessing the most comprehensive knowledge. Progress in all branches of biology owes much to the use of artificial radioisotopes, and the research they have made possible has given us a better understanding of the basic mechanisms of life.

A by no means negligible result of this work has been the bringing together of scientists working in different fields. Such collaboration is particularly indispensable in the study of the dangers inherent in the operation of reactors.

The problems of waste separation, storage and disposal will be studied at greater length towards the end of the session.

As soon as reactors producing electric energy begin to operate, we shall have to deal with large quantities of radioactive fission products. The storage of several hundred kilograms of radioactive isotopes each year is anticipated already.

It appears that several solutions have already been found which are satisfactory from the technical aspect, and it is chiefly their economic aspect which will have to be studied.

It is our hope that it will be possible to make use of a large proportion of the fission products, especially in the chemical industry.

In view of the care with which these problems are now being examined, the nuclear industries may be expected to continue to set a magnificent example of industrial safety.

Mr. AEBERSOLD (USA) presented paper P/308 as follows: Nations need not wait for power reactors before they realize rich rewards from atomic energy. Radioactive isotopes, as reactor by-products, have been distributed extensively for the past nine years and their use has grown rapidly. Thousands of users in over 50 countries have applied isotopes in science, agriculture, medicine and industry. Although they have already brought rich dividends, they are still far from attaining their vast worldwide potential for benefiting mankind.

Although most radioactive isotopes can be produced by other means, the reactor and the fission reaction itself produces them in such enormous quantities that their availability is incomparably greater since the dawn of the atomic era.

Radioactive hydrogen, tritium, is now sold by the United States Atomic Energy Commission for less than one cent per millicurie. When tritium was available from accelerators only, it cost around $10,000 per millicurie. Similar reductions in price or increases in availability apply to most other useful isotopes, though a few are not able to be produced in the reactor.

Perhaps the most important class of uses is in fundamental research. For this session however I shall restrict my remarks largely to applications of isotopes in technology and industry, with brief comments on uses in science, agriculture and medicine which have universal application.

In medicine, and the related pharmaceutical industry, radioisotopes are making important contributions. Most important is the great body of knowledge being gained on the biochemistry and physiology of humans and animals, both in health and disease.

Numerous practical applications are being made of radioisotopes in medical diagnosis and treatment. Over 1100 medical institutions in the United States of America are using these materials routinely for diagnosis and treatment. The number of patients who have benefited from radioisotopes as diagnostic tools is now around 500,000, and about 50,000 have received radioisotope therapy. These medical uses will continue to grow.

In medicine we should also see developed general diagnostic use of radioactive drugs and biochemicals, labelled either with radiocarbon or tritium, or both. With low-level counting techniques, the activity needed for labelling is so low that human administration will be perfectly safe, far below permissible limits. Applications of this type are not widespread as yet but they should soon constitute one of the great medical benefits.

Some as yet unrealized applications depend on development of methods of measurement. In our country industrial companies supply most instruments. They carry a great responsibility in that the rate of progress frequently depends on their vision and enterprise. For example, an urgent present need is the provision of a variety of cheap, efficient low-level detection devices. Such a service will eventually be initiated and will considerably increase the usefulness of isotopes.

In agriculture, hundreds of radioisotope tracer uses are being made in important studies of plants and animals. Many studies are concerned with the uptake and utilization of fertilizers and mineral nutrients, both by roots and foliage. Numerous tracer studies are also being made on the action of herbicides, insecticides, and fungicides, on plant diseases and on the basic process of photosynthesis. We can expect these contributions to agriculture to continue to increase.

In industry itself, the wide and important uses mentioned later will increase manifold in volume. Many new uses will also be developed. During the last year, industry in the United States of America effected savings in excess of 100-million dollars through the use of radioisotopes, and it is anticipated that within a few years it may well exceed one billion dollars per year in savings.

Several reasons for the great importance of isotopes should be noted. In the time available I can speak only briefly about each of these.

First, I wish to note the tremendous power of isotopes as tracers. The power of the tracer technique stems from the great extent to which commonly used radioisotopes can be diluted and still be detected.

TABLE I. Sensitivity of Radioisotope Tracing

Isotope	*Approx. carrier-free activity per gram dpm*	*Dilution theoretically detectable*
H^3	2×10^{16}	$\sim 10^{15}$
C^{14}	10^{13}	$\sim 10^{12}$
P^{32}	6×10^{17}	$\sim 10^{17}$

Table 1 shows the dilution theoretically detectable for three important tracer isotopes, H^3, C^{14} and P^{32}. The values are approximate and assume use of undiluted, carrier-free radioisotope and the latest low-level counting methods.

The dilution theoretically detectable ranges from 10^{12} for the long-lived isotope, C^{14}, to 10^{17} for the short-lived P^{32}. The degree of dilution easily detectable in practice is not as great, but still ranges from millions to millions of millions. This power to trace material is incomparably greater than by any other means.

Isotopic labelling techniques can also be unquestionably specific. Techniques of multiple labelling with two or more isotopes provide special means of achieving specificity in tracer experiments. Double or even triple labelling of compounds permits simultaneously tracing different portions of a complex molecule.

Great versatility also is possible in the labelling of materials with isotopes, either by chemical or biological means. Over 1000 labelled compounds are now readily available. Techniques have also been devised for labelling a wide variety of organisms such as blood cells, bacteria, viruses, and higher organisms. Fabricated objects, such as piston rings and machine tools can be activated directly by irradiation in the reactor. In total, a very large number of compounds, materials and organisms have been made radioactive for tracer studies.

The radiotracer technique thus has the combined power of tremendous sensitivity, unquestioned specificity and wide versatility. This great extension of man's power of observation may make isotopes as routinely and widely necessary as a microscope has become today.

In addition to use as tracers, radioisotopes have another broad category of application as versatile sources of radiation. As sources of alpha, beta, gamma rays, X-rays, and secondary neutrons their range of energies and half-lives covers a great variety of practical uses.

The only reactor-produced alpha-ray emitter now widely used in industry is polonium-210. Neutron sources, obtained by combining polonium and beryllium, are much used for "logging" oil wells and other techniques based on neutron reflection.

Reactor-produced beta-ray emitters are available in large variety. High-energy radiation is frequently desirable while, on the other hand, low-energy beta rays such as those of tritium are safer and more useful for some applications. With the latest low-level counting techniques both carbon-14 and tritium could be traced at very low levels in industrial processes and such utilization could possibly become extensive.

The availability of useful gamma-ray emitters is almost as good—with a lack only of long-lived, low-energy sources suitable for industrial use. Nevertheless, bremsstrahlung radiation, the X-rays produced by beta absorption, can frequently substitute for the needed low-energy gamma rays.

The intensity of gamma-ray sources produced with the reactor is very great indeed. It is routine practice to activate kilocurie sources of Co^{60} for teletherapy units. The activity is in the range of 20 to 60 curies/gram; even 100 curies/gram has been attained. In the latter case, 10 per cent of the stable cobalt was converted to the radioactive Co^{60}. United States reactors produce around 50,000 curies of Co^{60} per year and are scheduled to more than double this rate of production.

Useful long-lived gamma-ray emitters are also obtainable from fission products and a new multi-kilocurie fission product processing plant is under construction at Oak Ridge to be operated in late 1956. This plant will separate and purify thousands of curies of important fission products, and fabricate them into large radiation sources. Over 200,000 curies of cesium-137 per year will be produced, as well as related amounts of other potentially useful fission products.

A summary of the range of radiation sources available from reactors would not be complete without noting the reactors themselves and their coolants and fuel assemblies. Fuel elements removed from the Materials Testing Reactor in Idaho have been arranged to provide an irradiation facility with gamma intensities of about 10,000,000 r/hr.

The importance of isotopes is perhaps best indicated by the rapid and continued growth in utilization. In nine years the number of radioisotope using groups in the United States of America has grown steadily and the total now has reached approximately 5000.

Most striking is the recent growth in industrial users, from about 100 firms in 1950 to over 12 times this number in just five years.

Radioisotope shipments from Oak Ridge National Laboratory alone for nine years total 72,000. Although this laboratory still remains the primary supplier, several national laboratories now distribute some radioisotopes. Government laboratories supply only irradiated materials or simple compounds. Production of the very large number of special pharmaceuticals, fabricated sources, and other preparations containing radioisotopes, has been encouraged as a business for private enterprise. Shipments from private secondary distributors now total more than twice those from the national laboratories, thus making the total number of shipments reaching users over 35,000 per year.

The total activity of isotopes distributed in the first year of distribution—1947—was only 65 curies, whereas last year it was over 40,000 curies. The activity per shipment has also increased remarkably—from an average of just 30 mc the first year to ten times that amount—350 mc—at present, without including Co^{60}. If one includes Co^{60}, the average shipment now is about 2.5 curies.

Another measure of usage is the number of papers published in scientific and technical journals. Publications resulting in nine years through the use of United States-produced isotopes number over 11,000. An Eight Year Summary of United States Isotope Distribution and Utilization, containing over 7000 literature references, is one of the information volumes being furnished by the United States delegation to other delegations and may be reviewed in our Technical Library Exhibit here.

The United States has made isotopes available to other countries since September 1947. This was actually one of the first instances of "atoms-for-peace." Shipments have been made to over 660 institutions in 46 countries. Although international needs for radioisotopes are now usually met by reactors closer to the user, the United States continues to meet special needs.

If the above statistics on use were accumulated for reactor laboratories of all countries, the figures would be most impressive. They will become even more impressive as the worldwide use of reactors expands throughout the world.

In this first paper of the Conference dealing specifically with isotopes I will not attempt detailed illustrations of use but wish to outline the main principles of use. The tremendous variety of uses of radioisotopes stems from three very versatile basic principles which are applicable to radiation as seen in Table 2 below.

TABLE 2. Basic Principles of Radioisotope Utilization

Radiation affects materials	*Materials affect radiation*	*Radiation traces materials*
Food and drug sterilization	Clinical radiography	Metabolic studies
Static electricity elimination	Thickness (density) gauging	Fluid flow
Teletherapy	Industrial radiography	Abrasion and wear measurement
Radiation genetics	Liquid level and height indication	Study of reaction mechanisms

As may be seen, the first is where the radiation affects the material, the second is where the material affects radiation, and the third is where radiation is used to trace the material. I shall illustrate the first of these principles in Table 3.

TABLE 3. Radioisotope Radiations Used to Affect Materials

Chemical effects	*Biological effects*
Polymerization	Tissue destruction
Cross linking	Genetic changes
Induction of reactions	Food and drug sterilization
Electrical effects	*Emissive effects*
Static elimination	Scintillation for detection
Voltage production	Light production in phosphors
Pre-ionization for discharges	Activation of photovoltaic cells

Table 3 illustrates the use of radiation to affect materials. It is the least exploited at present, but is expected to play a large role in the future. Although radiation can have many different effects on materials, these effects all result from ionization or excitation of atoms or molecules.

Thus we find the radiation from radioisotopes applied to produce ionization of air in static eliminators, excitation of phosphorescent light sources, destruction of bacteria in foods and drugs, activation of chemical reactions, polymerization of organics, alteration of material strength and conductivity, gene changes, in addition to various therapeutic results in patients.

TABLE 4. Radioisotope Radiations Used to Gain Information on Materials (Penetration or Reflection)

Gauging	*Detection*
Thickness	Fluid levels
Density	Package contents
Composition	Assembly components
Radiography	
Welds and castings	
Clinical diagnosis	

In the second principle of use, illustrated in Table 4, radiation is directed at or through a material to gain information about the material. Thickness of a material, its density, atomic number and atomic structure, have different effects on radiation. Therefore much can be learned regarding these properties by observing their effects on absorbing or reflecting radiations.

The major applications of this principle are industrial and clinical radiography, thickness and density gauging, and analysis by radiation penetration. Other important uses include liquid level gauging and other applications where an object is revealed by its effect on a beam of radiation, as in industrial sorting and packaging operations.

TABLE 5. Radioisotope Radiations Used to Trace Materials

Bulk transfers	*Atomic and molecular transfers*
Fluid flow	Diffusion
Leaks	Vapor pressure
Wear and corrosion	Chemical reaction mechanisms
Hidden, moving objects	Complex chemical pathways
Analytical techniques	
Assays	
Volume and quantity determinations	
Neutron activation analyses	

In applications based on the third principle, illustrated in Table 5, radiation traces the material. The radioisotope is incorporated in or carried by the substance of interest. This substance can then be located or traced, or parts of it measured quantitatively. Radiomaterial can be traced in bulk, as in locating markers, determining fluid flow, following motion of material, detecting leaks and measuring wear and abrasion. Also, radioisotopes as tracer atoms are used to study all kinds of chemical, physical-chemical and biochemical problems. In technology, tracer uses include studies of corrosion, diffusion, detergency, catalysis, and kinetics and mechanisms of chemical reactions. Further application of these versatile principles to industrial use will be limited only by human ingenuity.

The total economic value of isotope utilization can be measured by success in (1) saving time of personnel in research, (2) increased speed in acquiring knowledge, (3) saving materials and labour in manufacturing, (4) improving performance of manufactured products, (5) increasing agricultural productivity, (6) decreasing losses from food spoilage, and (7) improving the general state of health of the public.

The sooner problems are solved, the sooner benefits can be realized. A good example is the great saving in studies of all kinds of wear and corrosion. With tracer techniques, extremely small amounts of wear or corrosion are measured accurately and the rate of obtaining results is speeded up by a factor of 10 or more. One group, studying improvement of lubrication, obtained results with tracer techniques in 4 years at a cost of $30,000 which would have required 60 years by previous methods and cost $1,000,000.

Ultimate savings in wear, from introducing improved lubricants so many years earlier, will far exceed the important savings in cost of research. Radioisotope techniques have already been applied to a host of wear studies such as wear of engine parts, rubber tires, and furnace linings. Since wear and corrosion of all kinds cost the world billions of dollars per year, the increased rate of finding ways to reduce this type of loss can result in enormous savings.

Millions of dollars are saved each year in the United States of America by the use of radioisotopes in radiography. Around 500 firms are using this principle. Other millions of dollars are saved by the hundreds of firms using various types of radioactive thickness gauges in product control.

Time does not permit illustrations of the economic value of isotopes in other applications, but as already indicated, industry in the United States of America alone now saves, from all applications of radioisotopes, over $100-million annually and these savings will grow rapidly.

In closing, I should like to remark on the over-all significance of isotopes in the development of atomic energy.

For nations undertaking a programme of atomic energy development, the use of radioisotopes is doubly rewarding. It not only provides many direct benefits, but helps develop a large source of professional and technical persons familiar with radioactivity. Since radioactivity is a uniquely important occupational factor of the atomic energy industry, an increasing number of persons trained with radioisotopes both precedes and permits an expanding atomic programme.

Fostering radioisotope uses will not only establish "seeds" from which interest in atomic energy will grow, but will create future use for the abundant amounts of radiation and radioisotopes which many nations will have as by-products of reactors in the near future.

Although world-wide need for propulsive and industrial power is the main interest of most countries in developing atomic energy, the by-product radioisotopes should not be overlooked as of equal importance in the long run to the over-all health and welfare of nations.

Mr. Seligman (UK) presented paper P/395.

DISCUSSION OF PAPERS P/308 AND P/395

Mr. W. F. Libby (USA): What would be your estimate of the total annual savings due to isotopes in British industry at the present time?

Mr. Seligman (UK): Well, I tried in fact to find this out myself and this is a very difficult thing to do. If I talk about economics it is very similar to an economist talking about science. In fact you heard figures from Dr. Aebersold which are the best figures available, I think, for America. I would say in England they are slightly on a different line. Perhaps not so much is achieved on saving in materials, in thickness gauging and so on; but by, for instance, the Thames mud experiment, the Comet crashes and this oil-brine interface business we can perhaps save several millions of pounds at one time. It is a very difficult figure to assess but my guess, if I may make a guess, is that it is several million pounds per year—but it is a very very rough guess.

Mr. Guéron (France): The speakers before me have set out the general principles, with some specific examples of the use of radioelements, and have assessed the benefits which that use may bring. I should like to indicate briefly how these elements are used in France (see P/330 and P/331).

Mr. Samarin (USSR): In connection with the very interesting papers presented by Mr. Aebersold and Mr. Seligman, I should like to introduce the following comments.

The use in industry of the method of continuous casting of steel has acquired great importance at the present time. The application of radioisotopes in determining metal hardening rates to which Mr. Seligman referred appears to me to be of particular interest.

Studies have been made in the USSR using radioactive isotopes to determine the hardening rates of steels of different compositions in both continuous and conventional casting. It should be noted that certain problems connected with this stage of the production process, i.e., with casting, could be solved only with the help of radioactive isotopes. Radioactive isotopes are widely used in various branches of industry. In the last two years, more than 200 papers have been published in the USSR, dealing with industrial research in which radioisotopes were used. A number of these papers will be reviewed in the course of this conference.

I hope that a wider exchange of experience in the future between workers in this field will contribute to the advancement of industry in all countries.

Mr. Libby (USA): I should like to ask Mr. Seligman whether tritium is receiving any appreciable industrial or medical use in Great Britain at the present time.

Mr. Seligman (UK): In industry, tritium is being used increasingly in England. For example, it is used for the purpose of filling cold cathode ray tubes so that they can start more easily. Another application which we envisaged was for checking underground connections in waterways using tritiated water because it is not absorbed in the soil as are all the other chemicals. I am not qualified to speak with respect to medicine. You should really ask a medical expert.

Mr. E. **Saeland** (Norway): I should like to ask Mr. Seligman a question about the interesting apparatus he described for measuring the corrosion by reflected gamma rays. Mr. Seligman mentioned an accuracy of 4 per cent for these measurements. It would be interesting to know what is the minimum area of the corroded spot which can be determined with such an accuracy.

Mr. **Seligman** (UK): The accuracy of 4 per cent is for steel up to three-quarters of an inch. I do not know offhand the minimum area. However, Mr. Putman, who did the experiments, is here and I am sure he can answer the question.

Mr. J. L. **Putman** (UK): Almost all the entire backscattering came from an area of about four centimetres in diameter. We tested this by changing the area steadily and found that it made not very much difference after the diameter of a flat sheet of metal of the thickness of about six millimetres was increased beyond about four centimetres. We did not go in very great detail into the specificity of the measurements of the limits of size of pit which could be detected on the inside of a tube. We did do one experiment, however, in which a three millimetre diameter hole, three millimetres deep, was bored on the inside of a tube which was six millimetres thick. We found that we could detect this quite easily.

Session 19C

GENERAL USES, PRODUCTION AND HANDLING OF RADIOACTIVE ISOTOPES

LIST OF PAPERS

The Use of Radioisotopes in Australia

By C. E. Eddy,* Australia

Australia has as yet no local source of supply of radioisotopes, so that all requirements must be obtained from the United Kingdom, the United States of America, or Canada. The great distance from these sources of supply has limited severely the range of isotopes which can be utilized in Australia to those with a half-life of at least a few days, and even then relatively expensive air transport is necessary in a large proportion of shipments. Notwithstanding these limitations, a steadily increasing use is being made of radioisotopes in medicine, industry and research. A general account of the methods which have been adopted on a national level to enable local workers to gain experience in the techniques of radioisotopes, and to use them in investigation and in routine applications, may be of interest to countries similarly situated.

ARRANGEMENTS FOR THE IMPORTATION OF ISOTOPES

The United States Atomic Energy Commission, in announcing in September, 1947, its arrangements for the export of radioisotopes to other countries, laid down certain conditions to ensure that valuable material in short supply was used to the best advantage, that users were safeguarded against health hazards, and that the results of scientific investigations with isotopes were made available as widely as possible to other workers. Because of its experience in radiological physics and the problems of protection against radiation, the X-ray and Radium Laboratory of the Commonwealth Department of Health was nominated as the authorizing, ordering, and distributing center for isotopes in Australia. Later, when Atomic Energy of Canada, Ltd., made isotopes available to users in other countries, a similar arrangement was necessary, and the Commonwealth X-ray and Radium Laboratory was again nominated as authorizing agent.

LOCAL ARRANGEMENTS FOR CONTROL OF USE OF ISOTOPES

The Atomic Energy Research Establishments in Great Britain did not require specifically that such organizations should be set up in importing countries, but the attention of intending users was drawn to the existence of such an organization in any country where one had been established.

To control effectively all importations into Australia and all use thereafter, the Commonwealth Government included radioisotopes in the Schedule of Prohibited Imports under Regulations of the Department of Trade and Customs, approval for entry of such imports being the responsibility of the Director-General of Health. The principal criteria for approval is that the isotope should be utilized safely and usefully.

In practice, this system operates quite satisfactorily. Intending users of radioisotopes approach the Laboratory for approval, discuss their proposals and requirements, and receive full information regarding suitable materials available, the specific hazards arising in their use, the types of monitoring and measuring equipment which would be required, and provisions for the safe disposal of radioactive wastes. In many cases, procedures are developed in association with the user, with members of the Laboratory staff visiting the site of operations. The provision of such an information and advisory service has been much appreciated.

For several years, a unit of the Commonwealth Scientific and Industrial Research Organization was located in the Laboratory to provide assistance in regard to radioisotopes to workers in that Organization and to research workers generally. On occasions, particularly when suitably labelled compounds were not otherwise available, this unit assisted prospective users by synthesising specially labelled compounds. C.S.I.R.O. also provided assistance in the actual clerical work involved in ordering isotopes, and through their Scientific Liaison Offices, in obtaining technical information from overseas suppliers. Since it was found, however, that C.S.I.R.O. used only a small proportion of the isotopes being imported, the whole of the activities concerned with isotopes are now the responsibility of the Department of Health.

All isotopes intended for use on human beings come under the general supervision of the Committee on Radioisotopes of the National Health and Medical Research Council. This Committee was appointed in 1947 to co-ordinate research in the therapeutic use of radioisotopes as well as in their use in tracer studies in humans, and to circulate material and literature dealing with these subjects. In particular, the Committee has been responsible for ensuring that full details of all clinical results of treatment are compiled and made available to other interested institutions. The

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* Commonwealth Department of Health.

Committee has been responsible for determining a policy of treatment for the different isotopes. When this has been decided, approval for the treatment of individual patients has been passed on to Therapeutic Trials Committees in the different States.

The rapid extension of the use of isotopes in industry, and the possibility of local production in the future has led the Health Departments of the Commonwealth and the various States to decide that legislative control should be introduced. Uniform legislation covering the use of both X-rays and radioactive substances has therefore been prepared, and has already been adopted in some States. This legislation is not intended to be unduly restrictive, and should enable the use of radioisotopes to be extended to all who are suitably qualified and experienced while still ensuring adequate protection of personnel and satisfactory disposal of contaminated wastes.

USE BEING MADE OF ISOTOPES

Isotopes, principally in tracer amounts and produced by cyclotrons, were obtained first in 1946. With the availability of pile-produced isotopes late in 1947, regular shipments of isotopes of different types have been obtained. Initially, the greatest interest was in those required for medical diagnosis and treatment, but in recent years the use in industrial applications has increased considerably.

Use in Medical Diagnosis and Treatment

Isotopes most in demand for medical diagnosis include: (1) iodine-131 for the assessment of thyroid dysfunction; (2) iron-55, iron-59, and chromium-51 for the investigation of blood diseases and abnormalities; (3) compounds labelled with iodine-131 and phosphorus-32 for the location of tumours.

Those most in demand for treatment include: (1) phosphorus-32 for the treatment of polycythemia vera and certain leukemias; (2) iodine-131 for the treatment of thyrotoxicosis and some forms of thyroid carcinoma; (3) gold-198 for the treatment of cancer in different sites; (4) strontium-90 and phosphorus-32 as external applications for the treatment of conditions of the eye and skin; (5) cobalt-60, in the form of needles and tubes for the treatment of malignant disease.

Because of the short half-lives of some of these [for example, radiogold (2.7 days), radioiodine (8.0 days), and radiophosphorus (14.3 days)], delivery must be made from overseas by air freight. The great majority of the isotopes required are obtained from the United Kingdom, in which case the transport time (4 days) is also the shortest. Although the suppliers there have developed very efficient lead-lined transport boxes, the freight is in many cases large compared with the cost of the isotope itself.

It has been found to be more economical in freight charges to obtain bulk supplies of isotopes in regular demand, dispense these in the Laboratory in the doses required for the treatment of individual patients, and then distribute them to hospitals in Australia which may still be up to 2000 miles from the Laboratory. Applications for tracer or treatment doses for individual patients are sent to the Laboratory, with an indication of a suitable date of administration. These doses are then allotted from supplies arriving, and the user notified of exact date of arrival. Doses are then dispensed to have the required content at the time and date of administration, and despatched usually by air freight. Individual hospitals are thereby saved much trouble in standardising doses. If a waiting list of patients tends to accumulate, additional supplies to the regular deliveries are obtained from overseas. With isotopes such as radiophosphorus and radioiodine, it is possible in this way to provide doses for patients with a waiting time not usually exceeding a fortnight, and to ensure that only about 15 per cent of the activity on arrival in Australia is lost in decay before use.

The organization of such a service requires care and attention to detail. The Laboratory has had, however, many years' experience with the preparation and distribution of radon (half-life 3.8 days) for use in hospitals hundreds of miles away. This experience in arranging rosters of patients and transport facilities proved of value when the problem of distributing imported isotopes arose.

All isotopes used in diagnosis or treatment are issued free of charge, the costs being borne by National Health Funds.

Figures showing the use made of isotopes for treatment purposes since the beginning of 1950 are shown in Table I. No great annual increase in this time has occurred in the use of radiophosphorus; this is due probably to the fact that the techniques of using radiophosphorus in treatment have been stabilized for some years, and the number of patients presenting annually is nearly constant. An increasing use is being made of radioiodine, as the methods of treatment are developed and their value assessed.

TABLE I. Isotopes Used in Medical Diagnosis and Treatment, 1950–55

Isotope	*Shipments received**	*Millicuries*	*Doses distributed*
Iodine-131	145	7350	2500
Phosphorus-32	109	3710	1135
Gold-198	6	1130	8

* In addition, 425 mc of cobalt-60, in the form of tubes and needles, 60 mc of strontium-90 in superficial applicators, and 10 mc of iron-59 for diagnostic studies, have been received.

The doses of radioiodine may be of the order of 20 microcuries for assessment of thyroid function, of 6 millicuries for the treatment of thyrotoxicosis, or occasionally of 50 to 100 millicuries for the treatment of metastases from thyroid carcinoma. Of the 2500 doses which have been issued in nearly 6 years, just over 1000 have been made in the last 10 months. This increase has been due to the adoption by several large hospitals of radioiodine as the treatment of choice in diffuse toxic goitre, and further increases can be expected.

The use of radiogold in treatment has only recently been introduced in Australia, and then only on an experimental basis.

Use of Isotopes in Industry

The use being made of isotopes by industry to a great extent depends upon the degree of industrialization in a country, and upon the need for improved methods in development and in production control. Isotopes can be used in industry in industrial radiography, in equipment for continuously measuring the thickness of rolled strips of various materials, in the elimination of static electricity particularly in the textile industry, in various types of inspection and package monitors, in tracing the flow of liquids through pipes, and in a number of tracer studies in chemistry and metallurgy. Intense sources of radiation from isotopes are also being used to assist in chemical processes, in the production of plastics, and in the sterilization of canned goods and biological products.

Prior to the availability of radioisotopes, X-rays and the gamma rays from radium and radon were used to an appreciable extent in industrial radiography in Australia. Certain isotopes are of particular value in this field, providing intense, reasonably cheap, and readily portable sources of radiation with a range of penetrating power. The construction in Australia during the last few years of a number of oil refineries, hydro-electric stations, oil and gas pipe lines, and welded ships has led to a sudden increase in the demand for different isotopes for the radiographic examination of welds.

The principal isotopes which have been used for industrial radiography are shown in Table II, together with the respective half-life, the energy of the gamma radiation emitted, the total number of sources imported in the period 1950–55, and the total number of curies.

TABLE II. Isotopes Used in Industrial Radiography, 1950–55

Isotope	*Half-life*	*Gamma-ray energy, Mev*	*No. of sources*	*Total curies*
Cobalt -60	5.3 yr	1.17–1.33	24	9.1
Caesium-137	33 yr	0.66	3	10.8
Barium-137	2.6 min			
Iridium-198	74 d	0.30–0.61	14	70.0

Sources for industrial radiography are selected to supply gamma rays of an energy suitable for the thickness of the material being investigated. Cobalt-60, with a reasonable half-life, is satisfactory for sections of steel from 2 to 6 inches in thickness. For thicknesses of from 0.5 to 2.5 inches, a less energetic gamma radiation is desirable. Originally, only iridium-192 was available in this range, with a half-life of only 74 days. It was therefore necessary to maintain a shuttle service, the partly spent sources being returned for re-activation. More recently, caesium-137 (which acts as a parent for the short-lived barium-137 which emits the required gamma rays) with a half-life of 33 years has become available, and is expected to replace iridium, at any rate in countries at a distance from the source of supply.

The isotopes used in equipment for thickness gauging, package monitoring, and static elimination are usually alpha or beta ray emitters, with relatively intense beams of easily absorbed radiation. Special attention is given to the instruction of personnel operating these units as to methods of avoiding over-exposure, and to the methods of disposal of decayed or damaged sources.

Appreciable use has been made of radioactive sources in "tagging" go-devils or pigs forced along pipe lines to clean the interior, or to mark the interface of different batches of petroleum products. Cobalt-60 is generally used, the source strength being selected to be adequate to enable the position of the pig along the pipe to be detected with sensitive instruments, even at a depth of some feet in the earth. Operating personnel are instructed in the precautions which must be taken to avoid radiation exposure during insertion and removal of the pig, or when recovering it after it has become stuck in the pipe.

Use of Isotopes in Research

Isotopes are used in research projects generally as tagged atoms in the investigation of chemical, metallurgical, biochemical and biological processes. The range of available isotopes is large, and the amounts of material required are generally very small. As many of the investigations are carried out in non-physical institutions, requests are frequently received for assistance in planning the isotope side of the work, and for advice on the type of counter to be used, and on the amounts of material required. In many cases, actual procedures for counting samples are recommended, and standard sources of a suitable radioactive material supplied to enable the constancy of the counting equipment to be checked.

The isotopes which have so far been used in re-

TABLE III. Isotopes Used in Research, 1950–55

Isotope	*Shipments*	*Isotope*	*Shipments*
Antimony-125	2	Phosphorus-32	12
Barium-140	1	Polonium-210	3
Caesium-137	3	Rubidium-86	2
Carbon-14	66	Ruthenium-106	2
Cerium-144	3	Scandium-46	2
Chlorine-36	3	Silver-110	1
Chromium-51	1	Sodium-22	6
Cobalt-56	1	Strontium-89	1
Cobalt-60	8	Strontium-90	7
Europium-154	1	Sulphur-35	7
Hafnium-181	8	Tin-113	2
Iron-55	4	Thorium-228	3
Iron-59	6	Thulium-170	2
Krypton-85	1	Tritium-3	4
Lead-210	1	Zinc-65	4
Neodymium-147	1		

search are listed in Table III. This shows that 168 shipments of 31 different isotopes have been employed in the last six years. In some cases, particularly with long-lived isotopes, one shipment of the minimum amount available has been divided for use in a number of institutions. The samples of carbon-14 have included a wide range of organic and inorganic compounds labelled in different ways.

The number and range of isotopes used in research can be expected to increase, particularly as a result of the training of chemists and others in the techniques and possibilities of radioactive tracer study in short courses being conducted this year by the Universities of Melbourne and Sydney, and by the N.S.W. University of Technology.

General

It will be evident that, even in a country far distant from sources of supply, opportunities for training in the use of radioisotopes, and for their utilization in medicine, industry, and research are still possible. In this connection, the manner in which the suppliers of isotopes have co-operated by making available full technical information and much useful advice should be recorded.

The existence of a local source of supply of isotopes would admittedly lead to the use of several elements which are important in tracer studies in biochemistry and biology and which have too short a life to permit their being obtained readily from overseas. These would include the following, for which some requests have been received: silicon-31 (2.8 hours); potassium-42 (12.4 hours); sodium-24 (15 hours); and bromine-82 (35 hours).

In addition, a local source of supply would undoubtedly increase the general interest in, and extend the range of, applications in which radioisotopes would be employed.

The Use of Radioisotopes in Czechoslovakia

By Chestmir Shimane, Czechoslovakia

This report will serve briefly to review the development of the use of radioisotopes in Czechoslovakia during the past four years.

The first radioisotopes, used in a number of scientific research and medical institutes, were those of phosphorus, iodine and sodium. As in any other branch of science which is just beginning to develop, this was predominantly meant for general orientation, work aimed at mastering the methodology of the use of the radioisotopes and the techniques of measurement. In those places where the necessary prerequisites for independent mastery of this work had not yet been achieved, the physics faculties of several institutes of higher learning and the physics institutes of the Academy of Sciences, along with several industrial research institutes, sponsored work with radioisotopes. In this way it was possible, from the very beginning, to create a great deal of interest in the use of radioisotopes in the most diverse areas, in fact, at approximately 15 different sites.

I want to touch on the work which was done during this period, mainly for the purpose of indicating the broad group of individuals who became interested in the use of radioisotopes from the very beginning of this work.

In the first place, medical institutes became interested in the use of radioactive phosphorus; they were concerned mainly with the therapeutic applications of the nuclide, for which no special instruments are needed. Radioactive phosphorus was used for polycythemia and leukemia. The results were so good that the initial interest in the use of radioactive phosphorus for these purposes immediately proved justified, and led to its consistent use.

Along with phosphorus, radioactive iodine also rapidly came into use, although mastery of the technique of working with this radioisotope for the diagnosis of thyroid disorders, and the analysis of the results, required somewhat more time.

Radioactive sodium was used during this period for exploratory experiments pertaining to the measurement of blood circulation time, and the diffusion of sodium io muscle tissue.

Furthermore, these three radioisotopes also were used in oncology.

Besides these basic medical applications, to which attention was given from the very beginning of the use of radioisotopes in Czechoslovakia, we should also mention the use of radioactive phosphorus in metallurgy (where the distribution of phosphorus in ingots was investigated by autoradiograms), the use of radioactive iodine in the histology of the invertebrates, the use of radioisotopes in the physiology of plants, and finally uses in dosimetry and in nuclear physics.

Since then, the use of radioisotopes has increased considerably, their variety has increased, as well as the number of chemical compounds in which they are used. As an illustration, I will indicate that more than 20 radioisotopes are imported into Czechoslovakia. The number of chemical compounds tagged with radioactive atoms has increased even more. Work with radioisotopes now is progressing in several dozen places.

Although the early use of radioisotopes was concentrated on medicine, and they were just beginning to be used in other fields, the center of gravity, in the pattern of use of the radioisotopes, is shifting to industry and other fields.

I will now attempt to outline the present status of the principal applications of radioisotopes which have been carried out in Czechoslovakia during recent years.

The use of radioactive phosphorus for the treatment of polycythemia and leukemia has become a clinical method. In the opinion of the physicians using it, radioactive phosphorus represents a very valuable contribution to the cure of polycythemia. Its use in the National Faculty Hospital in Prague, in Pilsen and Olomouc, on about 50 patients, had a positive effect in the majority of cases. In comparison with X-ray therapy, treatment with radioactive phosphorus is more advantageous, has a more lasting effect, does not require such frequent repetition, and is considerably more pleasant for the patient. As far as the use of radioactive phosphorus for leukemia is concerned, the treatment is not considered to be especially advantageous as compared to X-ray therapy. In lymphoid leukemia, however, the application of small doses of radioactive phosphorus to maintain the patient's condition has been justified. The average total number of leukemia patients being treated with radioactive phosphorus is approximately 30.

With the help of radioactive phosphorus, several questions having to do with the total volume of blood circulation in certain diseases of the blood were solved. The use of radioactive phosphorus for determining the total quantity of blood was compared

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Original language: Russian.

with the Evans method in a total of 111 cases made up of patients with various blood diseases.

A second basic radioisotope which is frequently used in Czechoslovakia for therapeutic, as well as diagnostic, purposes is radioactive iodine. For the diagnosis of thyroid disorders, a diagnostic method is being perfected. As a result of this work, a modified method using fixation and excretion curves emerged, as well as a new one, based on the measurement of thyroid and renal clearances. Their ratio serves to determine the degree and nature of the disturbance in thyroid function. The diagnostic investigations of thyroid disturbances are conducted in the Endocrinological Institute in Prague, where patients are grouped for diagnosis and treatment. Up to the present time, the number of diagnostic applications of radioactive iodine is about 2000. In many of these patients, the diagnostic procedure was repeated several times.

Several hundred patients are in the process of receiving therapy for hyperthyroidism using radioactive iodine; the therapy is presently completed for about 100 patients. Consecutive as well as single doses are used. Considerable success has been achieved using both methods. The number of cases of myxedema was reduced to 2–3% (as far as is possible to judge from available data).

Cancer of the thyroid was treated in 15 patients. All cases were in an advanced stage of the disease and were not operable. So far, the patients' lives have been extended by two years.

The patients who are to receive radioactive iodine therapy are strictly selected in accordance with fixation curves and with the aid of supplementary differential accumulation curves supplemented by γ-radiography.

Up to 400 millicuries of radioisotopes per year are used for each patient.

An interesting application of radioactive iodine was the successful cure of cancer of the larynx where the thyroid gland, containing an accumulation of radioactive iodine, acted as a therapeutic *moulage.* This method of treatment was recommended by Prof. Joliot-Curie. The treatment was verified in the Endocrinological Institute in Prague.

When a radioisotope of iodine is used in large therapeutic doses, the problem of recovering it from the patient's urine becomes of interest. On the one hand, this reduces the consumption of the radioisotope, and on the other hand, it limits the accumulation of waste radioactivity. In the Endocrinological Institute, which is the major user of radioactive iodine, a convenient method of recovery has been worked out.

Other medical applications may be mentioned. For instance, isotopes of iodine, sulfur, phosphorus and sodium have been used for investigating the dynamics of metabolism of shock in the living organism, from the standpoint of possible practical applications in surgery. These investigations gave valuable data on the possibility of introducing drugs into the organism under such conditions. A whole series of radioisotopes has been used in oncology for the treatment of malignant tumors. A method for localizing brain tumors with radioactive iodine has been worked out. Using radioactive iron, a method has been developed for determining the degree of iron absorption at the gastric level in various types of anemia; as well as a method for determining iron in the blood serum, and the use of radioactive iron for checking the rate of blood circulation. With the help of radioactive phosphorus the metabolism of the nucleic acids is being investigated.

The Institute of Plant Physiology of the biology department of Karl University in Prague has carried on interesting work in the field of plant physiology. The absorption of sprinkled liquids by the leaves of the higher plants was investigated with the help of radioisotopes of phosphorus and sulphur. These experiments were carried out in connection with others dealing with the feeding of plants by means of additives dissolved in the sprinkling liquid rather than via the roots. It was found that the additives enter the leaves and then move on into other parts of plants in measurable and sometimes substantial quantities, thus affecting the metabolism of the plant. The work is aimed at a solution of questions connected with increasing the yield of plants.

In the field of national water resources, the Research Institute of Water Reserves has used radioactive phosphorus experimentally to measure the velocity of rivers in order to ascertain whether this method has advantages over the use of the pigments. To date, the experiments carried out indicate that, in this application, the advantage of using isotopes is not great; on the contrary, applying them and measuring is more complicated than using pigments. However, since this method may be made more sensitive than the pigment one, it is possible that the use of the radioisotopes is more advantageous with large distances or for the measurements of rivers passing through water reservoirs.

Another example of the use of radioisotopes in water resources is a method for measuring the water equivalent of snow. This method is based on the well-known principle of measuring thickness by means of γ-ray absorption. Radioactive cobalt serves as the radiation source. The equipment includes means for remote reading, thus permitting its installation to be made in inaccessible locations. Results of measurements to date are very encouraging, and they are better than those of present methods which suffer from individual sampling errors. This method is very important for Czechoslovakia's water economy. If it becomes possible to obtain accurate information on potential water supplies in the form of snow in the watersheds of large rivers, then it will be possible to predetermine the levels in reservoirs so as to permit them to accept all of the melted snow. At the same time, it will not be necessary to empty the reservoirs unnecessarily; this will prevent the development of an undesirable power generating situation when it is desirable to have maximum possible amounts of water in the reservoirs.

The third application of radioactive isotopes in national water resources is still being developed. It depends on the use of neutrons for measuring fluctuations in the condition of deep waters. This problem has been partially solved by work on a neutron probe for measuring the moisture content of soil, reported separately. To date, in Czechoslovakia, only gamma emitters plus beryllium have been used as a source of neutrons; however, the possibility is being investigated of perfecting a scintillation counter for slow neutrons insensitive to γ-rays: this will permit the use of photoneutrons from artificial radioactive isotope sources, and consequently the manufacture of neutron probes in large quantities.

Let us now look into machinery manufacture, where to date the principle use of radioactive isotopes has been of cobalt and iridium in flaw detection. The first use of both isotopes dates back to 1953. Since then, the number of preparations used in radiography has increased several-fold; the range of γ-radiography has also increased significantly.

Radioactive iridium has been used constantly for inspecting seams and welds up to 40 mm thick, for illuminating boiler tube seams, and for inspection of thin-wall castings; radioactive cobalt has been applied to similar uses for thicknesses larger than 40 mm. Cobalt sources are used up to several hundred millicuries; those of iridium up to several thousand millicuries.

Use of these radioactive isotopes has meant considerable progress to our industry. While the usual X-ray equipment used in industry permits easy illumination of steel up to 80 mm thick, it cannot also be used exactly where it is needed, such as during assembly. Until radioactive cobalt was introduced for illuminating thick materials, mesothorium preparations had been used; however, they are expensive and were not available in sufficient quantities. Use of radioactive cobalt allows examining steel up to 150 mm thick. Since a considerable number of radioactive cobalt sources are in use in our industry, it is possible to use them not only in manufacturing, but also in assembly. It is now possible to effect quality control of materials and components even in those cases where it was not possible to do so earlier, although there was no doubt as to the desirability for such control.

Radioactive iridium permits inspection of seams of high-pressure pipes and boiler tubes, directly, during installation. For instance, nowadays, all pipe seams are illuminated and inspected during the construction of any electric power station in Czechoslovakia. With large diameter pipes, the source is placed on the axis of the pipe, and the film is placed on the seams from the outside; with pipes less than 40 mm in diameter, the source is placed outside the pipe but not in the plane of the butt, but consecutively in two planes perpendicular to each other. Several thousand seams are inspected during the construction of an electric power station. Just as pipe welds are inspected, so are the welds of steel structures, high-pressure tanks, welded structures in shipbuilding, castings which are subject to high stresses, and component parts of hydroelectric plants.

It has become obvious that regular inspection of welds has an important training effect on welders who are thus enabled to judge their work qualitatively. Regular inspection has led to a significant improvement in the quality of the work of welders, and therefore has resulted in fewer defects and improved safety.

The rather significant amount of experimental information that was obtained during fault inspection using radioactive cobalt and iridium, was used to assemble a display of weld gammagrams. This has been used for evaluating the quality of welds; and welding standards which specify evaluation of welds based on gammagrams are founded in this display. What remains to be established is the correlation between the gammagrams and the mechanical properties of welds.

Attempts to find other applications for radioactive isotopes in the machine building industry, principally having to do with the measurement of thickness without actual contact, and the evaluation of wear of machine parts, etc. are in the laboratory stage, for the time being, and they have not yet found widespread practical application.

Work on the study of the diffusion of carbon in steels during case-hardening is also in the laboratory stage. For this purpose, a nitrogen isotope (short half-life), is used. It is produced directly in the test sample upon irradiation with a stream of protons.

At the end of this report, I wish to draw attention to what made such widespread applications of the isotopes in Czechoslovakia possible. In the first place, it rests on the fact that the Soviet Union has furnished to us the necessary isotopes without any conditions regarding the control of industry, reports on their application, etc. It is further linked with the fact that our industry was able, from the very beginning, to furnish organizations with electronic instrumentation when these organizations were unable to develop it themselves. Here we have in mind mostly counting circuits, Geiger counters and integrators. Invariably, work with radioactive isotopes was begun by leaning on the cooperation of physicists who passed on their experience to other workers.

From the very beginning, the health of people working with radioactive isotopes was guarded by instruction lectures given by our specialists. Work was made safe by a government standard for work with radioactive isotopes and with radiation, which must be observed by all organizations and institutes. Radioactive isotope distribution is centralized for all of Czechoslovakia. This makes it easier to inspect the places of work and to evaluate their suitability for work with radioactive isotopes. This is mandatory, due to the danger inherent in the use of isotopes both to workers and their surroundings.

Interest in applying isotopes in our country increased many-fold after the agreement was signed between the USSR and Czechoslovakia for assistance in the fields of research and peaceful utilization of nuclear energy. To create suitable conditions for this broad new application of radioactive isotopes, the training of new specialists is being organized for high school and college graduates. These specialists will be completely acquainted with the problems of using radioactive isotopes in all fields, and they will assist in its wide dissemination while keeping all safety and health physics rules. In addition, courses and lectures are being prepared for senior specialists of different fields in the application of radioactive isotopes. Health organizations are also preparing to fulfill new demands for the inspection and safeguarding of the health of people working with radioactive isotopes; the number of the latter will grow considerably when Czechoslovakia will itself start manufacturing radioactive isotopes.

The government has been paying considerable attention to spreading the application of radioactive isotopes to all fields of science, medicine, industry and agriculture. One can expect a large increase in their applications, so as to obtain all the results which radioactive isotopes are capable of bringing to human health and the national economy.

Radioisotopes in Industrial Research

By S. E. Eaton,* USA

Radioactive isotopes are proving extremely potent as research tools for accelerating industrial progress. Developed as by-products of a military program, they are now creating new knowledge and understanding —of nature and of industrial processes and products —that will have positive and permanent value to mankind. The purpose of this paper is to discuss the unique and powerful features of radioisotopes which have been found particularly useful for industrial research, and to set forth some typical problems which they are solving in the United States of America.

You already know what radioisotopes are—atoms that give off radiation. They are what remains of the uranium atom after it has split; but they can also be made to order by irradiating ordinary atoms in nuclear reactors or particle accelerators. The radioactivity from massive quantities of radioisotopes can be used to promote chemical reactions, to preserve foods, or to measure or control the thickness of products during manufacture; but such uses are a subject in themselves outside the field of industrial research, and will not be covered in this discussion.

Radioisotopes are also proving tremendously important to agricultural researchers, nuclear physicists, and medical people; but what I would like to talk to you about today is the importance of radioisotopes to us industrialists and our research problems. Whether radioisotopes are going to be a necessary tool in the average company's research tool kit remains to be seen, but for those companies who value a microscope or chemical analysis or physical measurements, radioisotopes are certainly too powerful to be ignored.

Now just why are they so powerful? Why are they the only means of solving some problems? Let us put down four principal points:

1. They are easy to detect.
2. They provide a small, inexpensive, portable source of radiation.
3. They can be measured in minute quantities with high sensitivity.
4. They are specific and can be traced in the presence of other chemically identical atoms. Except for some minor differences in the low-atomic-weight elements, radioactive isotopes are identical in chemical behavior to nonradioactive atoms of the same element. Thus specific batches of tagged or radioactive atoms from one source can be differentiated in the presence of other chemically identical atoms from a different source. Radioactivity is unaffected by temperature, pressure, or chemical composition.

There are other advantages, but I think these cover the principal points. Now what are these advantages good for in industrial research? The fact that radioisotopes are easy to measure can simplify identification of the constituents of a sample by activation analysis.[1] Irradiation in a reactor or accelerator can make the sample radioactive in a manner characteristic of its constituents. A knowledge of these characteristics then allows interpretation of the sample's radioactivity in terms of its composition. The method is proving of particular value where a nondestructive test for traces of metallic impurities is desired.

Ease of measurement also makes it simpler to determine optimum mixing time for such materials as paint, ink, plastic products, powders for powder metallurgy, and the like. One of the components can be tagged with radioactivity, either by simple addition of a physical tracer, or synthesis of an isotope into the component. Alternatively, a nuclear reactor or particle accelerator could be used to activate the component if it is capable of surviving the destructive effects of the irradiation. After predetermined periods of mixing with the tagged material, samples from different parts of the batch can be compared for radioactivity and the batch mixed until activity is constant; or autoradiographs could be made by placing samples of the mix on photographic film, allowing them to sit for a suitable period of time, developing the film, and noting the uniformity of the samples by the darkened spots on the film. Once the optimum mixing time has been determined, the tagged batches can be disposed of in some safe manner, and it should not be necessary to add isotopes to the process again until some variable is changed.

There have been a number of suggestions for the routine addition of isotopes directly to a manufacturing process for the purpose of *controlling* the process, but to date these have not been put to practice in America. Aside from possible technical reasons for this, is the fact that agencies concerned with the public's health and safety consider very carefully any proposal for adding radioisotopes to a product that might reach the public, even though the quantity is small. They are particularly concerned about adding isotopes without the public's knowledge. Moreover, the general public is not yet educated (and may never be) to accept the addition of radioactivity to commercial products unless it serves a tangible pur-

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pose for the public, such as the addition of radium to watch dials to make them glow in the dark.

The subject of controlling processes brings us to the next feature of radioisotopes—the fact that they are a compact source of radiation. This feature does allow them to be used for industrial control purposes, but in such a way that they are not added directly to the process or product. Alpha, beta, gamma, and neutron radiations from isotopic sources are transmitted, absorbed, or reflected in a manner characteristic of the material they hit. Thus, an examination of the changes produced in a beam of radiation by an unknown material can provide useful information about the material, for example, its thickness. Radioactive thickness gauges are finding extensive use for measuring and controlling the manufacture of paper, plastic sheet, and tin plate.

More in the nature of a research tool is a device employing radiation from strontium-90 to measure the ratio of hydrogen to carbon in a hydrocarbon.[2] When beta particles from radioactive strontium are passed through a hydrocarbon sample such as gasoline or oil, the attenuation or loss of the particles depends on the number of electrons they encounter in the sample. Since hydrogen has the highest number of electrons per unit weight of any element (in fact, it absorbs just about twice as many betas per unit weight as does carbon), its absorption of the beta particles is greatest. Measurement of betas absorbed by the hydrocarbon sample thus becomes an indication of its hydrogen content. More specifically, when the absorption value is combined with a measurement of specific gravity (which tells the total weight of carbon and hydrogen per unit volume), one can calculate the weight of hydrogen per unit volume or the ratio of hydrogen to carbon in the pure hydrocarbon sample, which is the desired quantity. The measurement takes only about five minutes and has a statistical probable error of 0.02 weight per cent of hydrogen. This time compares with about one to four hours for normal carbon and hydrogen measurements which have a probable error of 0.05 per cent. The tool can be used not only for plant and product control but also for research on such problems as altering the percentage of hydrogen in petroleum fuels to improve their performance.

Portable radioactive sources using isotopes such as cobalt-60,[3] cesium-137,[4] and thulium-170,[5] are also used for low-cost radiography of cast and welded metals as well as lower density materials such as plastics and paper to diagnose for defects or to study internal structures, either routinely or as part of a research program.

Examples of the third feature of radioisotopes, their great sensitivity, are numerous. Several American oil companies have determined the rate of wear of automobile piston rings by irradiating the rings in a nuclear reactor, installing them in a test engine, and measuring the accumulation of radioactive iron worn from the piston rings and collected in the oil pan.[6] The results showed a very high rate of wear during the first few hours of break-in; this would have been very difficult to measure by other means, because of the small amount of metal worn away in such a short period of time. Examination of the cylinder wall showed also that relatively large amounts of radioactive iron were adhering at each end of the stroke with less in the middle. Such information is valuable in understanding the physical forces producing wear in an engine and in speeding up the development of longer wearing parts and better lubricants.

The chemical interaction of lubricants with the surface of radioactive steel is also being studied.[7] Any iron which reacts with the lubricant, and dissolves, can be measured with great sensitivity. It is believed that greater knowledge of such chemical interaction will lead to improved lubricants.

A tire manufacturer[8] has tagged automobile tire treads by incorporating in their formulation a plasticizer, triphenylphosphate, synthesized from radiophosphorus. He could then obtain a quick measure of the rate of wear of each tire by observing the radioactivity of the rubber worn from the tire and adhering to the road surface. Because of the simplicity of the test, tread wear could be determined as a function of the roughness and composition of the road surface, of the speed of the car, and of the rate of acceleration in a small fraction of the time and cost normally required. Usually, tire manufacturers maintain large test fleets of cars, and tires are run at least 5000 miles before a measurable amount of wear takes place. Such a procedure provides only a bare minimum of information on such variables as acceleration and deceleration. Yet the radioactivity tests showed that life expectancy of a tire at 60 miles per hour is only 57 per cent of that at 30 miles per hour, and that acceleration and deceleration cause even greater wear. Information of this kind should lead to tougher, longer wearing, and safer tires to meet the needs of modern high-speed cars.

Wear measurements have also been made on tagged floor wax, paint, and cutting tools,[9] usually with increased accuracy and in a shorter time because of the high sensitivity of the test. You can probably think of other wear problems in industry that would be solved if one could measure the rate of wear faster and more accurately.

A rubber company has tagged silicone lubricant[10] to study its effectiveness on rubber de-icing tubes for airplane wings. Results of this work showed that ice removal from the rubber tube surface is simplified when silicone lubricant is applied, because the silicone layer splits as the ice is torn from the rubber surface. A small amount of silicone is removed with the ice. The minute amount removed was still easily measured on the ice by its radioactivity.

Radioactive tungsten has been added to melts of high-temperature alloys and autoradiographs were made of the resulting product,[11] which showed that the added tungsten concentrated in the dendritic areas of the crystal structure. Much smaller quantities of tungsten can be detected by this method than by conventional etching procedures. Such a nondestructive technique also offers a method of studying the behavior of individual components in alloys or other systems without in any way disturbing the normal behavior of the system.

Another example of the high sensitivity of radioactive materials is an exploratory study[12] we have made for a silverware manufacturing company. The purpose was to learn more about the mechanism by which a thimble full of ammonium thiosulfate added to 100 gallons of electroplating solution makes the resulting silverplate much shinier. Not much is known about the way electroplating brighteners act, but it has been postulated that a minute amount of a brightening agent like ammonium thiosulfate might deposit from the plating bath with the silver and interfere with the formation of large, coarse crystals which produce a dull silver surface. If this were so, sulfur from the ammonium thiosulfate should be present in the silver. But no one has ever been able to find any sulfur there, so the theory could never be substantiated, and further progress toward a more effective brightener was slowed down.

Radioactivity, however, provides an extremely sensitive means of detecting sulfur, so we synthesized some of it into ammonium thiosulfate, added it to the plating bath, and analyzed the final silverplated surface for radioactivity. Definite activity was found throughout the plate, and a calibration of radioactivity in terms of sulfur indicated one sulfur atom to every 100,000 silver atoms. The surprising part of this study was not so much that sulfur was present in the plated silver, but that it was also present as a surface layer on the *other* electrode, the silver anode, whose surface was continually dissolving in the plating solution. This discovery led to the finding that sulfur from ammonium thiosulfate deposits on silver by a chemical action without the need for any electric current at all. This result is contrary to the previous theory of how this type of brightener acts. To throw light on whether atoms other than sulfur in the ammonium thiosulfate molecule are also retained in the silverplate, one would have to tag the nitrogen, oxygen, and hydrogen atoms of that molecule. As will be mentioned later, however, no good radioactive isotope of oxygen or nitrogen is available, and until fairly recently, radioactive hydrogen could not be obtained. So much for examples of sensitivity of the isotope technique.

An example of the *specificity* of radioisotopes, which allows them to be traced and identified even in the presence of other chemically identical atoms from other sources, is found in an old problem of rubber-product manufacturers. The problem was to learn more about the mechanism by which sulfur vulcanizes rubber in the presence of sulfur-containing vulcanization accelerators. One of the approaches used was to study the chemical structure of accelerator molecules by radioisotope techniques.[13] Radioactive sulfur in elemental chemical form was heated at vulcanization temperatures in contact with tetramethylthiuram disulfide, a vulcanization accelerator, and the accelerator was then separated and assayed for radioactivity. Activity was a measure of freedom of exchange of the sulfur atoms, and thus indicated the bond strengths of sulfur in different positions of the accelerator molecule. Further evidence on the structure of tetramethylthiuram disulfide resulted from radioassays of sulfur found in the products obtained by destructively heating the tagged accelerator and also from products isolated from vulcanized rubber prepared with the accelerator. The fact that activities of the sulfur in all of these experiments were identical indicated that the tagged sulfur atoms must have distributed themselves uniformly throughout the original tetramethylthiuram disulfide molecule. Therefore, all the sulfur atoms of the molecule must be chemically equivalent. These results lent support to the theory that this accelerator molecule is a planar six-membered ring hybrid with all sulfur atoms having equivalent bond strength. This concept combined with other evidence from related studies threw further light on the complex mechanism of rubber vulcanization.

A cattle-feed manufacturer also has used radioisotope techniques to determine[14] the proper chemical form in which iodine and other vital minerals should be added to salt cake. The object was to prevent the minerals from being washed away by the rain while the cake was lying out in the pasture, and yet to allow the animal to absorb and utilize them after it had licked the salt. Various chemical forms of iodine were prepared from radioactive iodine, and these were incorporated in the salt cake. Their leachability from the salt could be determined by the radioactivity in the rainwater contacting the cake. Radioactivity measurements on the animals' vitals and excreta could show the amount utilized by the animal. The combined evidence showed that dithymol diiodide had optimum solubility for use in salt cake.

In another study,[15] a large oil company examined the mechanism by which carbonaceous coke deposits on the solid catalyst beads during catalytic cracking of petroleum. Formation of this coke decreases catalytic efficiency and lowers product yield. The aim of the study was to determine whether certain types of hydrocarbon molecules, or certain chemical groups within the molecule, are preferentially involved in the coke formation. In spite of the voluminous literature on theoretical aspects of catalytic cracking, no information on this point had been developed, but with radioactive tracers definite evidence could be obtained. Radioactive carbon-14 was synthesized into normal heptane with the middle carbon atom of the straight chain molecule labeled with carbon-14, normal octane and one-octene with the end carbons labeled. Each of these materials was passed over an experimental catalyst bed of silica-alumina beads at cracking temperatures, and the resulting coke samples were measured for radioactivity. Within experimental error, each sample of coke had the same specific activity as the labeled hydrocarbon from which it was formed. This indicated that all carbon atoms of a given hydrocarbon molecule are equally involved in coke formation. This finding suggests that molecules first adsorb on the catalyst surface and then either undergo catalytic cracking, with all fragments being desorbed as final product, or the molecules desorb completely unchanged. A few molecules must remain on the catalyst surface and be completely changed to coke, probably through dehydrogenation and polymerization. The possibility of part of the molecule forming coke preferentially seems essentially ruled out by the study.

Radioactive tracers sometimes make it possible to do research on a full plant scale. For example, we made a study of the behavior of the sulfur in a commercial coke oven for a steel company several years ago.[16] The purpose of this work was to learn whether one of the two principal forms of sulfur in coal is preferentially eliminated with the off-gas in the coking process. If it were, one could produce low-sulfur coke from high-sulfur coal providing most of the sulfur in the coal was of the type eliminated in the off-gas.

Radioactive sulfur was synthesized into one of the forms occurring naturally in coal, that is, pyrites (FeS_2). This was mixed intimately with ten tons of powdered coal which itself contained ordinary nonradioactive pyrites. The coal was charged into a full-scale commercial coke oven, continuous samples of the sulfur were extracted from the off-gas, and final samples of the coke were measured for radioactivity. Results showed that pyritic sulfur was evolved somewhat more slowly than organic sulfur for the first six hours of the run and more rapidly for the next seven hours. The net result for the entire run, however, was an almost equal rate of evolution for both types.

Thus, in a single experiment it was proved that with normal conditions of operating a coke oven the two major forms of sulfur in coal remain in the coke in about the same proportions as they existed originally in the coal. In practical terms, therefore, there would be no advantage to using a coal with a high or a low ratio of pyritic to organic sulfur. If one wanted to produce a low sulfur coke (which is always a goal of steel manufacturers), the most important point is to see that the coal has a low *total* sulfur content.

Until recently, I believe, this work was the largest scale industrial tracer experiment ever conducted, but a few months ago an automobile manufacturer ran a bigger one.[17] They were studying the usefulness of some very finely divided iron ore powder obtained by concentrating low-grade ores. The purpose was to find out whether this unusually fine powder could be charged into a blast furnace satisfactorily or whether it was going to be blown right out again by the high-velocity air going through the furnace. Five pounds of the fine ore was irradiated in a nuclear reactor, and was mixed intimately with 22 tons of identical ore, then with 54 tons more of a different, coarser type ore. This was fed into a blast furnace, and samples of the pig iron, slag, and expelled dust were checked for radioactivity. The radioactive measurements indicated that sixty per cent of the fine tagged ore dust remained in the furnace as pig iron. Though surprisingly high, this percentage was not considered good enough to be practical. The manufacturer concluded that further research is necessary into the way iron particles can adhere together or agglomerate, and that such research might show how little particles can be made into big ones so that low-grade ore concentrates can be used in a blast furnace.

My company is currently making another full-scale plant study for a client, this time on molten glass, to determine the flow pattern in a continuous glassmaking process. The work has not yet been published.[18] Radioactive sodium, phosphorus, and barium are being added intermittently to specific small portions of the glass feed materials. Radioactivity of the exit glass and of samples taken at certain key spots in the tank are noted as a function of time after the tagged batch is introduced. This work is yielding information on the holdup time of the glass in the tank as well as on the degree of mixing and channeling of the feed materials in flowing through the tank.

By simultaneous addition of radioactive isotopes such as phosphorus and barium, which have completely different radiation characteristics, we can compare the two and obtain evidence of the adequacy of each isotope as a true tracer of glass flow. Phosphorus-32 is a pure beta-ray emitter, and its radiation can be stopped by a thin sheet of metal. Barium, on the other hand, emits gamma rays which can easily penetrate the sheet. Thus it is possible to measure one isotope in the presence of the other by noting the total phosphorus plus barium activity from the samples without the filter, then measuring the barium gamma rays alone by inserting the filter between sample and counter, and stopping the phosphorus beta rays. If the phosphorus and barium are both true indicators of glass flow, they will behave alike, and their ratio in exit samples will remain constant. Results so far indicate considerable channeling and nonuniform over-all mixing. Final results will be used as a basis for designing an improved glass melting tank.

In the field of metallic corrosion, we used tracers to obtain evidence which helped settle a law suit for one of our clients.[19] This work is also unpublished. The problem revolved in part around the role of carbon disulfide as an inhibitor of corrosion of brass fire extinguishers filled with carbon tetrachloride plus small amounts of chloroform and ethyl alcohol. To throw light on the mechanism by which carbon disulfide inhibits essentially all corrosion, carbon disulfide was synthesized from radioactive sulfur, and ethyl alcohol was tagged with radioactive carbon. These materials were used separately and in various combinations with carbon tetrachloride and chloroform, and were stored inside the brass extinguishers for several months. Radioactivity showed up on the brass surface in a very thin but tenacious film which was judged by radioactivity measurements to be only about fifty to eighty molecules thick. This was well below normal limits of detection.

The fact that the film contained both radioactive carbon from the alcohol and radioactive sulfur from the carbon disulfide, showed both had contributed to the film. The relative amounts of radioactive carbon and sulfur found in the film suggested two simultaneous mechanisms of corrosion inhibition: first, a direct reaction between carbon disulfide and brass, probably to form zinc and copper sulfide; second, a preliminary reaction between carbon disulfide and ethyl alcohol to give xanthic acid (a strong organic acid) followed by the reaction of this acid with the brass surface to give zinc or copper xanthate. The radioactivity could not be removed by vacuum treatment, washing, or ionic exchange in inactive solutions, and was thus not due simply to surface adsorption of the radioactivity. Its presence indicated that there was a chemical reaction between carbon disulfide, ethyl alcohol, and brass to form a film on the brass. The film thickness is believed sufficient to account for the corrosion protection observed with normal fire extinguisher fluid containing carbon disulfide.

The varied fields from which these examples have been selected illustrate the great versatility of radioisotopes for industrial research.

Radioisotope techniques are not always simple and straightforward, and in fact, have a number of limitations. For instance, one may not always be able to obtain a proper isotope with a suitable half-life, intensity of activity, or purity (as mentioned earlier, no good radioisotope of oxygen or nitrogen is available; neither is one of silicon). Sometimes the amount of tracer that must be used is so large that safety precautions become very difficult, as in some types of atmospheric pollution studies. Synthesis of an isotope into a true tracer compound identical in chemical and physical form with the material being studied is not always possible, particularly when the material is a naturally occurring complex substance.

The high sensitivity of the tracer method sometimes imposes unusually stringent requirements on analytical techniques. For example, in a chemical separation of two products *A* and *B* for analysis, removal of perhaps 99.999 per cent of *A* from *B* would normally represent a clean separation. Yet if *A* were highly radioactive and *B* were not, the 0.001 per cent of *A* remaining might completely invalidate activity measurements of *B*.

Special equipment is usually needed to measure the radioisotopes and to monitor the levels of radioactivity for health purposes. It is usually advisable to set aside separate laboratory space for work with radioisotopes so that radioactive contamination can be controlled and unauthorized (but curious) people can be excluded. At least one person on the staff must be trained to use isotopes in order to qualify to receive them from the United States Atomic Energy Commission. Sometimes there are problems of public relations to consider, particularly if the work is going to involve the public in any way.

In the last few years, however, some former limitations have been overcome. Availability of isotopes in many chemical forms has improved. Safety procedures such as decontamination, waste disposal, shielding, and remote control have now been well worked out, and industry should feel no qualms about using radioactivity providing qualified personnel are responsible for the work. Measuring instruments are now much more accurate, reliable, and simple to operate than those available a few years ago. There are many more technical people familiar with the use of isotopes and qualified to obtain them.

As for over-all cost of doing industrial tracer research, generalizations are hazardous because the research problems vary from easy to difficult, and therefore, from low cost to high. The simplicity of the tracer principle and the ease of radioisotope detection lead one to think that tracer research in general should always be quick, easy, and cheap. As a matter of fact, it usually is simpler, quicker, and cheaper than other research methods for the kinds of problems we have just been talking about—the kinds for which tracers are best suited. But these problems are frequently the tough, complicated ones which cannot be solved easily by other methods. Thus, even with tracers the total research cost may sometimes seem high to those who have thought of the technique as a delightfully simple panacea. As in most research, the major direct cost is for the time of the research worker—to plan, carry out, interpret, and report the work. We have found that a good research man familiar with solving industrial research problems can quite easily become proficient in using radiochemical techniques, so that at least for modest scale operations, a full-time radioactivity specialist is usually not required.

Use of radioisotopes for industrial research in America is increasing steadily. Future rate of growth depends not on cost but on the industrial researcher himself. This man is in a key position to understand the capabilities of radioisotope techniques and also to recognize the particular problems which they can solve. Radioisotopes are ready to reward the ingenious researcher with new knowledge for the benefit of industry and mankind.

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The Use of Radioisotopes in Uruguay and Production Prospects for Them in the Republic

By Walter S. Hill,[*] and German E. Villar,[†] Oriental Republic of Uruguay

The peaceful use of atomic energy has three aspects which are of great interest to Uruguay, as follows: (*a*) the technical and scientific use of radioisotopes; (*b*) the production of radioisotopes; and (*c*) the generation of nuclear power.

In so far as point (*c*), in view of its importance, has been dealt with in a separate paper, this communication will refer to points (*a*) and (*b*), with special emphasis on their importance for Uruguay.

THE TECHNICAL AND SCIENTIFIC USES OF RADIOISOTOPES IN URUGUAY

The use of the radioisotopes in Uruguay began in 1950, at the Nuclear Physics Laboratory of the Institute of Physics of the Faculty of Engineering and Surveying. The laboratory immediately went into the study of the scientific and medical applications of the radioisotopes, in cooperation with units of the Public Health Ministry, which later were to set up their own laboratories.

At the present time, five laboratories, in our country, specialize in radioisotope techniques. They are: (*a*) the Nuclear Physics Laboratory of the Physics Institute of the Faculty of Engineering and Related Arts; (*b*) the Radioisotope Laboratory of the Institute of Endocrinology of the Public Health Ministry; (*c*) the Institute of Radiotherapeutics of the Public Health Ministry; (*d*) the Institute of Biophysics of the Faculty of Medicine; and (*e*) the private "mc^2" Laboratory.

The Nuclear Fission Laboratory of the Institute of Physics of the Faculty of Engineering (Avenida Herrera y Ressig 565, Montevideo), concentrates on scientific research, with the help of the Rockefeller Foundation.

The Radioisotope Laboratory of the Institute of Endocrinology (Larravide 74, Montevideo) works in cooperation with the Institute of Physics of the Faculty of Engineering. Its activity is oriented toward scientific research and medical applications, mainly as regards the thyroid.

The Institute of Radiotherapeutics of the Public Health Ministry (Hospital Pereyra Rossell, Bulevar Artigas 1550, Montevideo), concentrates mainly on the medical applications of the radioisotopes.

The Biophysics Institute of the Faculty of Medicine (Avenida General Flores 2125, Montevideo), carries out scientific investigations in the field of the application of the radioisotopes to medicine.

The "mc^2" Laboratory (Avenida 8 de Octubre 2874, Montevideo), concentrates on scientific investigation, the medical applications of the radioisotopes, and their use in industrial X-ray work.

Even though the laboratories just mentioned still are in the first phase of their development, they are well equipped, and an idea of the importance of their activity is given by their consumption of radioisotopes which, for the year 1954, reached a total of some 2000 millicuries.

The radioisotopes in use in Uruguay come, for the most part, from England (Harwell and Amersham).

PRODUCTION OF RADIOISOTOPES IN URUGUAY

Allowing for the rapid development which the studies related to the use of the radioisotopes is taking in Uruguay, as well as their medical applications, the need for the installation of a small nuclear reactor in the country becomes more imperative every day, for it would make it possible to widen the field of these studies and research with short-lived radioisotopes, the use of which is now impossible, in view of our distance from the production centers.

In addition, a nuclear reactor would be a valuable tool for the training and experience of the engineers who might specialize in nuclear engineering.

The Institute of Physics of the Faculty of Engineering and Agriculture is currently studying the possibility of carrying out, in the near future, an installation of this type, the essential characteristics of which will be determined in agreement with such information as may be gathered at the International Conference on the Peaceful Uses of Atomic Energy.

Original language: Spanish.

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Large-Scale Production of Radioisotopes

By A. F. Rupp,* USA

In June of 1946 there appeared a significant article in the magazine *Science*,[1] announcing the availability of radioisotopes from Oak Ridge. From a very modest beginning, the radioisotope program has grown into a business with sales over 1.5-million dollars per year from Oak Ridge National Laboratory alone; and thousands of dollars of additional business is done annually by secondary processors receiving material from Oak Ridge National Laboratory and other production sites in the United States.

The first large reactor (so-called X-10 graphite reactor) was used to produce the first small amounts of radioisotopes for distribution, and in August of 1946 the first shipment (of carbon-14) was sent to Bernard Free Skin and Cancer Hospital† for cancer research. Only laboratory-type chemical processing facilities were available for work on radioactive material, but the program was pursued vigorously, and by 1948 the business had grown sufficiently and experience had been acquired to allow the design and construction of a radioisotope processing area. The growth of the radioisotope program is shown in Fig. 1. It may be noted that there has been a steady increase in shipments during the years that the program has been in operation. The radioisotope program in the United States is set up to encourage the processing of materials by commercial laboratories; and it may also be noted that the amount per shipment has grown rapidly in recent years, partly because of a change in the character of the business—from the retailing of small amounts to many users, to wholesale-type sales to commercial processors who reprocess and package radioisotopes in smaller quantities for special kinds of uses. The average shipment has grown from 1 or 2 millicuries per package at the beginning of the program to an average of 2000 millicuries per shipment at the present time; however, the average figure is strongly influenced by very large shipments of cobalt-60 for teletherapy made in the last few years—but it is not unusual to send shipments as large as 1000 millicuries of other radioisotopes such as iodine-131 to secondary processors. Many special compounds and devices are made by the commercial processors; for example, the preparation of ampoules from which the physician can take standardized doses of radioisotopes for diagnostic work is being performed by well-established pharmaceutical houses, long familiar with the practices and requirements of the medical profession. The magnitude of the business by such secondary processors is considerable. In 1954, one such secondary processor made a total of 15,000 shipments, principally to medical and biological users. The total amount of activity in these 15,000 shipments was about 800 curies; however, this activity came almost exclusively from the facilities at Oak Ridge National Laboratory and was sent out in approximately 1000 shipments.

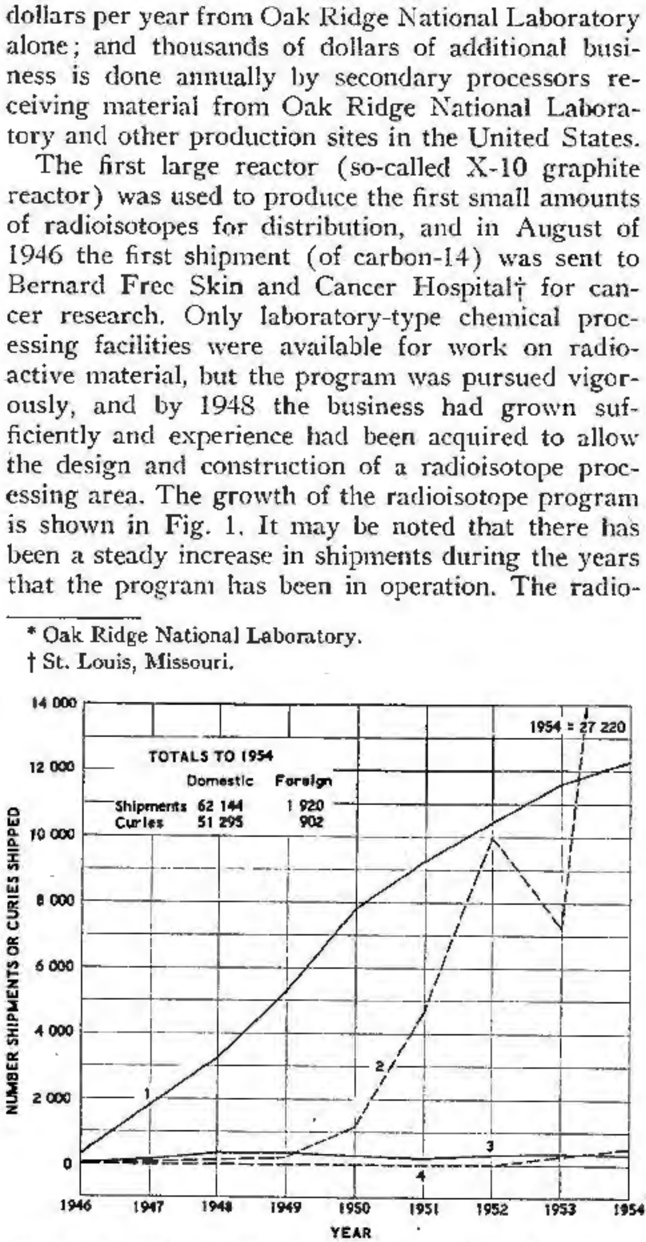

Figure 1. Growth of the radioisotope program at Oak Ridge National Laboratory: (1) domestic shipments; (2) domestic curies; (3) foreign shipments; (4) foreign curies

RADIOISOTOPE PRODUCTION OPERATIONS

Shown in Fig. 2 is a flowsheet of the radioisotope production operations. It will be noted that production centers around the nuclear reactor, although small amounts of certain radioisotopes are now being made on cyclotrons, such as the very important radioisotopes sodium-22, manganese-54, and beryllium-7. Radioisotopes are produced in the reactor by two main processes: irradiation of target materials with neutrons in (n, γ), (n, p), or similar processes, and by the formation of fission products in the reactor fuel.[2] Some isotopes can be made in several ways; e.g., iodine-131 was originally made by irradiation of tellurium, and after the demand exceeded the amounts that could be easily produced by that

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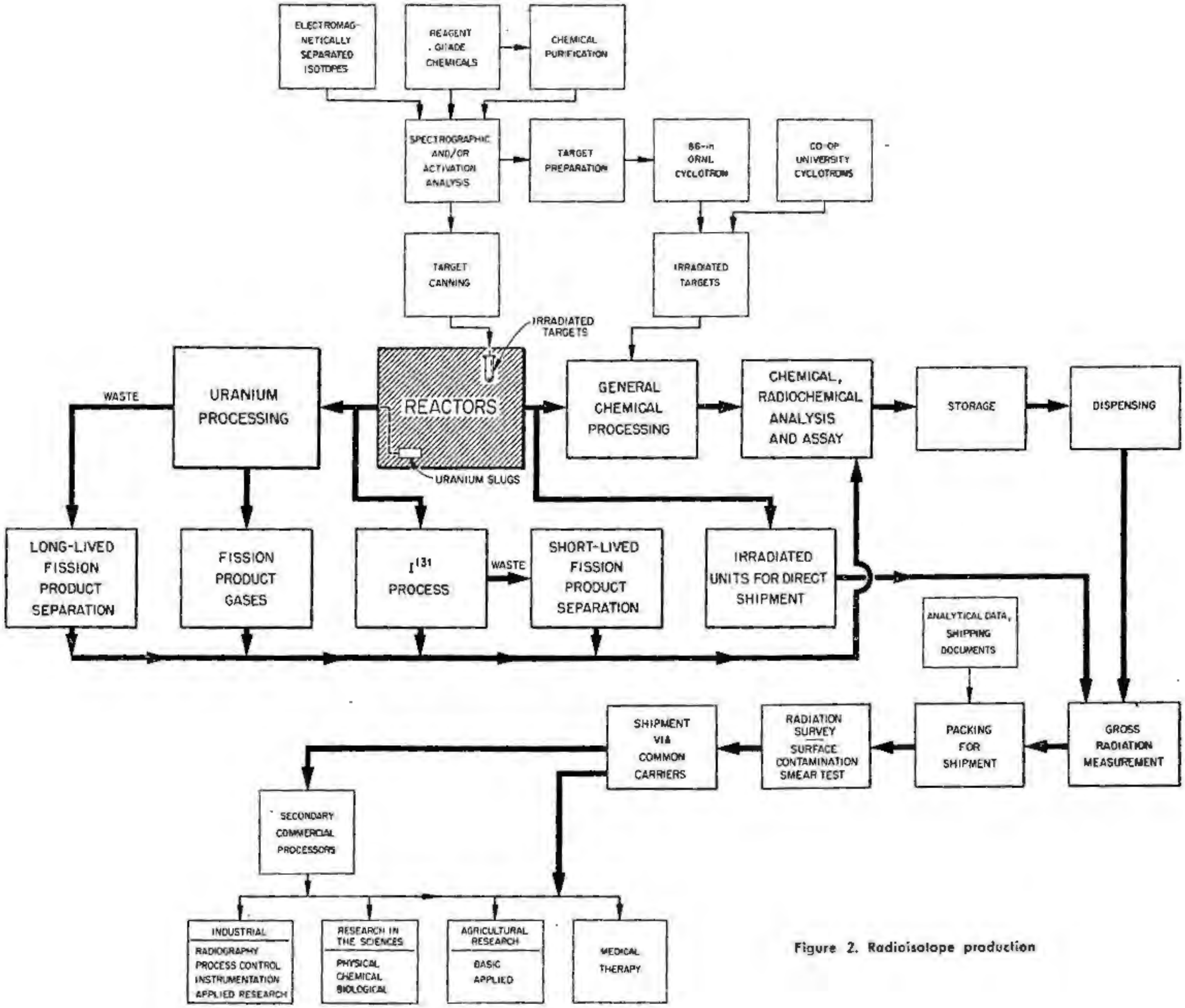

Figure 2. Radioisotope production

method, the separation of fission product iodine from uranium fuel was started. After materials are removed from the nuclear reactor either as irradiated targets or as uranium fuel, the next principal step is chemical processing. A wide variety of chemical processes are employed, some of the more important of which are discussed later in this paper. After chemical processing, it is necessary to test the products carefully for identity, radiochemical and chemical purity; tests for sterility or pyrogenic substances are not made, although high standards of cleanliness are maintained during all operations. The preparations are stored, principally those radioisotopes having half-lives of over thirty days. The next operation is dispensing, which is done by remote control facilities. Packaging for shipment is the next operation; radioisotopes are shipped out either in sturdy wooden boxes containing lead shields (designated as "returnable" containers on which the user must pay a deposit and return to the Laboratory) or a newer type "disposable" container which consists of a small internal lead shield surrounded by highly absorbent paper, sealed in a metal container, which is then enclosed in a pasteboard box. The use of the disposable-type container has developed to the point where over 90% of all shipments from Oak Ridge National Laboratory are made in this manner.

All target materials used in the program are carefully selected or purified before irradiation in the nuclear reactor; spectrographic analyses and in some cases activation analyses are made to determine the purity, in addition to the manufacturer's analyses. An effort is made to collect fairly large batches of highly-purified material with known properties for use as target irradiation material. In some cases, when exceedingly high specific activities are required (e.g., iron-59) and no suitable processes are available for producing carrier-free or unusually high specific activity material, it is necessary to use electromagnetically-enriched stable isotopes as target material. This has proved very successful and very high specific activity iron-59 has been made from iron enriched to 50% in the target atom (Fe^{58}), from the natural abundance of 0.31%. The specific activities obtained are in the order of 5000 millicuries per gram, compared with that previously available from normal iron of only 25 millicuries per gram.

Figure 3 is a photograph showing the main part of the isotope processing area. A number of small separate operating buildings are used for several reasons. It was found desirable to separate high-radiation level from low-radiation level operations and processing of chemically similar materials in which cross-contamination would be a serious problem. Also, it is possible to reduce the spreading of contamination by use of separated working areas. There has been no serious general contamination in the ORNL radioisotope area since it was built in 1948. This design has proved highly satisfactory for the many special types of operations required in a radioisotope production area. Much of the heavy work is done on the outside of the buildings, where heavy lift trucks pass along the roadways and work into the doorways of cells which open to the outside of the buildings. Complete underground facilities are provided for discharge of hot waste liquids to the main Laboratory waste system. Contaminated gases are removed through filtering and decontamination equipment and discharged to the atmosphere through a 250-foot-high stack (Fig. 4).

RECENT IMPROVEMENTS IN SHIPPING RADIOISOTOPES

The shipment of radioactive materials usually requires heavier containers and consequent shipping expenses not encountered in the transportation of similar amounts of non-radioactive materials. Although many improvements have been made to reduce container weights and shipping costs, occasionally there are some small shipments where the cost of container and transportation charges exceed the actual money value of the product.

ORNL has developed new containers that are fairly inexpensive and effort has been made wherever possible to substitute light "disposable" containers for the heavy returnable type. The use of a dis-

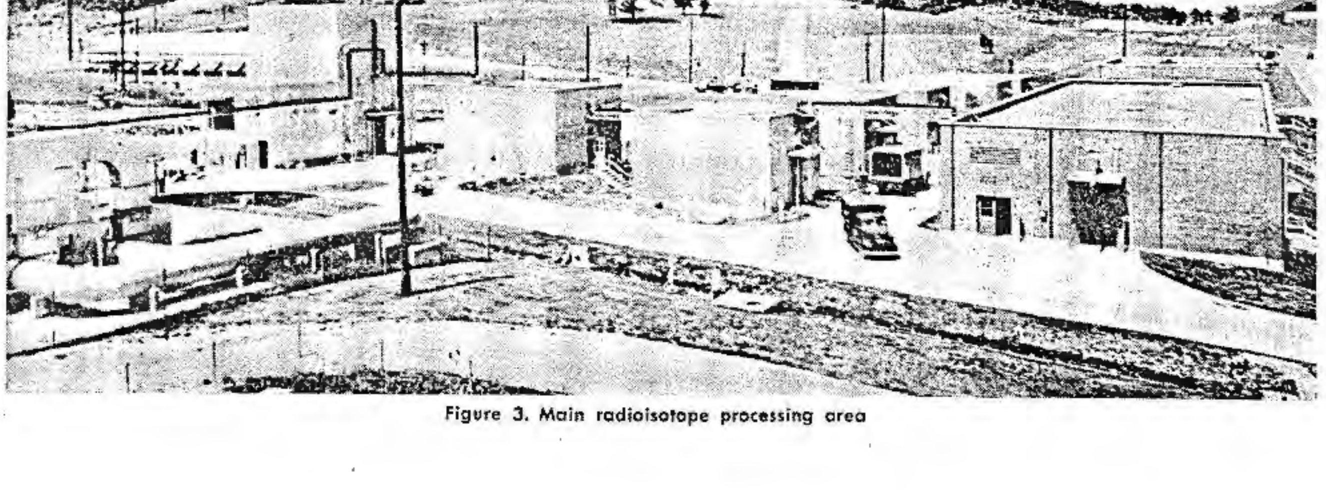

Figure 3. Main radioisotope processing area

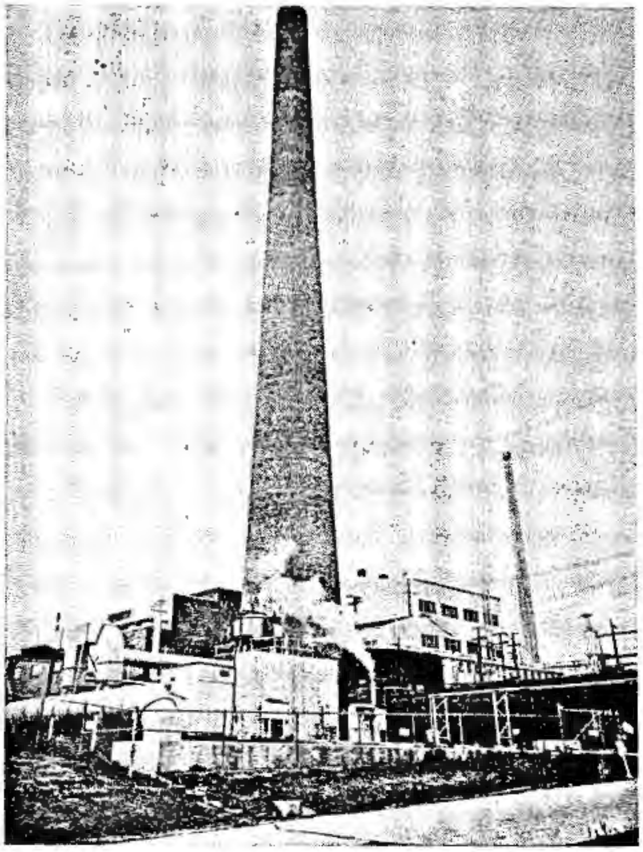

Figure 4. Stack and decontamination equipment for exhaust gases

posable container saves the customer up to 70% of the transportation costs; and the Laboratory saves the cost of handling, storage, decontamination and inspection of returned containers. From 1948 to 1954 the shipping weight per curie has been reduced from 1400 pounds to 12 pounds. While most of this great reduction can be attributed to the growth in size of individual shipments (especially cobalt-60) and relaxation of shipping regulations, a large share may be attributed to the use of light disposable containers.

The advantage of using disposable containers was first shown with shipments of beta emitters, such as P^{32}. The original returnable container used for P^{32} was composed of a steel and lead shield in a wooden box and was the lightest container in use at that time, weighing about 9 pounds and costing approximately $65 each. The disposable containers now used for P^{32} shipments (see Fig. 5) weigh only 2 pounds and cost 30¢ each. It is estimated that the saving to the customer is about $4, or 60% of the cost per shipment in the returnable container.

The use of disposable containers was later extended to small quantities of gamma emitters, saving customers about $6.50 per shipment. This was accomplished by enclosing the glass bottles (processed radioisotope solutions) or aluminum inner containers (reactor-irradiated units) in small lead shields with wall thickness varying from ⅛ inch to ½ inch (see Figs. 6 and 7). To prevent the inner assembly, made heavier by the addition of lead, from shifting within the fiberboard box while in transit, it was necessary to design a new type of box insert. The thickness of lead used in disposable containers has been limited to ½ inch in order to keep the cost less than the cost of using returnable containers. In an effort to further increase their use, the economics of increasing the shielding thickness to ¾ inch is now being studied.

Another feature of the design of disposable containers is the use of large fiberboard boxes which reduce the radiation at the surface of the package by increasing the distance to the radioactive material. The two types of boxes now in use have cubical dimensions of 8¼ inches and 12 inches.

Other standard containers in addition to the ones already mentioned are shown in Figs. 8 to 10. The containers shown are used for approximately 98% of all shipments; the remaining 2%, not described, covers a great variety of containers designed for use in shipments of special products. The quantities and types of radioactivity shipped in the various standard containers and the combinations of boxes and shields used are given in Table 1.

IODINE-131

Iodine-131 is one of the most important radioisotopes distributed in the radioisotope program. It was originally produced by irradiating tellurium in the reactor and then distilling off the iodine (produced by decay of Te^{131}) after dissolution of tellurium in a mixture of sulfuric acid and chromic acid.[3] Millicurie amounts of iodine-131 per batch

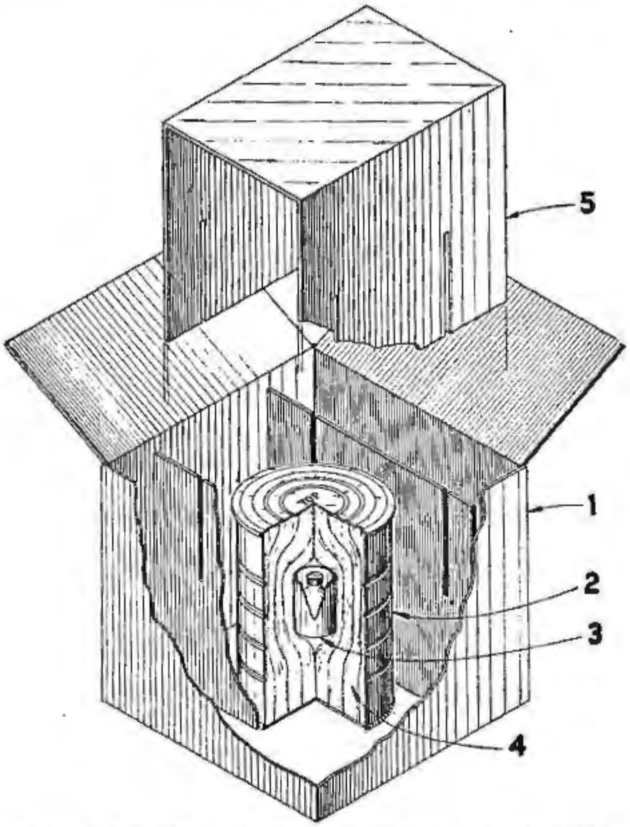

Figure 5. Disposable container for separated radioisotopes: (1) fiberboard box (350 lb test); tin can; (3) fiberboard tube; (4) absorbent wadding; (5) fiberboard insert

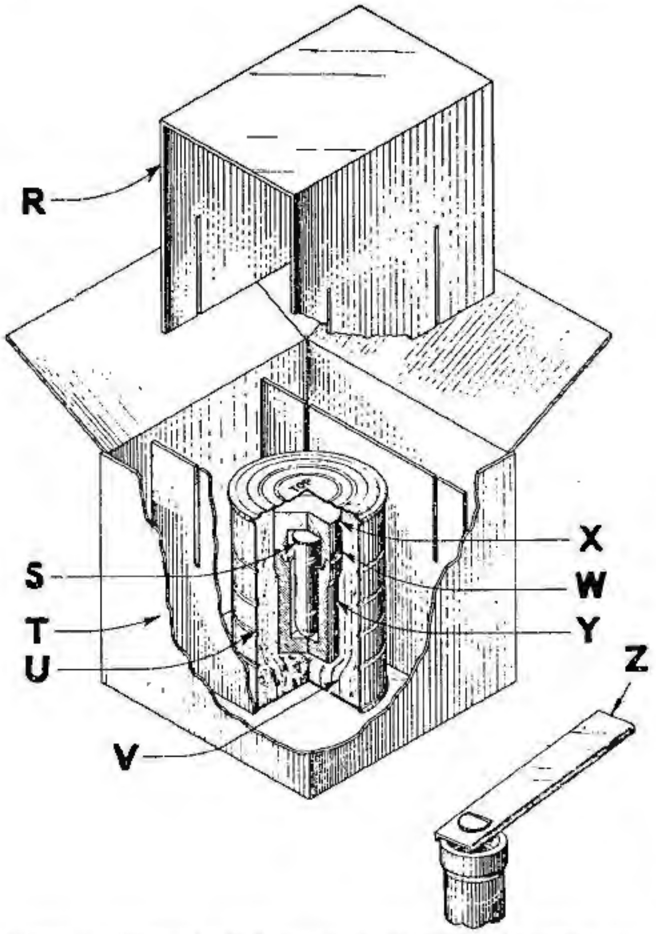

Figure 6. Disposable container for unseparated radioisotopes (reactor units). *R* is fiberboard insert; *S* is aluminum can; *T* is fiberboard box (350 lb test); *U* is tin can; *V* is absorbent paper wadding; *W* is Scotch tape seal; *X* is top section of lead container; *Y* is bottom section of lead container; *Z* is decapper

were made in this way but the growth of the program required larger amounts, so the production of fission product iodine from uranium metal was started in 1948. The original small plant could produce approximately 3 curies per batch and was operated from 1948 to 1954. In 1954 a new plant was installed with a capacity of approximately 25 curies per batch, using 30–60-day irradiated uranium metal available from the graphite reactor (Fig. 11). The essential parts of the original I^{131} process were the dissolution of the uranium metal in strong nitric acid, followed by a steam-air distillation of the elemental I^{131} through a condenser where part of the iodine was removed by the condensed liquid, and the vapor was passed through a caustic solution to remove most of the remaining iodine-131; 99.9% of the iodine passing the condensers was removed by the 50% sodium hydroxide scrubbing solution. Part of the iodine comes off promptly during the dissolution of the uranium and accompanies the NO_2 gases to the condensing and scrubbing sections; the remainder must be distilled out with steam-air sparging, giving large volumes of nitric acid, nitrous acid, water and (from the scrubber) alkaline sodium ni-

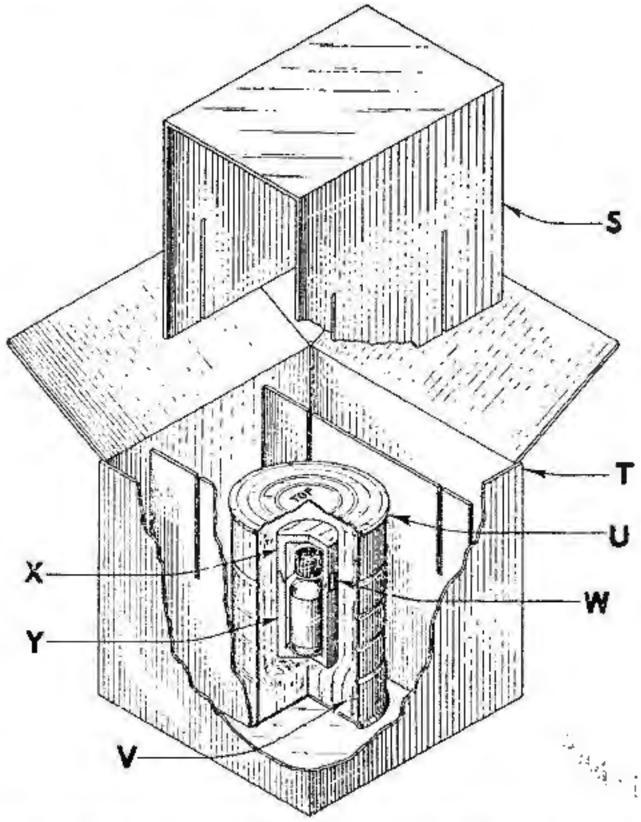

Figure 7. Disposable container for separated radioisotopes. *S* is fiberboard insert; *T* is fiberboard box (350 lb test); *U* is tin can; *V* is absorbent paper wadding; *W* is Scotch tape seal; *X* is top section lead container; *Y* is bottom section lead container

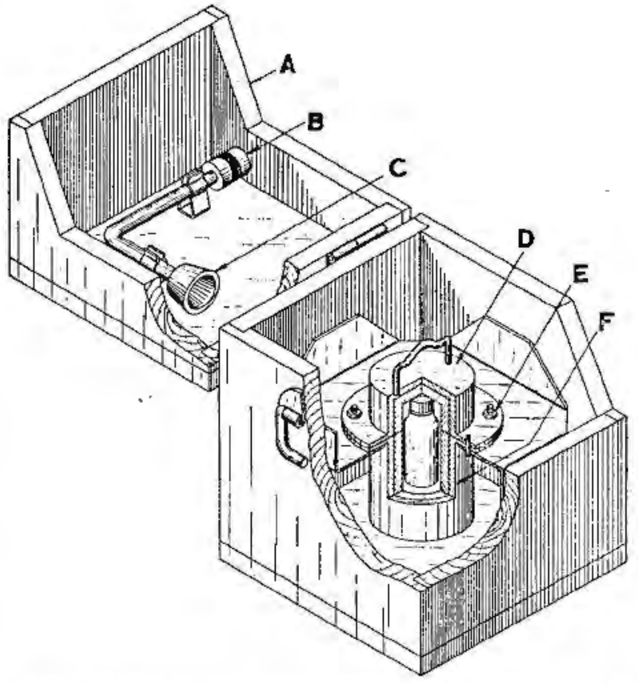

Figure 8. Returnable containers for separated isotopes. *A* is ¾ in. plywood box; *B* is wrench; *C* is bottle cap remover; *D* is top lead shield; *E* are nuts; *F* is bottom lead shield

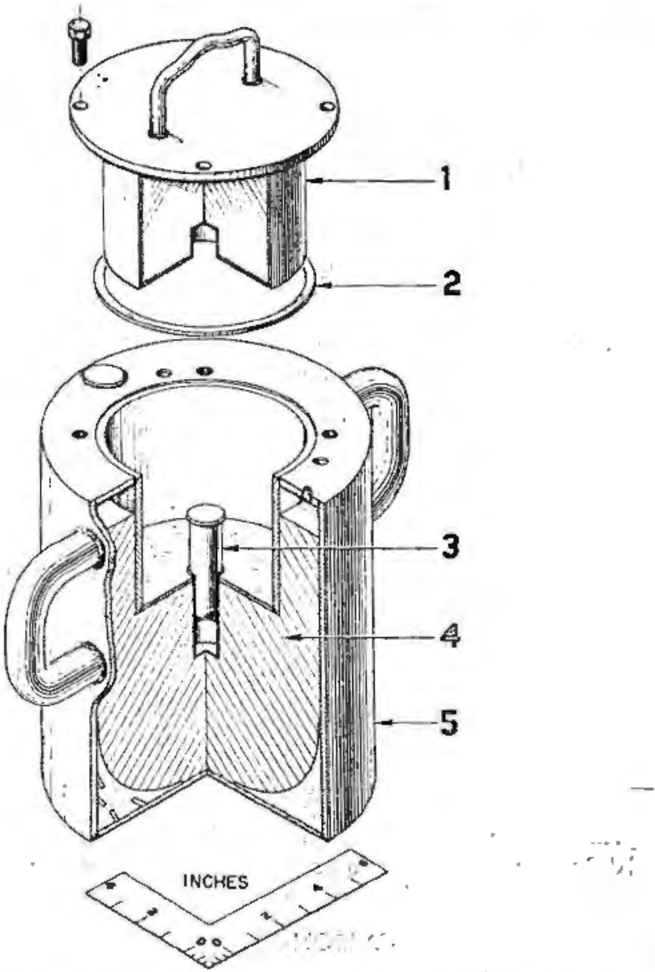

Figure 9. Returnable container for unseparated isotopes (reactor units) for larger quantities: (1) plug; (2) neoprene gasket; (3) aluminum can; (4) lead; (5) stainless steel body

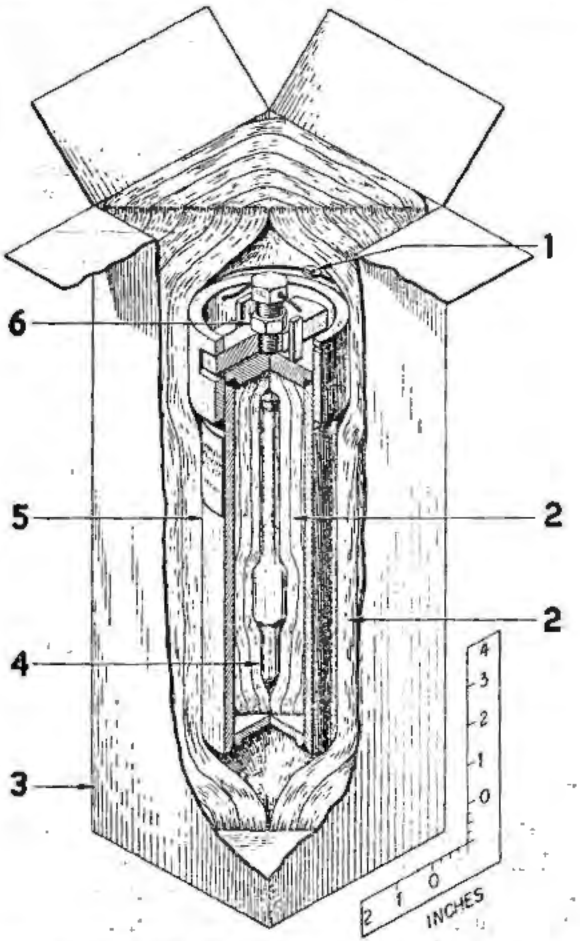

Figure 10. Returnable gas container: (1) wire and seal; (2) paper wadding; (3) fiberboard box, (4) Pyrex ampoule; (5) aluminum container; (6) locking bolt

trate-nitrite solution. It is necessary to distill and concentrate the iodine from these liquors, and at the same time remove as much as possible of the contaminating nitrate. This is accomplished by treating the liquors with hydrogen peroxide to oxidize the nitrite, and subsequent distillation of the elemental iodine into a small volume of dilute sodium hydroxide solution. It was found necessary to add small amounts of hydrogen peroxide continuously throughout the distillation in order to obtain good yields.

With the first fission product process, approximately 40% of the iodine passed on through the condensers into the alkaline scrubber. Experiments indicated that it was possible to remove almost all of the remaining iodine by passing the vapors through an efficient bubble-cap scrubbing tower containing very cold water, at about 3°C. In the present plant the condensers, condensate tank and water scrubbing tower are cooled with refrigerated water. Using this technique, almost all of the iodine is captured in a watery solution containing relatively small amounts of nitrous or nitric acid and it is not necessary to process a large amount of liquid from the caustic scrubber. In addition to this technique, it has been found advisable to introduce a small stream of oxygen at the base of the cold water scrubbing tower to oxidize the nitric oxide (NO) continuously to nitrogen dioxide (NO_2), thus forming relatively pure nitric acid in the scrubbed liquor rather than unstable nitrous acid. The final alkaline scrubber is used only to remove the remaining 1% of iodine from vapors so that the gases can be passed on to charcoal traps for the absorption of xenon-133. The gases from the final part of the process are discharged into the area off-gas system, where it is passed through precipitrons, filters, and then is discharged through the 250-foot stack to the atmosphere.

The concentrated iodine distillate from the previously-mentioned hydrogen peroxide distillation is about 1 liter in volume and may contain 25–100 curies of I^{131}. The distillation is conducted through a four-plate bubble cap column which tends to prevent the distillation of the less volatile hydrogen peroxide and nitric acid, thereby giving an almost pure distillate. Nevertheless, it is desirable to repurify this final distillate in glass equipment in order to achieve the extremely high purity required for I^{131} used for medical purposes. In the 1948 plant, the final puri-

TABLE 1. Radioisotope Shipping Container Data

Figure number	Type of Container	Type of material shipped	Maximum activity		Thickness of lead shield (in.)	Outside dimensions of boxes (in.)	Volume of bottle (ml)	Total weight of assembled container (lb)
			Typical radioisotope	Amount				
5	Disposable	Separated, liquid,	P^{32}	350 mc	None	8¼ × 8¼ × 8¼	15, 25, 50, 100, 200, 500	2
		beta emitters,	I^{131}	9 mc				2
		and small	P^{32}	1 curie	None	12 × 12 × 12	Same	4
		amounts of	I^{131}	30 mc				4
		gamma emitters						
7	Disposable	Separated, liquid,	I^{131}	50 mc	⅛	8¼ × 8¼ × 8¼	15	3
		gamma emitters		95 mc	¼		15	4
				145 mc	⅜		15, 25	Min. 5 Max. 6
				320 mc	½		15, 25	Min. 6 Max. 8
				600 mc	½	12 × 12 × 12	25	10
6	Disposable	Unseparated solids	I^{131}	95 mc	¼	8¼ × 8¼ × 8¼	Aluminum container,	5
		(reactor units)		300 mc	½		no bottles	7
				500 mc	½	12 × 12 × 12	Same	10
8	Returnable	Separated, liquid,	Co^{60}	5 mc	½	9½ × 9½ × 12	15, 25, 50, 100	Min. 23 Max. 35
		gamma		25 mc	1		15, 25, 50, 100	Min. 35 Max. 56
		emitters		30 mc	1½		15, 25	Min. 55 Max. 57
				5 mc	½	20¼ × 20¼ × 16¼	200, 500	Min. 90 Max. 98
				25 mc	1		200, 500	Min. 118 Max. 132
				40 mc	1½		50, 100	Min. 124 Max. 136
				100 mc	2		15, 25, 100, 200	Min. 133 Max. 226
				500 mc	3		15, 25	Min. 228 Max. 250
9	Returnable	Unseparated	Co^{60}	1 curie	4	None used	Aluminum irradiation	450
		solids, gamma		5 curies	5		can, no bottles	650
		emitters		25 curies	6			810
				18 curies	7½			1320
10	Returnable	Gases	H^3	25 curies	None	5 × 5 × 16	Ampoules of 1, 5, 10, 25	4
			A^{37}	15 mc			cm^3	4

fication was accomplished by oxidizing the iodine to iodate with potassium permanganate, adjusting to 20% by volume with sulfuric acid, reducing the iodate to the elemental iodine with phosphorous acid catalyzed with a small quantity of hydrogen peroxide, and then quickly distilling the elemental I^{131}. Any nitric acid contamination that was present was removed by a pre-distillation during the acid permanganate part of the cycle, the iodine remaining in the non-volatile iodate form. This procedure worked fairly well for many years, but being a somewhat temperamental procedure, a better one for routine use was desired. This was accomplished very simply by adjusting the liquor to approximately 20% by volume with sulfuric acid, and then distilling the iodine through an eleven-plate, Bruun-type glass fractionating column, collecting the distillate in dilute sulfurous acid, reducing the iodine promptly and completely to the iodide form. After filtering through a bacteriological-type filter and adjustment of volume, the pH is adjusted to 9.0 with sodium bicarbonate and the product is removed for analysis and dispensing. Hydrogen peroxide is also used to assist the distillation of iodine in this final procedure; however, the rectifying column effectively keeps the hydrogen peroxide and nitric acid from passing into the distillate. Iodine-131 with no detectable radioactive contamination and of highest chemical purity is routinely produced in this manner. The over-all yield in this process, based upon the calculated amount of iodine-131 in the original uranium slugs, is approximately 85%. The yield loss that is incurred is principally in the distillation from the dissolver since 10–15% invariably remains in the uranium solution; subsequent distillations are over 90% efficient.

CARBON-14

The flowsheet for the production of carbon-14 is shown in Fig. 12. This important radioisotope has also been produced in a number of ways at Oak Ridge National Laboratory, the first production being from the irradiation of ammonium nitrate solution in a loop in the graphite reactor. Although some very high specific activity material was made this way, the process was difficult to operate and therefore was used only a short time. Calcium nitrate was then used as target material and this process was continued until 1948, when production of carbon-14 using beryllium nitride target material was started. Beryllium nitride was selected as an ideal target material because of its very stable nitride and favorable nuclear properties, there being little parasitic absorption of neutrons in the beryllium. The compound also contains approximately 50% by weight of nitrogen, making it a very compact form of target.[4] Large quantities of beryllium nitride of sufficiently high purity have been produced and are being irradiated to produce a continuing supply of carbon-14 of steadily increasing specific activity.

The aluminum jackets that are used to enclose the Be_3N_2 pellets are removed from the irradiated pellets by melting them off in a furnace. The pellets are then dissolved in 65% sulfuric acid and varying amounts of H_2O_2, and the carbon-containing compounds are

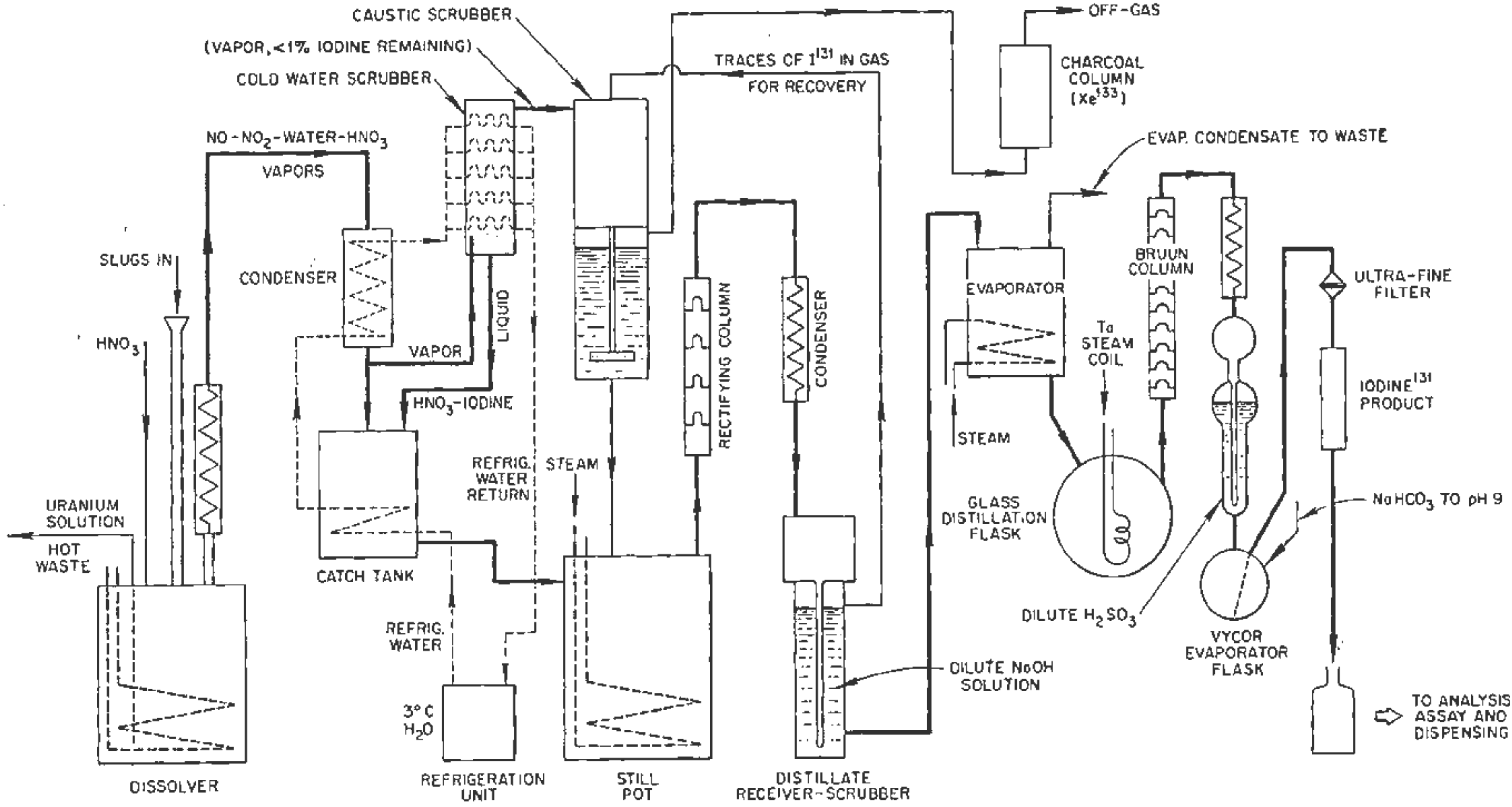

Figure 11. I^{131} process flowsheet

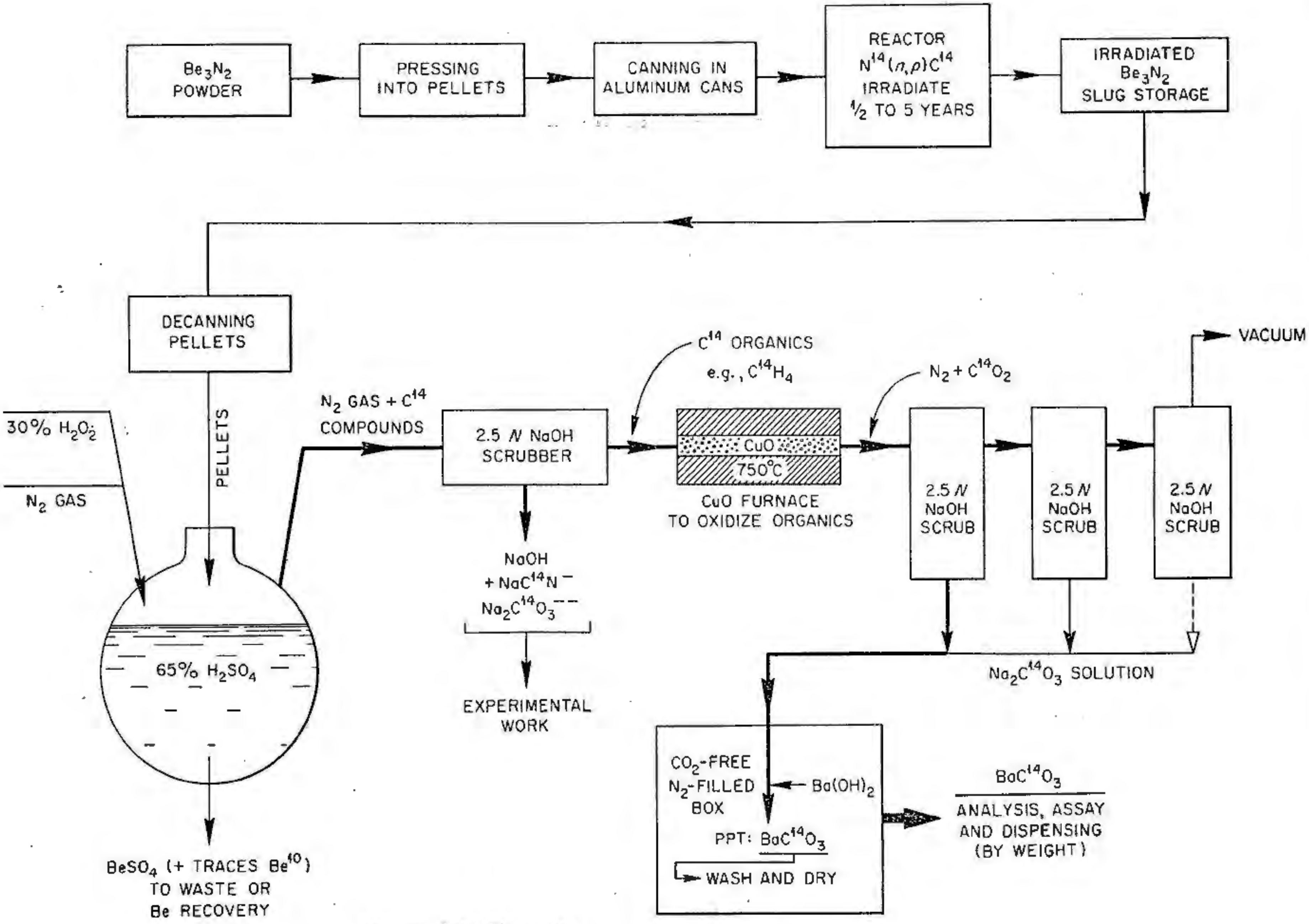

Figure 12. Carbon-14 production

swept out of the solution in a stream of highly purified nitrogen gas.[5] A variety of carbon compounds are given off during the dissolution process, the principal ones being methane, carbon dioxide, and carbon monoxide; small amounts of cyanide and miscellaneous organic compounds are also formed. The gases are passed through a bed of copper oxide at a temperature of approximately 750°C, to oxidize all the carbon compounds to carbon dioxide. Some water is formed by the oxidation of hydrogen compounds and this is removed in a small trap before the gases are passed to scrubbers containing highly-purified sodium hydroxide solution. $NaC^{14}O_3$, which is formed in the solution, is then removed to a box filled with CO_2-free nitrogen and the carbonate is precipitated with barium hydroxide. The precipitate is carefully washed and dried before chemical analysis and assay. The isotopic abundance of the carbon-14 produced has ranged from about 2% with the calcium nitrate process up to a maximum of 30% with the present beryllium nitride process. Small amounts of carbon are included in the target material and contribute almost all of the excess carbon that is found in the finished product. Tests have been made showing that only very minute quantities of carbon are introduced from the chemicals and nitrogen gas used in the process.

Some of the organic compounds in the gas generated during the dissolving of Be_3N_2 can be separated, and experiments have been run showing that the cyanide that is present has an extremely high specific activity. There is very little demand for extremely high specific activity carbon-14, but it is believed that by isolating some of the various organic compounds small quantities of very high specific activity material, approaching 100% abundance, will be obtained.

PHOSPHORUS-32

Phosphorus-32, a radioisotope useful for medical and agricultural research, is made in relatively large quantities. The production capacity of the equipment at ORNL is approximately 6 curies per batch. Because the flux available in the graphite reactor is fairly low, it is necessary to irradiate large quantities of sulfur to achieve the necessary production [by (n,p) reaction]. Kilogram amounts of highly-purified sulfur are melted into large aluminum target cans and the temperature of the reactor is such that the sulfur remains molten in the reactor during irradiation; however, the aluminum container is not appreciably attacked. Gas pressure sometimes forms in the cans during irradiation, which has been traced to small quantities of moisture in the cans and organic matter in the target sulfur which react with the molten sulfur to produce hydrogen sulfide. Some effort has been devoted to removing the organic matter by treating the molten sulfur over long periods of time with magnesium oxide, followed by a very fine filtration to remove the organic matter as a tar-like material adhering to the MgO. Almost all grades of sulfur produced in the United States are free from arsenic, so that it is not necessary to be concerned about arsenic contamination in the final product.

While the first extractions of phosphorus from sulfur were made by the vigorous method of pouring the molten sulfur into boiling nitric acid, this method was used only a very short while and it was replaced by pressure extractions with dilute nitric acid.[6] The first extractors were made of stainless steel and the dilute 0.1 N nitric acid was mixed with the molten sulfur by using a propeller-type agitator. Good yields were obtained, but the mechanical difficulties were considerable and this method was abandoned in 1949 in favor of the present stationary autoclaving method. This consists merely of melting the sulfur out of the target container into a large Pyrex glass tube enclosed in a stainless steel autoclave. Dilute nitric acid is placed on top of the sulfur and the tube closed and the autoclave lid is bolted on. The temperature is brought up to 135 to 138°C, by applying steam to a jacket on the autoclave. The autoclaving is allowed to continue overnight, during which time over 90% of the phosphorus (as orthophosphate) is extracted into the aqueous phase from the molten sulfur. The efficiency and rate of extraction during this process could of course be increased by better agitation, but since it is allowed to extract overnight and the extraction is accompanied by a minimum of trouble from leakage of radioactive materials out through seals (one of the troubles with the process where power-agitation was used), it is a quite satisfactory production method. After the extraction is completed, the aqueous phase is drawn off and sent to a quartz vessel for the first step of the purification process: evaporation to reduce volume and destroy organic matter by boiling with aqua regia. The glass tube in which the extraction was accomplished is slightly tapered so that a small rod may be placed in the molten sulfur and then upon cooling, the hard sulfur cake can be lifted from the tube for easy disposal.

During the purification process, the main problem is to separate the microgram quantities of P^{32} (as phosphate) away from the accompanying impurities, which are sulfuric acid from oxidation of sulfur, corrosion products such as ion, chromium, nickel, impurities extracted out of the sulfur which consist of organic matter, magnesium, aluminum, potassium, sodium, lead, and many other trace elements. During the evaporation and the original treatment with aqua regia in the quartz evaporator, much of the organic matter is destroyed. Water is added and the acidity is adjusted to as low a point as possible, and still allow complete removal of the phosphorus from the walls of the evaporator. The solution is passed through a cation exchange bed (Amberlite IR-120) to remove the bulk of the iron, chromium, nickel, and other cations. The phosphate-containing liquid in the effluent is then put in a precipitator of the Stang-reactor type. About 300 milligrams of lanthanum is added as carrier, and by adding ammonium hydroxide, lanthanum hydroxide is precipitated, carrying with it all the P^{32}, possibly as lanthanum phosphate; sulfate and other anions are left in solution. The precipitate is washed and then dissolved in dilute nitric acid and passed through another cation exchange column to remove the lanthanum. The effluent then contains only nitric acid and the P^{32} as phosphate. A small amount of phosphate, 25 micrograms per millicurie of P^{32}, is usually added in the evaporator to cut down losses by adsorption during the process. The dilute nitric acid containing the phosphorus is passed to the final evaporator, made of Vycor (96% SiO_2) glass, evaporated to dryness, and dissolved in 0.1 N hydrochloric acid. In order to remove any further traces of cations that might be present, the solution is again passed through a very small cation column containing Amberlite IR-120 resin, evaporated to near dryness, dissolved in 0.1 N hydrochloric acid, and passed through a small bed of highly-purified activated carbon. The product is then diluted, analyzed and assayed for dispensing.

SULFUR-35

Sulfur-35 is produced by irradiation of chlorine-containing compounds and at Oak Ridge National Laboratory the compound that has been used since the beginning of the program is potassium chloride. Several other important radioisotopes are obtained by irradiation of the same target material; e.g., chlorine-36 is produced by (n, γ) reaction on chlorine-35. Since the radioactivity of the potassium-42 produced by (n, γ) reaction with potassium is short-lived, it can be allowed to decay out before the target is processed. Small amounts of phosphorus-32 are also produced by (n, α) reaction on the chlorine.

The target (KCl) is dissolved in water, usually under carefully-controlled conditions so that the small quantity of stable argon-38 which is captured within the crystal can be salvaged when it is released as the crystals dissolve. During our first work on this procedure, it was thought necessary to oxidize the sulfur by adding small quantities of bromide or other oxidizing agents to the solution. It was later found that the sulfur is oxidized to sulfate without the use of oxidizing agents, presumably because it is present in the irradiated crystal in a highly-active chemical form. In the next step, the solution, properly adjusted for concentration, is passed through a column containing cation-exchange resin, Amberlite IR-120, and the potassium is quantitatively removed. The effluent contains hydrochloric acid and carrier-free sulfur-35 in the form of sulfuric acid, which is continuously passed to a small evaporator where the HCl is distilled off under a slight vacuum. The distillation is carried to dryness and the sulfur-35 that is found adsorbed on the walls of the flask is removed by boiling with a small quantity of 0.1 N hydrochloric acid. With the exception of small traces of phosphorus-32, the radiochemical purity of the resulting product is quite satisfactory. The traces of phosphorus-32 are either allowed to decay out, or are removed by passing the starting KCl through a column of aluminum shavings, which selectively takes up traces of phosphate. HCl^{36} is further purified by distillation through a small rectifying column, hydrochloric acid being collected as its constant boiling mixture at 20% strength. The specific activity of the resulting sulfur-35 is quite high—carrier-free for all practical purposes.

SHORT-LIVED FISSION PRODUCTS

The production of short-lived fission products is closely associated with iodine-131 production. In addition to the xenon-133 removed from dissolver off-gases, already mentioned in the iodine process, the dissolved uranium solution is a ready source of almost all of the other fission products. The uranium solution is transferred to another cell and the uranium is removed by batch extractors with a suitable solvent. Zirconium-95-niobium-95 is previously removed from the uranium solution before extraction by passing the uranyl nitrate solution slowly through a bed of silica gel. Over 90% of the Zr^{95}-Nb^{95} is taken up, presumably as a radio-colloid, on the surface of the silica gel. The silica gel can then be washed with reagents such as sulfuric and nitric acid to remove most of the contaminating fission products, and the Zr^{95}-Nb^{95} is eluted in 0.5 M oxalic acid. This is an example of an almost ideal type of process where a product is selectively adsorbed in relatively pure form. The other fission products are separated into groups of rare earths, alkaline earths, and ruthenium and cesium. The separation into groups can be accomplished by solvent extraction, ion exchange, or by precipitation techniques with added carrier. Two methods have been used for radioisotope work, the principal one being separation by cation exchange, using selective elution with ammonium citrate solution at various pH values. More recently, the rare earths group has been separated from the alkaline earth group by solvent extraction of the rare earths into tributyl phosphate solution. The alkaline earth group is further separated into its constituents of barium-140 and strontium-89 by elution with ammonium citrate on a Dowex-50 cation exchange column. The ion exchange method is practical if one does not have too much radioactivity per unit volume on the ion exchange column. Severe radiation damage results if more than one or two curies of an energetic beta-emitting radioisotope per cubic centimeter of ion exchange resin is placed on the column. The rare earths group is also further fractionated by ion exchange, this time the separation being divided into several main groups and then a refractionation of these groups is made to separate them into the individual components.[7] Yttrium is removed first from the rare earth column, followed by neodymium and prascodymium, then cerium, and finally residual strontium and barium contamination, traces of which which accompany the first rare earth group separation. Usually it is necessary to put the rare earth groups from this second fractionation on 4-foot-long cation exchange columns approximately one centimeter in diameter, steam-heated to 98°C, in order to give a very efficient fractionation into pure components. The results obtained on such columns indicate that they have as high as 500 theoretical plates for separation of the rare earth components; products of extremely high purity (99.9%) are obtained.

COBALT-60, KILOCURIE GAMMA SOURCES

The part of the program in which interest has been growing most rapidly during recent years is the production of kilocurie amounts of cobalt-60 and fission products, such as cesium-137, for use as teletherapy sources, radiographic sources, and sources for irradiation of chemicals, biologicals, and agricultural materials. Cobalt is an ideal material to use for production of gamma sources, since Co^{60} is produced by (n, γ) reaction by merely irradiating cobalt metal, which fortunately is monoisotopic, containing 100% cobalt-59. The neutron capture cross section is quite high, 34 barns, although the production rates obtained are somewhat lower than one would calculate, because of the shadowing in the nuclear reactor and the local depression of flux when large amounts of cobalt are placed in the reactor. The practical cross section for large-scale production is about 15–25 barns, depending upon a number of variables. The practical production rate that we find for cobalt-60 in a flux about 3×10^{13} n/cm^2/sec is about 5 curies Co^{60} per gram of cobalt per month.

The cobalt metal is prepared in various sizes, the most popular being small discs 1 cm and 2 cm in diameter by 1 mm or 2 mm thick. These sources may be placed in various configurations to form teletherapy sources or radiographic sources. Various other shapes and sizes used are cylinders ranging from 1 cm in diameter by 1 cm thick to very small pieces $\frac{1}{16}$ in. in diameter by $\frac{1}{16}$ in. thick. Canadian workers also use pellets 1 mm in diameter for cobalt irradiation. Various kinds of needles and wire have also been irradiated for special purposes, such as insertion into radium-type needles for medical use.

Since cobalt metal is subject to corrosion in moist air (and we have observed that corrosion is accelerated by radiation), the surface of the cobalt has been covered by a very thin electroplated coating of gold; more recently nickel has been used. The main purpose of such a coating is to prevent dusting and flaking of the cobalt during handling. Any Co^{60} to be used for radiography or other purposes must be encapsulated within sealed metallic containers. The various sizes and shapes of Co^{60} pieces are assayed in an especially-designed gamma chamber which has been calibrated carefully against cobalt pieces of the same sizes and shapes that were dissolved, aliquotes removed, and assayed by gamma counting against standards in the laboratory in a large argon-filled gamma chamber. Cross-checks were also made against standard radium sources. Using pieces of cobalt of various sizes calibrated for the geometry of each individual size and shape, the wafers or small cylinders can be assayed to ±5%. By combining various shapes and sizes of these capsules, kilocurie teletherapy and industrial radiation sources can be reasonably well-calibrated. The small pieces of cobalt rods and wafers can be put into almost any configuration for irradiation that is desired, which is especially useful for making special gamma sources for specimen irradiation.

GENERAL METHODS USED FOR MISCELLANEOUS PRODUCTS

In addition to radioisotopes produced on a fairly large scale, many other radioisotope preparations are made, most often involving the purification of high specific activity reactor-produced materials or certain important cyclotron-produced radioisotopes. Several examples are given below.

Iron-59 with a specific activity up to 10 curies/gm is produced by irradiating iron which has been electromagnetically-enriched to 50% Fe^{58}. After irradiation, some traces of impurities are present, notably Co^{60}. Purification is accomplished by extracting the iron as the chloride from 12 *N* HCl into dichlordiethyl ether, a solvent that is remarkable in that its extraction efficiency becomes greater at low concentrations.[8] The iron is back-extracted with dilute acid and extremely pure $Fe^{59}Cl_3$ is obtained in one or two cycles.

Carrier-free Sb^{125} is isolated from irradiated tin by a method typical of many used for radioisotope preparations.[9] The tin is dissolved in 4 *N* HCl and passed through a column filled with 30-mesh granular tin metal. The Sb^{125} is deposited in a thin layer on the top of the granulated tin column. The tin-Sb^{125} layer is removed, dissolved in HCl, and the antimony recovered by co-precipitation with CuS, followed by distillation of $SbCl_5$. The $Sn^{113}Cl_2$ is also recovered, the specific activity being changed only an insignificant amount by passage through the bed of tin metal.

Anion exchange resins are quite useful for purification of radioisotopes,[10] although this method must be used at fairly low radiation levels, since anion exchange resins are easily damaged by radiation. Cobalt-58 is formed by (n,p) reaction during irradiations to produce Ni^{63}. The nickel metal is dissolved in 12 *N* HCl and passed through a column of anion exchange resin (polystyrene-divinyl benzene type); Co^{58} is taken up on the resin and the pure nickel solution passes through; Co^{58} is eluted with water.[9]

Carrier-free Ca^{45} is produced by (n,p) reaction with scandium. A large amount of gamma-emitting Sc^{46} is present, making it desirable to use a procedure easily adapted to remote control. The separation method used is chelation and extraction with a solution of 0.5 *M* thenoyltrifluoroacetone (TTA) in benzene. With a pH of 4.0 in the aqueous layer, scandium is chelated and is extracted into the benzene layer. The water layer contains carrier-free Ca^{45}, which may be extracted into TTA-benzene at pH 9 and back-extracted into water for Ca^{45} purification.[11]

These general methods, or combinations of methods, are used for almost all radioisotope preparations. Co-precipitation, adsorption of radio-colloids, distillation, gasification, and chromatographic separations from adsorption media or cation exchange resins are also important methods in radioisotope production work.

CYCLOTRON-PRODUCED RADIOISOTOPES[12]

The cyclotron can be used in supplementing the supply of available radioisotopes, since the radionuclides produced in the cyclotron offer a wide range of energies and types of activities; they include neutron-deficient positron and alpha emitters.

The ORNL 86-inch cyclotron is an exceptionally good production instrument, having a steady internal proton beam in excess of one milliampere at an energy of 22 Mev and 2 milliamperes at 18 Mev. These high currents are made possible by the use of wide dees, a high-output ion source, and a radio-frequency oscillator with sufficient power to maintain extremely high dee voltage.

Grazing-incidence targets are designed to allow the beam to strike their surface at a very small angle to increase the effective beam area, thus reducing the power density to a manageable level.

During the past two years, some 20 elements have been bombarded for the production of millicurie to curie quantities of radioisotopes. Some typical ones are: 54 d Be^7 by $Li^7(p,n)$; 1.87 hr F^{18} by $F^{19}(p,pn)$; 310 d Mn^{54} by $Cr^{54}(p,n)$ from calutron-enriched Cr^{54}; low specific activity Mn^{54} by $Mn^{55}(p,pn)$; 80 d Co^{56} by $Fe^{56}(p,n)$; 270 d Co^{57} by $Ni^{58}(p,pn;$ $p,2n;$ $p,2p)$; 250 d Zn^{65} by $Cu^{65}(p,n)$; 17.5 d As^{74} from $Ge^{74}(p,n)$; 78 hr Ga^{67} by $Zn^{66}(p,2n)$; and 2.93 yr Po^{208} by $Bi^{209}(p,2n)$.

These isotopes were chosen for initial investigation because of their suitability for medical, biological, or industrial application.

LONG-LIVED FISSION PRODUCTS

Cobalt-60 is an extremely good source of radiation, having two gamma rays per disintegration (1.17 Mev and 1.33 Mev), and it can be obtained in extremely high specific activity (a maximum of 70 curies per gram obtained up to this time). However, the only moderately long half-life of 5.3 years and the fact that it is not a true by-product of the nuclear reactor suggest that it may eventually become of secondary importance, especially as a source of industrial gamma radiation as compared with fission-product cesium-137. Cesium-137 has a half-life of 33 years, gamma radiation of 0.67 Mev from its short-lived barium-137 daughter, and is a true by-product, being a fission product which occurs in relatively high yield (6%). Therefore, much attention has been devoted to the separation of large quantities of cesium-137 from reactor wastes. Other fission products in the reactor wastes can be separated at the same time if there is a market for these materials. Other long-lived radioisotopes in wastes which are of greatest interest are strontium-90, cerium-144, ruthenium-106, europium-155, prnmethium-147, and technetium-99.

In 1948, work was started on the development of industrial-type processes for economically removing these important fission products from reactor wastes and fabricating the radioactive material into suitable sources. The processes have been taken through the laboratory and semi-works stages, during which 1000-curie quantities of Cs^{137} and 100-curie quantities of Sr^{90}, Ce^{144}, and Ru^{106} have been produced. A simplified flowsheet for the process which has been developed is given in Fig. 13. Most emphasis has been placed on the cesium separation and purification process, since it was deemed most valuable of all the fission products. During the development period, many types of processes were tried for the separation of cesium. Since cesium has relatively few insoluble compounds, the first methods tried were all based upon isolating cesium by removing all the other elements in the solution by ion exchange or precipitation. However, many of the reactor wastes are contaminated to a very great extent with Group I elements, such as sodium and potassium. Therefore, a more direct means of isolating cesium was necessary. Various precipitants were tried, such as silico-tungstic acid, phospho-tungstic acid, ferrocyanides, and other compounds of this type which have been long known and used for analytical purposes. However, most of these were not well-adapted to industrial-type processes and the most promising process appeared to be the one in which advantage is taken of the co-crystallization of the relatively insoluble cesium alum with potassium or ammonium alum.[13] By saturating the solution with the potassium alum (or as now used, ammonium alum) at approximately 80–90°C, and then cooling to bring down a crop of alum crystals, it was found that the cesium was quite selectively co-crystallized with the batch of crystals that was brought down, more than 99% of cesium being removed from the solution.[14] By decanting off the supernatant liquor, another batch of material containing cesium could be put into the crystallizers and the previous crystals redissolved. More ammonium alum was added to saturate the solution at 80°C, and a new crop of crystals grown, this time including all the cesium that has been brought down during the first step plus the newly-added cesium.

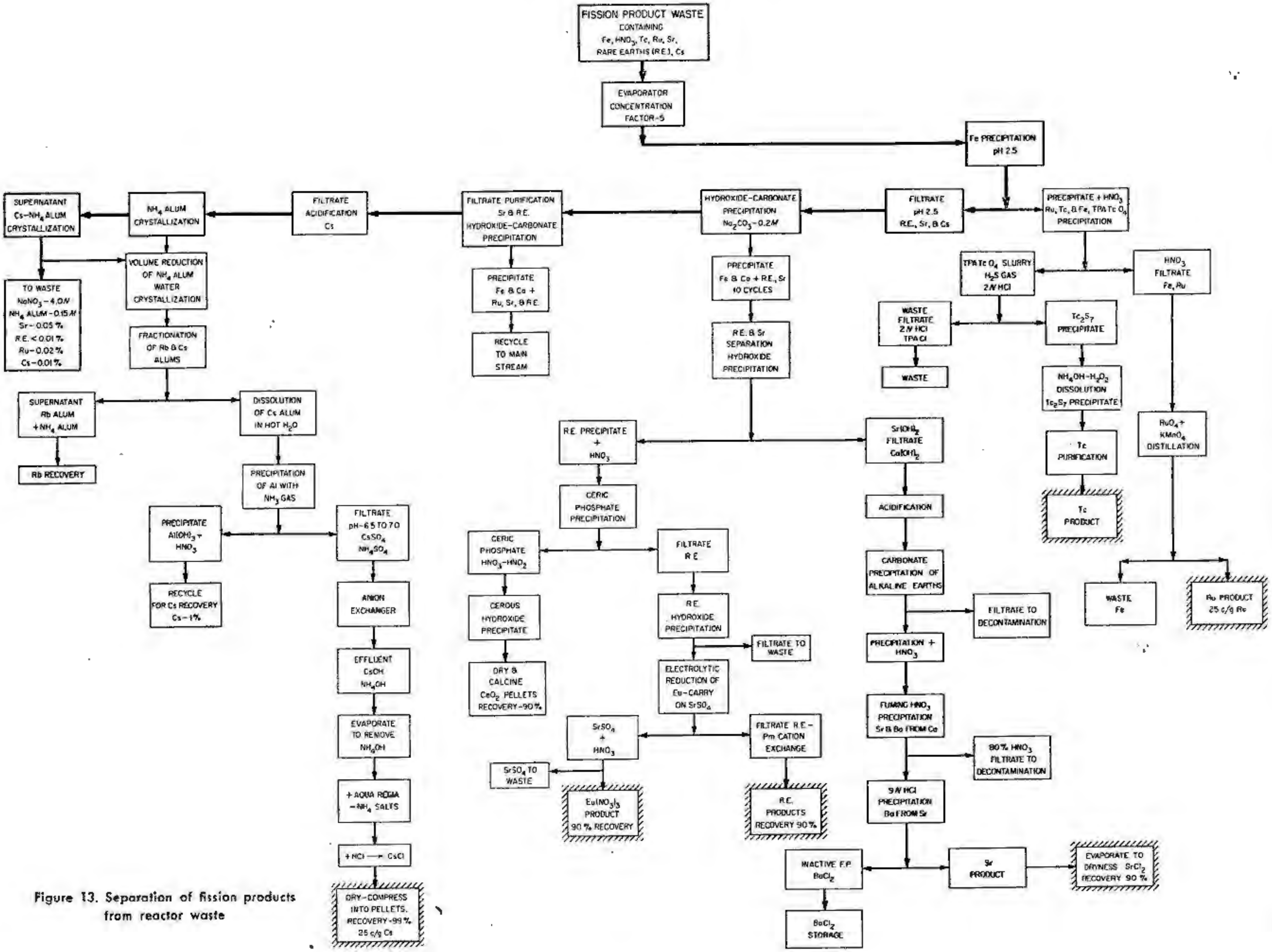

Figure 13. Separation of fission products from reactor waste

Successive crystallizations of this type are made and the crop of crystals grown after each crystallization becomes progressively richer in cesium. This procedure can be followed until cesium losses in the supernatant liquor become higher than desired. In the semi-works equipment designed for this process, a number of crystallizers are used, some of which handle the supernatant liquor for recrystallization and recovery of small amounts that have been passed into the supernatant liquor; others are used to concentrate the cesium by dissolving the alum crystals in pure water and then growing smaller batches of crystals to get the cesium into progressively smaller volumes of liquid. If enough cesium is accumulated, it is possible to precipitate pure cesium-ammonium alum. Cesium-137 in this process comes down almost radiochemically pure ($> 99\%$) after one crystallization. Only insignificant amounts of cerium and ruthenium-106 superficially contaminate the first batches of crystals. However, some inactive fission products, such as rubidium, accompany the cesium and must be removed by selective, controlled crystallizations near the end of the process; by these successive crystallizations, one can enrich certain fractions in cesium and others in rubidium. At all stages of the process, the various fractions can be recycled to an appropriate previous point in the crystallization cycle and in this way, losses can be easily kept to a minimum.

The crystallization process is also well adapted to removing cesium, either before or after the other fission products are removed from a fission product mixture. However, in most cases it is probably desirable to remove the other constituents, such as alkaline earths, ruthenium, and rare earths, before the cesium is removed.

Because of its complex chemistry, ruthenium-106 has always been one of the most difficult of the fission products to cope with in fission mixtures. The procedure developed for separating ruthenium from fission product waste consists of controlled homogeneous precipitation of iron at a pH of 2.0 by the hydrolysis of urea, or the controlled addition of very small quantities of ammonia gas into the mixture with high-speed agitation, producing a granular precipitate of hydrated iron oxide which co-precipitates over 90% of the ruthenium and technetium in the solution. This fraction is then removed for storage and combining with other similar fractions from succeeding batches passing through the plant.

All second-stage processing is done by combining portions of similar fractions going through the plant until a large enough quantity is accumulated to process in the later steps in the process, thereby giving a more economical operation.

The effluent from the ruthenium-technetium-iron hydroxide precipitation contains cesium, alkaline earths, and rare earths. The next step is to remove the rare earths as the hydroxides by precipitation from a carbonate-free solution, thus allowing the alkaline earths to pass into the filtrate. The rare earth precipitation is made by passing airborne ammonia gas into the solution with high-speed agitation. The rare earth group precipitates quite completely, there being a plentiful supply of inactive neodymium, lanthanum, and other rare earths to carry down all of the cerium, promethium, and europium that are present. It may be well to remark at this point that with procedures of this kind, where one is dealing with large quantities of fission products, classical methods are usually quite satisfactory, since high enough concentrations of the fission products are present for ordinary precipitations. This is in contrast to the early radiochemical work where only minute concentrations of fission products were available.

After removal of the rare-earth group as the hydroxides,* the filtrate containing cesium and the alkaline earths is treated with sodium carbonate to precipitate the alkaline earths, mostly inactive barium and radioactive strontium-90. After this precipitation, there remains only cesium in the effluent for removal by the previously-described alum crystallization process.

The constituents of the main groups are further separated and purified in the following manner. The iron-technetium-ruthenium precipitate is dissolved and the technetium is selectively precipitated as the tetraphenyl arsonium (TPA) pertechnetate.[15] The effluent containing iron-ruthenium is re-precipitated as $Fe(OH)_3$ and is dissolved in concentrated sulfuric acid, treated with potassium permanganate and the ruthenium is distilled, as ruthenium tetroxide, into hydrochloric acid or nitric acid containing sufficient quantities of hydrogen peroxide to reduce the ruthenium to the Ru^{+3} state. The ruthenium chloride solution is then used for the preparation of large sources.

Ruthenium is one of the few fission products that can be easily prepared in metallic form as an electroplated metallic source. The ruthenium chloride (or nitrate) solution is treated with red fuming nitric acid and/or hydrochloric acid, depending upon the starting solution, to produce ruthenium nitroso-chloride. Very satisfactory ruthenium electroplating can be done from a bath of ruthenium nitroso-chloride, producing smooth metallic surfaces. Sources have been made by electroplating ruthenium-106 having a specific activity of approximately 40 curies/gm onto copper surfaces, in sufficient thickness to indicate that 10,000 curies could probably be plated onto an area of one square foot. Immediately after electroplating the radioactive ruthenium, the plated source is transferred to a wash solution of sulfurous acid and then placed in a ruthenium nitroso-chloride bath, where a flash covering of inactive metallic ruthenium is placed over the active ruthenium. The plate is then rewashed and placed in a silver cyanide

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* Or, the hydroxide and carbonate groups may be brought down together for later separation, as shown on the flowsheet.

TABLE 2. Characteristics of Separated Fission Products

	Cs^{137}	Sr^{90}	Ce^{144}*	Ru^{106}*	Pm^{147}	Eu^{155}	Co^{60}†
Stable nuclides also present	Cs^{133} Cs^{135}	Sr^{88}	Ce^{140} (Ce^{142})	Ru^{99} Ru^{101} (Ru^{102}) (Ru^{104})	None	Eu^{153}	Co^{59}
Active nuclide, weight/kilocurie	12.7 gm	5.1 gm	0.31 gm	0.3 gm	1.07 gm	0.87 gm	0.88 gm
Active + stable, weight/kilocurie	31.7 gm	9.5 gm	~2 gm	~26 gm	1.07 gm	~5 gm	20 gm
Weight/kilocurie compound	40.0 gm (CsCl)	11.2 gm (SrO)	~2.5 gm (CeO_2)	26 gm (Metal)	1.25 gm (Pm_2O_3)	~64 gm (Eu_2O_3)	20 gm (Metal)
Weight/kilocurie, obtained experimentally	39.8 gm (CsCl)	~70 gm (SrO)	2.6 gm (CeO_2)	30 gm (Metal)	~7 gm (Pm_2O_3)	—	20 gm (Metal)
Practical volume/kilocurie	15 cm^3 (CsCl)	~23 cm^3 (SrO)	1.3 cm^3 (CeO_2)	2.5 cm^3 (Metal)	~1 cm^3 (Pm_2O_3)	—	2.25 cm^3 (Metal)

* After one year of decay.

† For comparison purposes, using 50 curie/g material.

electroplating bath and plated with a thin coating of silver. The plate is finally slipped into an envelope made of thin stainless steel and sealed by welding. The stainless steel envelope has handling projections for placing the source in carriers and inserting it into radiation devices.

The technetium precipitate of phenyl arsonium pertechnetate is further purified by several re-precipitations as the pertechnetate and then as the technetium sulfide, followed by dissolution in ammonia and hydrogen peroxide to produce ammonium pertechnetate solution as a final product.

The rare earth group contains three main constituents, cerium-144, promethium-147, and europium-155. Of these constituents, cerium-144 is present in by far the largest quantity. Since it provides most of the radiation in moderately old fission product mixtures, it is important to separate it from promethium and europium so they can be further purified in lightly-shielded equipment. Several methods are available for removing cerium-144; namely, selective extraction from concentrated nitric acid into tributyl phosphate solutions, or a carefully-controlled precipitation of ceric phosphate in acid solution. Cerium may also be separated from the other rare earths as a ceric iodate; however, this process produces a relatively large quantity of the very voluminous ceric iodate precipitate in which large losses of europium-155 occur. Furthermore, the conversion of the iodate to more usable forms of cerium oxide or hydroxide is very difficult because of the large quantity of iodine to be removed. However, by close pH control, cerium can be removed with about 90% efficiency by precipitating as the ceric phosphate. If the cerium is then reduced to the Ce^{+3} state, it can be re-precipitated as the hydroxide and thus separated from phosphate. The next step after dissolving the cerium hydroxide is to re-precipitate cerium as the oxalate, in which form it can be ignited to ceric oxide. Ceric oxide can either be packed by vibration into thin stainless steel tubes which are sealed by welding, or combined with silica and other ceramic ingredients and fused into thin layers on iron plates, which are then encapsulated within thin stainless steel jackets and sealed by welding. Experiments are still in progress to determine the best way of fabricating cerium sources. However, it appears that it will be necessary in any case to fabricate the cerium in fairly thin layers to prevent self-absorption of the radiation, since only a fraction of the Ce^{144} radiation is hard gamma radiation.

The fractions of rare earths containing promethium and europium are purified by taking up on a steam-heated cation exchange column and selectively eluting with ammonium citrate solution, as previously noted. Radioactivity "peaks" are obtained indicating which fractions are rich in europium and promethium; after separation, these fractions are again fractionated on ion exchange columns to produce products of the desired purity. Europium may be further purified by reducing to the Eu^{+2} state by electrolytic reduction or passage through a zinc metal reductor, followed by co-precipitation with barium sulfate, or direct precipitation if the quantity of Eu is large enough. By oxidation of the europium to Eu^{+3}, it can be separated from barium carrier and sulfate. Further purification may be made by reduction with sodium amalgam and re-extraction from the amalgam as the hydroxide, finally precipitating as the oxalate.

The characteristics of the fission product preparations are shown in Table 2.

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The Production of Beta-Ray Sources for Industrial and Clinical Use

By P. E. Carter and L. C. Myerscough,* UK

Although sealed gamma-ray sources, containing at first the natural radioelements radium and radon, and recently suitable artificial radioisotopes, have been used for industrial radiography and in radiotherapy since early in the century, it is only during the last few years that beta-emitting sources of robust construction have been produced for industrial and clinical purposes.

In industry, the use of sources containing beta-emitters of widely differing energies, and made to a variety of designs is developing rapidly. Perhaps the most important industrial applications at present are the continuous absorption thickness gauging of a wide range of products, varying from thin paper or plastic to sheet metal, and the measurement of the thickness of metallic or plastic coatings applied to metal by the reflection or back-scattering technique. Beta sources are also being used extensively to disperse electrostatic charges which accumulate on textiles during manufacture, and to eliminate electrostatic effects which may occur during the production of paper and plastic sheet. Sources containing tens or hundreds of millicuries suffice for thickness measurements and the dispersal of static charges, but the development of radiation chemistry and radiation sterilisation on the commercial scale may soon require beta-ray sources at the multicurie level.

In radiotherapy, beta-ray sources containing radium have been used for many years in the treatment of diseases of the skin and eye. The construction of these radium sources proved insufficiently robust for hospital use, while the energetic gamma-radiation increased manipulative hazards and was clinically undesirable. The replacement of radium by radioisotopes such as strontium-90 and ruthenium-106 has been a major advance. Such sources contain only up to 50 mc but now that fission product radioisotopes have become plentiful, the use of multicurie sources, simulating the clinical effect of particle accelerators, is proposed as a line of development.

Although several hundred beta-emitting nuclides have been identified, the number whose physical characteristics render them suitable for industrial and medical use is surprisingly small. The properties of the six most important nuclides used as sources of beta radiation are shown in Table 1.

Because of the cost of replacement and the inconvenience of working with a rapidly decaying source, it is desirable to use a nuclide having a half-life of not less than about one year. On the other hand, nuclides of very long half-life are less useful as they are not readily available in large amounts or at acceptable specific activities. The most widely used beta emitter is strontium-90, which has the convenient half-life of twenty years. Several of these beta-emitting nuclides are suitable for exciting phosphors, and may replace alpha-emitters such as radium which have been used for many years. Discussion of this subject, however, is beyond the scope of the paper.

With the exception of thallium-204, the nuclides listed are all fission products which can be extracted without the addition of isotopic carriers. Although inactive isotopes of these beta emitters are also present in fission products and the total weight per element may be considerably more than that of the active isotope, the specific activities obtainable are still in all these cases very high, e.g., 50–100 curies strontium-90 per gm strontium. In the case of thallium-204, which is produced by radiative capture, the maximum specific activity readily attained is much lower, now about 1 curie per gm thallium.

The separated nuclides for source preparation are required to be of a high standard of chemical and

TABLE 1

Nuclide	*$T_{\frac{1}{2}}$, years*	*Maximum beta energy Mev*	*Gamma emission*	*Beta half-thickness, in Al, mg/cm²*
Promethium-147	2.6	0.22	None	5
Krypton-85	9.4	0.7	Weak	24
Thallium-204	4.0	0.76	None	27
Strontium-90	20.0	2.2, from Y^{90} daughter	None	56
Cerium-144	0.8	3.0, from Pr^{144} daughter	Present	110
Ruthenium-106	1.0	3.5, from Rh^{106} daughter	Present	270

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radiochemical purity. The specifications for chemical purity have become increasingly severe as the source strengths demanded have risen, for impurities may have an adverse effect on the operation of the preparative processes and on the quality of the product, particularly as regards specific activity. The radiochemical purity of the nuclide is normally required to be of the order of 99%, excluding of course any short-lived daughter product. It is usually necessary to work with well-aged mixed fission products in which the shorter-lived radioisotopes have decayed to a satisfactorily low level. For example, strontium-90 is frequently required substantially free from strontium-89, which has a half-life of 53 days and is initially present at a higher activity level than the useful isotope, and an ageing period of 18 months to two years is required. In the case of ruthenium-106, the fission product ruthenium-103, half-life 40 days, is an unwanted contaminant. Radioactive impurities in the final source may affect the efficiency of operations; for example in thickness gauging, they may necessitate more frequent adjustment of equipment to allow for decay, or they may increase the radiation hazard so making increased shielding necessary. It should be borne in mind, in connection with this latter point, that even with the pure beta emitters, sources emit penetrating radiation (X-rays) which cannot always be ignored. This is particularly so with an energetic source such as Sr^{90} (+ Y^{90}), commonly enveloped in an inactive metal of high atomic number such as silver.

It will be observed that the maximum beta energies of the nuclides listed cover a useful range from 0.22 Mev for promethium-147 to 3.5 Mev for ruthenium-106. In the case of strontium-90, cerium-144, and ruthenium-106, the high energy emissions are from the daughter products yttrium-90 ($T_{1/2}$ 61 hours), praseodymium-144 ($T_{1/2}$ 17.5 min) and rhodium-106 ($T_{1/2}$ 30 sec). The choice of nuclide for a particular application is mainly governed by the beta energy required but considerations of half-life and acceptability of primary gamma emission are also involved.

The choice of chemical form for the radioactive component is governed by the following considerations:

1. Method of preparation to be used.

2. Radiation stability, e.g., nitrates or organic compounds are usually unacceptable.

3. Specific activity. This is sometimes required to be the maximum attainable and therefore the elementary form of the nuclide is usually needed. If the use of a compound rather than the element is desirable on other grounds, then evidently one containing a high proportion of the element is to be preferred.

4. Solubility and chemical stability. To minimise the consequences of any failure of the inactive sheathing material, the active component should be in a stable chemical form and have a low solubility in water or in other liquids which might come into contact with the source. Low volatility at elevated temperatures is also desirable to minimise dispersal in event of fire.

With all these considerations in mind, it is the usual practice to use the elementary forms of ruthenium, thallium and of course krypton, strontium as carbonate, and cerium and promethium as oxides.

The toxicity of these nuclides varies considerably, strontium-90 being the most hazardous; the maximum permissible level of this in the body is only 1 microcurie. As sources normally have an activity at least at the millicurie level, it is evidently a matter of great importance that the source is adequately sealed.

The manufacturer has therefore to produce a layer of radioactive material, sufficiently thin to minimise self-absorption; for most applications this must be of uniform thickness. This radioactive layer has then to be completely enveloped and sealed in a sheath of inactive material, so that there is no danger of dispersal of the radioisotope under normal working conditions. The construction of the source must also be sufficiently robust to withstand accident. The permissible thickness of the sheathing material will evidently depend upon the maximum beta energy of the nuclide, and a compromise must be sought between safety and reasonable efficiency of beta emission.

METHODS OF MANUFACTURE

The production methods which have been considered in the United Kingdom fall into four main groups:

1. A source is made of inactive material, which is then activated by irradiation in a nuclear reactor. This method has the great advantage of demanding very little manipulation of active material, but since few of the useful beta emitters are made by (n,p) or (n,γ) reactions, its application is limited. Useful sources of phosphorus-32 ($T_{1/2}$ 14.3 days; β_{max} 1.7 Mev) have been made using polythene containing 20 wt % of red phosphorus, which may be irradiated to give a maximum dosage rate at the surface of about 13 rep/min; the intensity attainable is limited by the stability of the plastic under neutron bombardment while the short half-life of this nuclide severely limits its application. Sources containing thallium-204 enveloped in aluminium could be made similarly, though the cost of irradiation in this form might prove rather high compared with bulk irradiation of thallium metal or thallous oxide.

2. Radioactive material is introduced into a thin-walled metal tube, and sealed off. Low-intensity sources of strontium-90 for applications where uniformity of loading is unimportant have been made by evaporating a solution of the radioactive salt in a thin-walled stainless steel tube. Alternatively energetic beta-emitting nuclides available at high specific activity have been adsorbed onto clays of high exchange capacity, e.g., montmorillonite, and the ac-

tivity fixed by firing at 1000–1100°C. The product, from which the nuclide cannot be extracted even by dilute mineral acids, may be packed in thin-walled aluminium tubing.

Similarly, beta-sources containing krypton-85 have been made by filling carefully degreased, thin-walled aluminium tubing with the purified gas, and sealing the ends by cold welding.

3. Sources of most beta-emitters may be made by depositing a thin film of the element or of a convenient compound on a backing plate, and then protecting it by applying a thin covering layer, usually of metal. The films may be made either by evaporating a solution or suspension of a salt of the nuclide, or, if the properties of the metal are suitable, by electrodeposition, by vacuum deposition or by sputtering. The protective covering layer may either be a metal deposited similarly, or it may be a metallic foil secured by adhesive or solder. Sources made by evaporation and covering with metallic foil are in common use, but it is difficult to obtain a reasonably uniform coating. The covering foil is usually secured to the backing plate only around its edges, and not to the active area of the source. Since the evaporated layer is unlikely to be bonded firmly to the backing metal, there is some danger of dispersal of particles of radioactive material if the protective foil is accidentally punctured.

The electrodeposition method, which was used in classical radio-chemistry to prepare beta-ray sources of radium-D (lead-210), has been applied successfully to the routine production of thallium-204 sources for industrial use. Using metal of specific activity 60–70 mc/gm, sources are made containing up to 1 mc/cm^2 in a variety of shapes; the active thallium deposit is protected by electrodeposits of copper and cadmium.

4. The fourth method comprises making (e.g., by powder metallurgy) a metallic matrix in which the radioactive material is uniformly dispersed, and enveloping this in a sheath of inactive metal. The composite billet is extended by forging or rolling to the required dimensions. Sources made by this technique combine uniformity of distribution and robust construction, with consequent freedom from leakage even under unfavourable conditions. They are usually made of corrosion-resisting metals, such as platinum, silver, or gold, and a wide variety of radioactive nuclides can be incorporated. The protective face, which for low energy beta-sources such as promethium-147 must be as thin as 0.005 mm, is an integral part of the strip of metal. If it is scratched or abraded accidentally, only a minute proportion of the radioactive material is exposed; even this is fixed firmly in its metallic matrix, and is not easily dispersed. Since the radioactive material is sealed effectively and permanently, it is considered to be unnecessary to make routine periodical tests for external contamination or leakage on sources after installation.

Using fission products of high specific activity, sources containing up to 0.3 curie/cm^2 have been made already, but further development should enable the intensity to be increased to several curies/cm^2. In general this method is most successfully applied to the production of sources in the form of strips which are made conveniently by rolling; the manufacture of sources of small area, and particularly of circular shape is not always possible. Compared with most of the methods reviewed, this one requires somewhat more elaborate equipment; it is, however, very well suited to routine production of large numbers of similar sources. Examples of these are given in Table 2 and Figs. 1–3. In order to achieve the very high standard of sealing demanded by the tests described in the next section, it is most important that tests for external contamination of workpieces and of metal working machinery be carried out at every stage of the process.

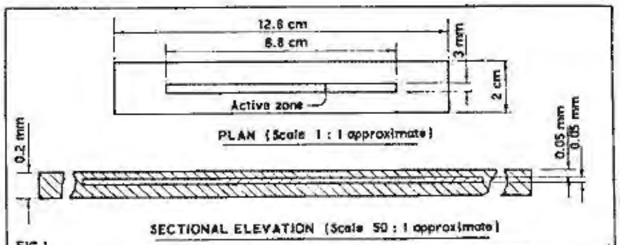

FIG. 1

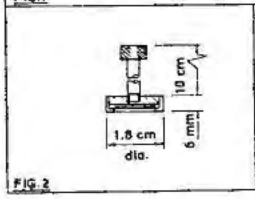

FIG. 2

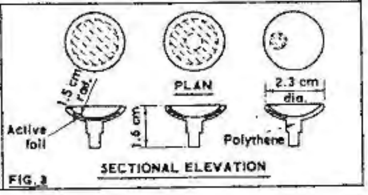

FIG. 3

CRITICAL EXAMINATION AND TESTING OF BETA-RAY SOURCES

Sources manufactured at the Radiochemical Centre are subjected to the following tests before issue:

Emission

The absolute calibration of sources is made with an extrapolation chamber; in routine production, sources are compared with a standard of the same nuclide using a small ionisation chamber with thin window and variable gain control. Alternatively strip sources are compared conveniently with an empirical standard using a larger ionisation chamber of the type incorporated in a thickness gauge.

Uniformity of Distribution

A small ionisation chamber with a narrow window may be used to scan a source, but autoradiography is often more convenient and furnishes a permanent record.

Except with the weakest sources, a process type film is to be preferred to an X-ray emulsion, while with the more intense sources, the use of contact printing paper permits a convenient exposure time.

TABLE 2. Typical Beta-Sources in Current Production in the United Kingdom for Industrial Use

Nuclide	*Type*	*Active dimensions*	*Screenage, mg/cm²*	*Strength, mc*	*mc/cm²*	*Uses*
Promethium-147	Silver strip sandwich with platinum protective face. Inactive surround	Length 5–10 cm Width 0.3–1.25 cm	5	5–20	0.5–1	Absorption thickness gauging of material up to 10 mg/cm², e.g., thin paper, plastic sheet
Thallium-204	Electrodeposit on copper strip, protected by deposits of copper and cadmium	Length 2–30 cm Width 0.5–2.5 cm	15	up to 80	up to 1	Absorption thickness gauging of material up to 120 mc/cm², e.g., paper, plastics, metal foil. Back-scatter thickness gauging of metallic coatings. Static dispersion
Strontium-90	Silver strip sandwich with inactive surround (Fig 1)	Length 2.5–30 cm Width 0.3–2.5 cm	30–50	up to 500	up to 250	Absorption thickness gauging of thicker sheet-materials and metals up to 500 mg/cm². Static dispersion
Cerium-144 Ruthenium-106	Silver strip sandwich with inactive surround (Fig 1)	Length 2.5–30 cm Width 0.3–2.5 cm	50	5–20	1–6	Absorption thickness gauging of metals up to 700–1000 mg/cm²

For Medical Use

Nuclide	*Type*	*Active dimensions*	*Screenage*	*Strength*		*Dosage, rate of surface*
Strontium-90	Plaque. Silver sandwich element mounted in silver case (Fig. 2). Circular or square	Area 1–4 cm²	100	5–40	2.5–10	100–350 rep/min
Strontium-90	Eye applicator. Silver foil elements of various shapes mounted in polythene (Fig. 3)	Area 0.3–2.5 cm²	60	0.5–7.5	1.5–3	100–180 rep/min
Ruthenium-106	Plaque. Silver sandwich element mounted in silver case (Fig. 2). Circular or square	Area 1–4 cm²	100	5–40	2.5–10	180–600 rep/min

The beta-emission from sources containing about 1 mc/cm² of the more energetic beta-emitters excites a fluorescent screen sufficiently brightly to be easily visible under photographic darkroom illumination, while the shape of sources of 5 mc/cm² or more can be observed in normal artificial light. This property provides a ready means of locating the active area of a source, and of detecting gross lack of uniformity.

External Contamination

All faces and edges of the source are swabbed with solvent. The removable activity should not exceed 0.01 microcurie.

Activity Extractable by Water (Immersion Test)

The source is immersed for eight hours in distilled water at 50°C; the activity of the extract should not exceed 0.05 microcurie.

CONCLUSION

Although up to the present beta-ray source strengths have barely reached the curie level, several of the methods reviewed could be applied to the manufacture of much stronger sources, containing hectocurie or even kilocurie amounts.

Mixed fission products are available even today in very substantial quantities, while methods of separating and purifying the individual nuclides on a large scale are under investigation.

Gamma-emitting sources are being made already at the kilocurie level, and higher intensities are envisaged. As uses are found for beta-ray sources at these high levels of activity, there is likely to be concomitant development in their manufacture and application.

Separation of Carrier-free Isotopes by Diffusion Methods

By K. Samsahl and K. Taugböl,* Norway

I. INTRODUCTION

Separation of carrier-free radioactive isotopes by means of diffusion from solid target materials seems in some cases to be convenient and is therefore here tried for the two isotopes of great medical importance, iodine-131 and phosphorus-32.

The diffusion methods render possible high specific activity as the target material in many cases can be used repeatedly, and a minimum of other chemicals are needed.

In the present two cases few working stages and simple apparatus are typical advantages.

II. DIFFUSION OF RADIOACTIVE IODINE-131 FROM IRRADIATED TELLURIUM DIOXIDE

1. Irradiation Material

The present method consists in dry distillation of the iodine from tellurium dioxide under vacuum at elevated temperatures. Owing to the fact that some of the possible iodine compounds formed by irradiation require a rather high temperature for distillation, elementary tellurium with its low melting point (452° at 760 mm) was considered unsuitable. Of the oxygen compounds, tellurium dioxide, only, is stable at temperatures above 400°C. Its melting point was determined to be approximately 735°C, and the vapour pressure below 700°C negligible. In addition it is nonhygroscopic and therefore seemed to us to be the best starting material.

Since commercially available tellurium oxide was found to contain a rather high degree of impurities undesirable for our purpose, we prepared our own irradiation material from exceedingly pure tellurium. Hydrated tellurium dioxide, which is amphoteric, was precipitated from hot acid solution by adjusting the pH to 6.2 with ammonia, when an almost 100% yield was obtained. After standing overnight, the solution was filtered and the precipitate washed well with distilled water, dried, and then heated to 700° for half-an-hour.

Small amounts of ammonium chloride, when retained in the tellurium dioxide, cause reduction to elementary tellurium on heating.

On heating to about 520°C the colourless, crystalline tellurium dioxide turns pale-yellow and becomes successively darker as the temperature is raised.

It was found that no iodine distilled below 500°C, even after prolonged heating, (about two hours) at 2×10^{-2} mm pressure, but on heating above 550°C iodine begins to distill. This may be due to changes in the crystal structure of tellurium dioxide at this temperature.

Tellurium dioxide as a fine powder, if not exceedingly pure, showed a marked tendency to sinter at temperatures above 600°C. The size of the particles does not seem to affect the distillation rate to an extent of practical importance.

2. Apparatus and Operational Procedure

An apparatus as shown in Fig. 1 was used. It is made of quartz with the exception of the ground joint, *e;* the bulb, *f;* and the two-way stopcock, *g;* which are made of Pyrex glass. The flask, *a*, has a volume of about 300 ml and is provided with a ground joint, *b*, in the bottom of a short, wide tube, *c*. *d* is a freezing trap. The connexion between *c* and *e* consists of a thick rubber tube.

The connexion to the vacuum line can be broken by pulling out the ground joint cone at *h*. A two-step oil pump and a water aspirator can alternately be brought into use. The flask *a* is filled and emptied through *b*. The tube between *a* and *d* is insulated with asbestos tape, bound with resistance wire and finally lagged with asbestos yarn. Thus the tube can be electrically heated up to 700°C. The other circuit, *c2* in Fig. 1, is made in a similar way. A special problem was the sealing at *b*, as the combination of high temperature and vacuum which is necessary here, rendered the use of all sorts of greases known to us, including graphite, impossible. The valve

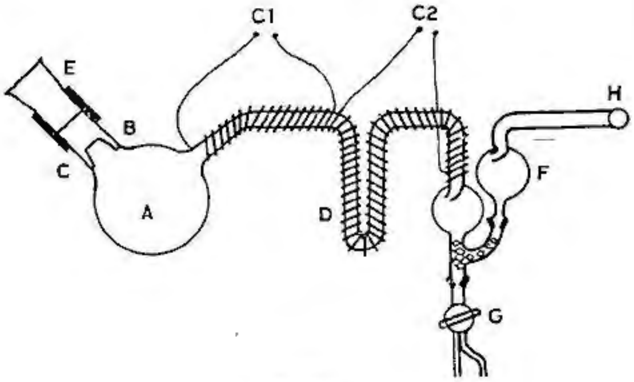

Figure 1

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[*] Joint Establishment for Nuclear Energy Research, Kjeller near Lilleström.

shown in Fig. 2 was therefore constructed. With this arrangement the vacuum seal is moved to the cold part of the apparatus, but a glass disc, *a*, can be pressed tightly against the grounded edge of the cone by means of a strong spring, thus preventing the escape of vapour into the valve.

The stopcock, *b*, is prepared in a special way. A groove is ground around the middle of the cone with an extension to one side. In one position of the cone this coincides with a small hole, *c*, in the socket and thus opens to the atmosphere.

A thin stainless steel wire is fastened at one end to the cone, passed inside the glass tube, *d*, and fastened at the other end to the lower part of the tube. By turning the stopcock, the disc can be lowered and raised. It opens to the atmosphere when the disc is in a particular elevated position. The disc and glass tube, *d*, are connected flexibly by a short tube, or plug, made of heat resisting material such as silicone or neoprene rubber. This allows the disc to fit closely to the cone.

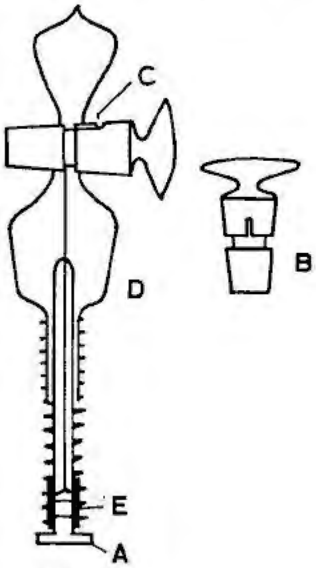

Figure 2

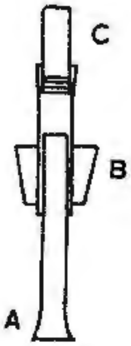

Figure 3

The apparatus is mounted in a frame which can be rotated over to the left, thus allowing the contents to be emptied through *b*. The powdered tellurium oxide is filled directly back into the irradiation can by means of the device shown in Fig. 3. The female ground joint, *a*, fits to the cone, *b*, in Fig. 1. The rubber stopper, *b*, fits in the opening *e* (Fig. 1), and the can, *c*, sticks in the polyethylene tube. The filling device used is described in an earlier report,[1] together with the method for remote control and opening of the irradiation can.

When the apparatus is turned into the horizontal position, the flask, *a*, is inserted into a furnace (Heraeus TiT) equipped with a pyrometer.

A Dewar flask filled with liquid air can be raised and lowered under the freezing trap, *d*, Fig. 1, by an elevator, all manipulations being done outside the lead wall. The elevator consists simply of a platform connected to the operating handle by wires passing over a pulley system and fixed in any desired elevation.

Asbestos covers are fastened to the top of flask *a* and trap *d*, thus providing heat insulation to the furnace and Dewar flask, respectively. After filling of the apparatus with irradiated tellurium dioxide, 50–150 gm, the trap *d*; is cooled in liquid air and the furnace and circuit 1 are switched on, the heating being regulated by means of variacs. The temperature in the flask and tube is kept between 680 and 700°C for 2–4 hours; the pressure maintained at about 2×10^{-2} mm with an oil pump. The iodine activity now appears in the first part of the trap. A suitable volume of sodium bisulphite solution is drawn up through the stopcock, *g*, by means of the water aspirator. By heating flask *a*, and then in turn the tube and the trap by the circuits *c*1 and *c*2, respectively, the activity is carried over into the solution by a gentle stream of air drawn through the apparatus from *e* to *h* and trapped in the solution.

The activity is now in a volume of only a few ml. The solution is run out through stopcock, *g*, directly into a distillation flask, and purified by distillation as described for the previous method.[1] Up to the present time the method has not been used in routine production, but single runs have shown very high yields, even after a processing time of only a few hours. Radioactive assay of the residue in flask *a*, showed no iodine activity. The activity collected in the trapping solution proved to be almost pure iodine-131.

III. DIFFUSION OF RADIOACTIVE PHOSPHORUS-32 FROM IRRADIATED SULPHUR

1. Experimental

The introduction of a suitable wetting agent has made possible the extraction of radioactive phosphorus from powdered sulphur into boiling water.

The course of extraction is found to be strongly influenced by the transition from rhombic to monoclinic sulphur (transition point 95.5°C), and the time for the extraction, about 1½ hours is assumed to coincide with the transition time under these conditions.[2] Figure 4 shows the extraction yields from

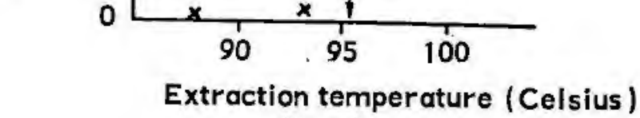

Figure 4

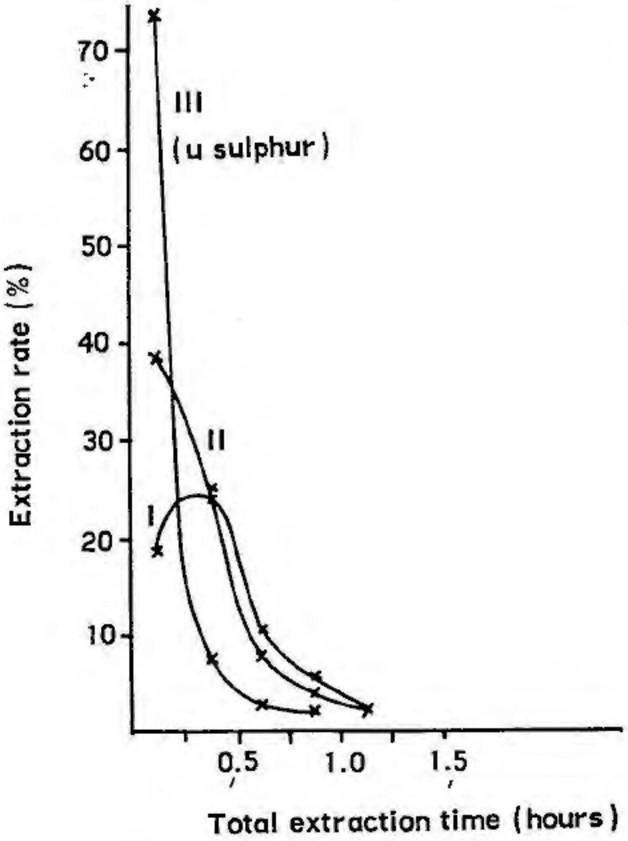

Figure 5

equal amounts of sulphur heated in water (without stirring) for one hour at different temperatures.

Figure 5 shows the extraction rate. The shape of the curve I is typical for transition curves;[3] the low extraction rate at the beginning is probably due to a lack of monoclinic crystal seeds. It may also partly be caused by the poor heat conductivity of the sulphur.

The points on Fig. 4 and the curve I, Fig. 5 are obtained with sulphur of relatively great particle size, 100 μ, because the effect of the crystallization is then more significant.

The crystallization back to the rhombic form in water and a repeated boiling did not increase the yield to a degree of practical importance. The reasons may be that the phosphorus only diffuses to a certain extent out of the particles, depending on the particle size,[4] and that it occurs in different chemical states with different diffusion rates. For instance, extraction of phosphorus activity from sulphur dissolved in benzene by shaking with water, shows that a few per cent of the activity is not extractable by water and is probably elementary phosphorus.

Changing the pH and the oxidation-reduction potential of the solution showed no marked effect on the extraction yields, indicating that the phosphorus activity, which has been in contact with the water, has been extracted, regardless of its chemical state.

The only factor of importance to change the yields is the particle size of the sulphur. With decreasing particle size and increasing porosity of the particles, the yields are rapidly increased. Common, commercially, sublimated sulphur (pharmaceutical quality) thus gives yields up to 70%, but on using "micronised" sulphur with particles down to 5 μ the yields are raised to above 80%.

The course of extraction by use of the latter types is shown in Fig. 5. The curve II shows a more rapid extraction and the influence of the crystal change is less predominant. Amorphous sulphur (μ-sulphur) obtained as a residue on treating sublimated sulphur with carbon disulphide, thus gave very high yields (86%). Microscopic examination showed the particles to be very porous.

Extraction at elevated temperature and pressure (110°C, 2 atm) gave only a slight increase in the yield.

The high purity sulphur contains, after irradiation, only phosphorus-32, phosphorus-33 and sulphur-35 as activities, stated by following decay-curves for two months. Absorption curves are shown in Fig. 6. The sulphur activity was found to be retained in the sulphur as shown by the absorption curves I and II, taken on the sulphur before and after extraction, respectively. This proves that the sulphur-35 is in its elementary state during the extraction, and a separation of sulphur-35 by the Szilard-Chalmers method is rendered impossible in this case.

2. Apparatus and Procedure

The apparatus used up to the present time consists of a 10 1, wide beaker closed on the top with Pyrex glass plate. Five holes are bored through the top plate. One in the middle is for mechanical stirring, the others are for sucking off the active solution, filling, emptying and refluxing, respectively. The stirrer is made of glass and constructed as a screw to draw the sulphur down in the water by slow rotation. After about 10 minutes the whole sulphur batch is thoroughly wetted by the boiling water which contains only a few drops of the wetting agent.

Most of the experimental runs have been carried out with a wetting agent of the anionic type, consist-

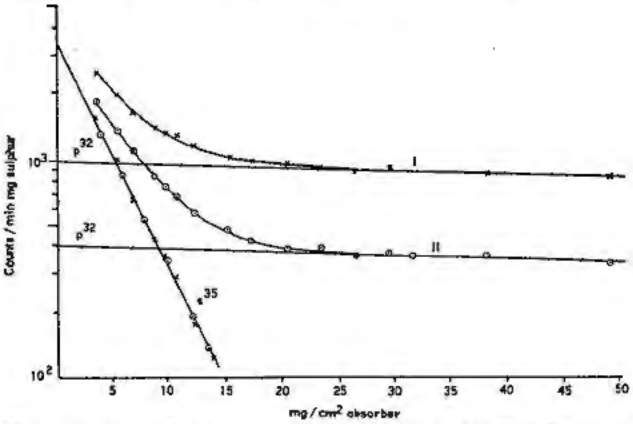

Figure 6. Absorption measurements on sulphur, irradiated for one month, after three months decay

ing of a sodium salt of sulphonated higher alcohols. In this case, however, a quite time-consuming process is needed for removal of the organic matter after the extraction; requiring high temperature and therefore resulting in strong adsorption of the phosphorus activity. By use of a non-ionic wetting agent such as octyl alcohol, this difficulty is overcome, because the wetting agent can then be readily distilled off with water. The solution is therefore sucked off from the extraction vessel through a fine porcelain filter dish into a distillation flask and carefully evaporated down to dryness with nitric acid.

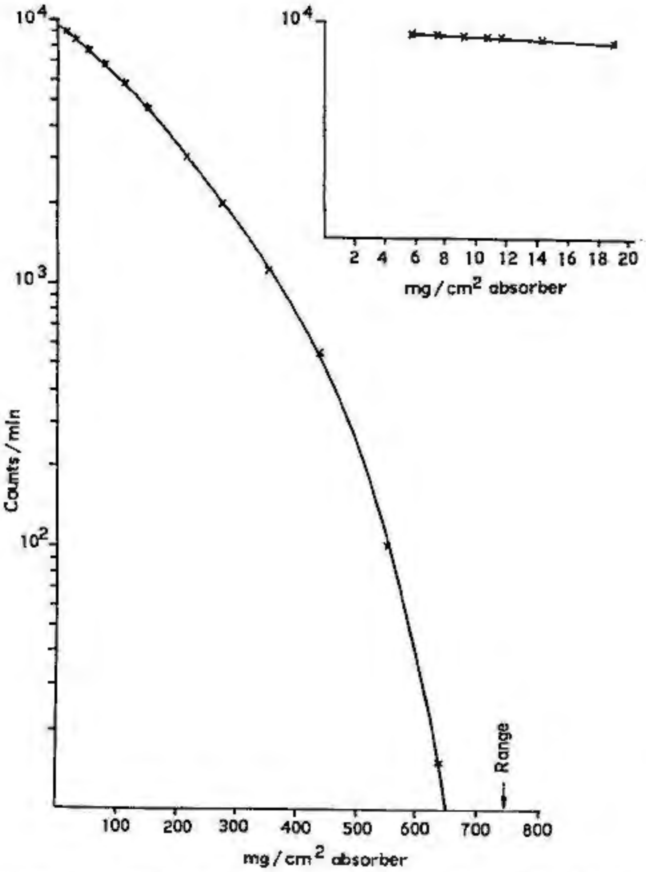

Figure 7. Absorption measurements on final solution obtained from sulphur, irradiated for one month

The phosphorus activity is now present as orthophosphate which is taken up in a suitable volume of water.

The absorption curve, Fig. 7, taken on an aliquot of the final solution after evaporating to dryness, shows a pure phosphorus-32 activity. Determination of the range according to Harley and Hallden[5] was found to be 740 mg/cm^2 in aluminium. No γ-activity could be detected. By comparing a pure phosphorus-32 standard solution with our solution in a scintillation counter, the same counting rate per μc P^{32} was found for the two samples. The chemical purity was controlled by spectrographic analysis, and the products were found to be very pure, impurities not exceeding 1 γ/ml for the elements considered. The aggregate time is about 3 hours. The sulphur, which is sucked out of the apparatus as a slurry in water, may be used repeatedly, but this seems to be of little practical importance.

The specific volume of the irradiation material is about twice that of molten sulphur.

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The Preparation of Kilocurie La^{140} Sources

By R. P. Hammond and J. W. Schulte,* USA

The Los Alamos Scientific Laboratory has for some years utilized for research purposes high intensity point sources of gamma rays. Forty-hour lanthanum-140 was chosen as a convenient emitter for these sources because of its high energy gammas, high specific activity, and because of its conveniently prepared parent, barium-140. This paper will outline the remote control process used to separate essentially carrier-free lanthanum from barium material, and package it in a convenient capsule for use as a point source. As much as 20,000 curies has been processed at one time.

PARENT MATERIAL

The process begins with the arrival at Los Alamos of a shipment of barium-140 material. Barium-140, having a half-life of 12.8 days, is a routine product of the Oak Ridge National Laboratory, where it is produced by dissolution of pile slugs. The product is of high specific activity, but is not carrier-free, because of the stable barium-138 formed in fission. The shipment, weighing only a few grams, is received in a platinum-lined cup of 600 cm^3 capacity, contained in an 8000-pound shield.

As shown in Fig. 1, the barium material, if originally freed from lanthanum, accumulates both radioactive lanthanum and its stable daughter-product cerium-140. The curves marked "Barium," "Lanthanum" and "Cerium" indicate the weight of these substances relative to the original weight of radioactive barium present. Since lanthanum has a specific activity ahout 7.5 times that of barium, the number of curies of lanthanum activity, shown in the fourth curve, soon equals that of the barium. If at any time the accumulated lanthanum and cerium are chemically separated from the barium, the accumulation begins anew as shown, taking the remaining barium as 100 per cent.

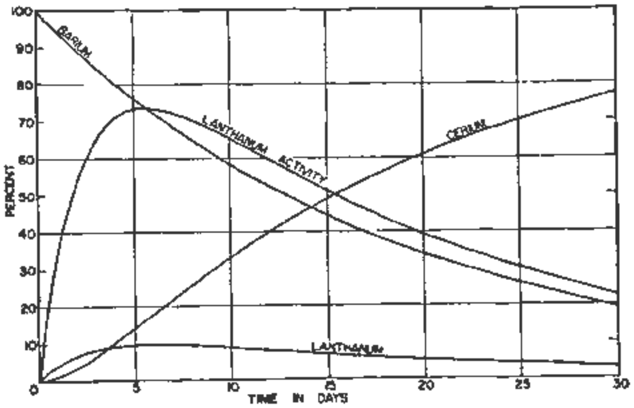

Figure 1. Barium-lanthanum activity chart

Thus, if 10,000 curies of barium-140 is separated after four days growth, the barium has decayed to 80 per cent of the original value or 8000 curies, and 7200 curies of lanthanum-140 are present, or about 13 milligrams. An equal weight of inert cerium-140 has also accumulated from decay of lanthanum. Cerium and lanthanum are chemically similar in this process and consequently both are found in the final source. After another four days about 5750 curies of lanthanum will have grown in, and thus a succession of sources of diminishing size can be prepared from one shipment of parent material.

REMOTE CONTROL FACILITY

The separation of lanthanum and accompanying cerium from parent barium, and the source fabrication and packaging are accomplished in an underground cell 11 ft by 22 ft and 9 ft high, lined with stainless steel for ease of decontamination. On the same level but shielded by 5½ feet of ordinary concrete is the control room from which the cell machinery is operated (see Fig. 2). The principal mechanism of the cell is the transfer crane, which is of the jib type, pivoted on one wall and covering a semicircular area. A carriage moves in and out along the boom of the crane, carrying a quill mechanism with vertical and rotary motions. All four crane motions are driven by external variable speed dc motors and flexihle shafts. Selsyn generators geared to the motor shafts drive repeaters which give at each control station a numerical indication of the position of the crane tool. Although viewing devices give excellent visual control of the crane, the routine transfer operations are carried out by numerical sequence from the position indicators, with the viewing devices as a check.

The other equipment in the cell includes a device for pouring from one cup to another, one for adding any of several reagents from external burets, a stirring motor with platinum coated stirring rod, heaters for evaporating solutions, refrigerators for chilling, and various miscellaneous appliances, such as filter apparatus, cup washing tool, etc. The pelleting machine for fabricating and packaging the finished source material will be described in connection with that operation.

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* Los Alamos Scientific Laboratory.

Figure 2. Remote control cell and control room

The source materials and the chemical separation operations are assayed and controlled by means of neutron counters, shown in Fig. 3. Built into the walls of the cell, these devices are able to measure accurately the radio-lanthanum content of a container even though mixed with radio-barium, and even though other containers of lanthanum are present in the cell. This is accomplished by utilizing the 2.3-Mev gamma-ray of lanthanum striking beryllium to produce photoneutrons which are then moderated and counted. Barium sources and lanthanum sources not in line with the collimator will not be counted, as the lower energy radiation will be below the beryllium threshold.

CHEMICAL SEPARATION

The lanthanum and cerium accumulated after a suitable growth period are separated from the barium by an extremely simple but effective process. The dry mixture is dissolved, then a large excess of chilled fuming nitric acid is added with stirring, which precipitates barium nitrate quantitatively. A filter stick containing a sintered platinum disk about 2 cm in diameter is lowered into the cup, and the solution of lanthanum and cerium drawn by vacuum into another cup. Additional portions of nitric acid complete the transfer.

After rinsing off the filter stick with water, the original cup, now containing only barium, may be stored for accumulation of more lanthanum. The solution in the second cup is evaporated nearly to dryness, a process which is assisted by the self-heating of the radioactive material. Conversion to the fluoride is then made by addition of hydrofluoric acid, and this salt is completely dried and ignited at red heat.

PELLETING AND PACKAGING

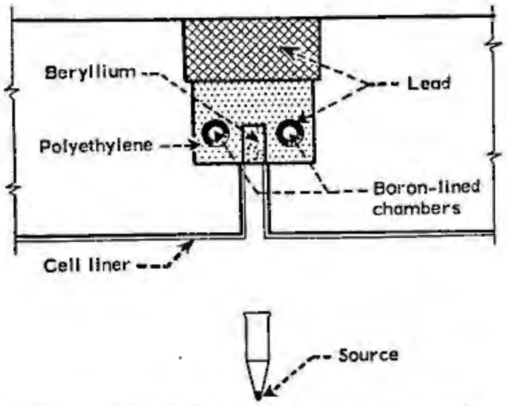

Figure 3. Neutron counter for gamma sources

The result of the foregoing operations is a flaky powder weighing about 50 milligrams, having an activity of several thousand curies perhaps, and consisting of a mixture of lanthanum and cerium fluorides. If there is insufficient radioactive lanthanum and accumulated cerium available to make up 50 milligrams of fluoride, inert lanthanum carrier is added at the start of the process. The next operation is the conversion of the powder into a coherent pellet of high density. This is accomplished by means of an hydraulic plunger and die operated from the control room (see Fig. 4).

This machine has two main stations, the dumping station and the pressing station. Various tools, dies, and fixtures may be loaded by the crane onto two revolving tables, and moved under the appropriate station by rotation of the table.

The cup in which the fluoride salt is dried and ignited is equipped with a platinum-lined removable tip. After the tip is removed in an air powered vise the powder is loaded into the pelleting die by means of a guide funnel and dumping fixture. After compacting the powder by a plunger pressure of 38,000 pounds per square inch, the length of the cylindrical pellet is determined by a remotely-operated micrometer which measures the plunger travel to within 0.0005 inch. Facilities are available for trimming the pellet to the exact size required to fill the final source cavity.

The bottom of the die is now removed, the tip cleaned of loose powder by means of a brush fixture and the final source container revolved into position under the die. The cavity in the container will just receive the cylindrical pellet when ejected from the

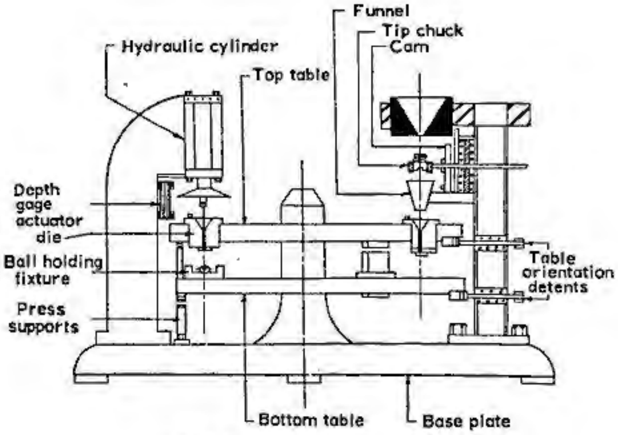

Figure 4. Pelleting machine

die. The container with the pellet is located on the bottom table; it can remain in position while another fixture containing the final plug is superimposed by revolving the upper table. Seating the tapered plug by the hydraulic ram completes the spherical shape of the container and forces the pellet to assume the shape of the central spherical cavity. The aluminum container must be fairly large to provide sufficient surface for the dissipation of the heat energy of the source. The finished source is cleaned, measured and placed in a shield for transport to the user. Approximately ten hours is required for the entire operation.

Preparation of Kilocurie Quantities of Xenon-135

By G. W. Parker, W. J. Martin, G. E. Creek and P. M. Lantz,* USA

In the following discussion, two separate investigations involving entirely different methods are reported on the preparation of Xe^{135} sources for neutron absorption studies. The later work is shown to surpass the former in scale by two orders of magnitude; however, the earlier one was perhaps the more complex and in many ways the more significant. For the reason of its being current the kilocurie-scale work is presented in more detail. In another paper of this Conference, the methods and results of the actual measurement of the xenon cross section are discussed separately.[1]

PREPARATION OF CURIE QUANTITIES OF I^{135} AS PdI_2

At the time it was undertaken (1948), the direct measurement of the Xe^{135} cross section was an experiment of such unusual difficulty that it required the combined efforts of a number of people to solve the problems in both radio chemistry and neutron-physics.

As the result of several considerations, Shapiro and Bernstein[2] devised a system for a neutron beam experiment based on a preparation of only five to ten curies of xenon. A beam of pile neutrons was collimated to pass through the sample in a two inch length of 1.5 mm Pyrex capillary tube (0.03 cm^2 in cross section) in order to observe sufficient neutron attenuation with their curved crystal spectrometer. The Xe^{135} was produced as the daughter activity of 6.7-hr iodine contained in a small amount of PdI_2.

Some introductory work by Borst[3] had indicated feasibility of processing irradiated uranium metal for the 6.7-hr I^{135} and precipitating palladium iodide as a carrier suitable for neutron transmission; however, most of the credit for the chemical methods finally employed is due to the work of Brosi, Freed, Zeldes and others.

PRODUCTION OF PALLADIUM IODIDE, PdI_2 (6.7 hr. I^{135})

The mass-135 chain[4] from which 6.7-hr iodine and 9.2-hr xenon are derived is summarized as follows with a yield of 5.6–5.9 per cent.

$$\begin{tikzcd}
 & & 15.3\text{ min Xe} \arrow[dd] & & \\
<2\text{ min Te} \arrow[r] & 6.68\text{ hr I} \arrow[ur, "30\%"] \arrow[dr, "70\%"'] & & 2.1\times10^{6}\text{ yr Cs} \arrow[r] & \text{stable Ba} \\
 & & 9.2\text{ hr Xe} \arrow[ur] & &
\end{tikzcd}$$

An essentially all glass system was used for the dissolution of uranium metal and the distillation of iodine into a reducing solution of sulfurous acid. In Fig. 1 the schematic arrangement of the cell, slug chute, decoating vessels and dissolvers is shown with the distillation equipment and precipitation vessels.

Uranium, dissolved in hydrochloric acid, to form UCl_4, was oxidized with $\text{Li}_2\text{Cr}_2\text{O}_7$ in the presence of ferrous sulfate as a buffer to prevent over oxidation of iodine. The potentiometric end point for UCl_4 was observed with a platinum-tungsten couple and ferric sulfate was then added as the final oxidant. Iodine was distilled by refluxing vigorously with the addition of small amounts (1–2 milligrams) of iodine carrier (the distillation rate was observed to follow an exponential rate) in order to maintain a 10–20 minute removal half-time.

CRYSTALLINE PdI_2

Of considerable importance to neutron transmission of the sample was the strict freedom from water. After some adverse experience, a method of forming a crystalline PdI_2 precipitate was devised by an adjustment of the HCl and PdCl_2 concentrations and by removing SO_2 slowly by boiling.

CAPILLARY LOADING BY CENTRIFUGATION

The very small mass (10 milligrams) of PdI_2 thus formed was then transferred to a SiO_2 (quartz) coated plastic (fluorothene) funnel, attached to the mounted capillary tube assembly (Fig. 2). A silica coating evaporated onto the plastic funnel as proposed by Freed was found to be strikingly effective in preventing hold-up of PdI_2 on the plastic funnel. When the precipitate was distinctly crystalline, it was easily dried in the capillary by passing anhydrous solvents (ethyl ether, perfluoro triethylamine) through the bed of solids. However, a flocculent precipitate was invariably observed to become mobile (from the radiolytic decomposition of water) and was partly driven back up the capillary tube whenever the centrifuge was stopped.

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* Oak Ridge National Laboratory. Including work by S. Freed, Brookhaven National Laboratory; A. R. Brosi, H. Zeldes, J. R. Sites, G. M. Hebert, G. W. Parker, W. J. Martin, G. E. Creek, and P. M. Lantz, Oak Ridge National Laboratory.

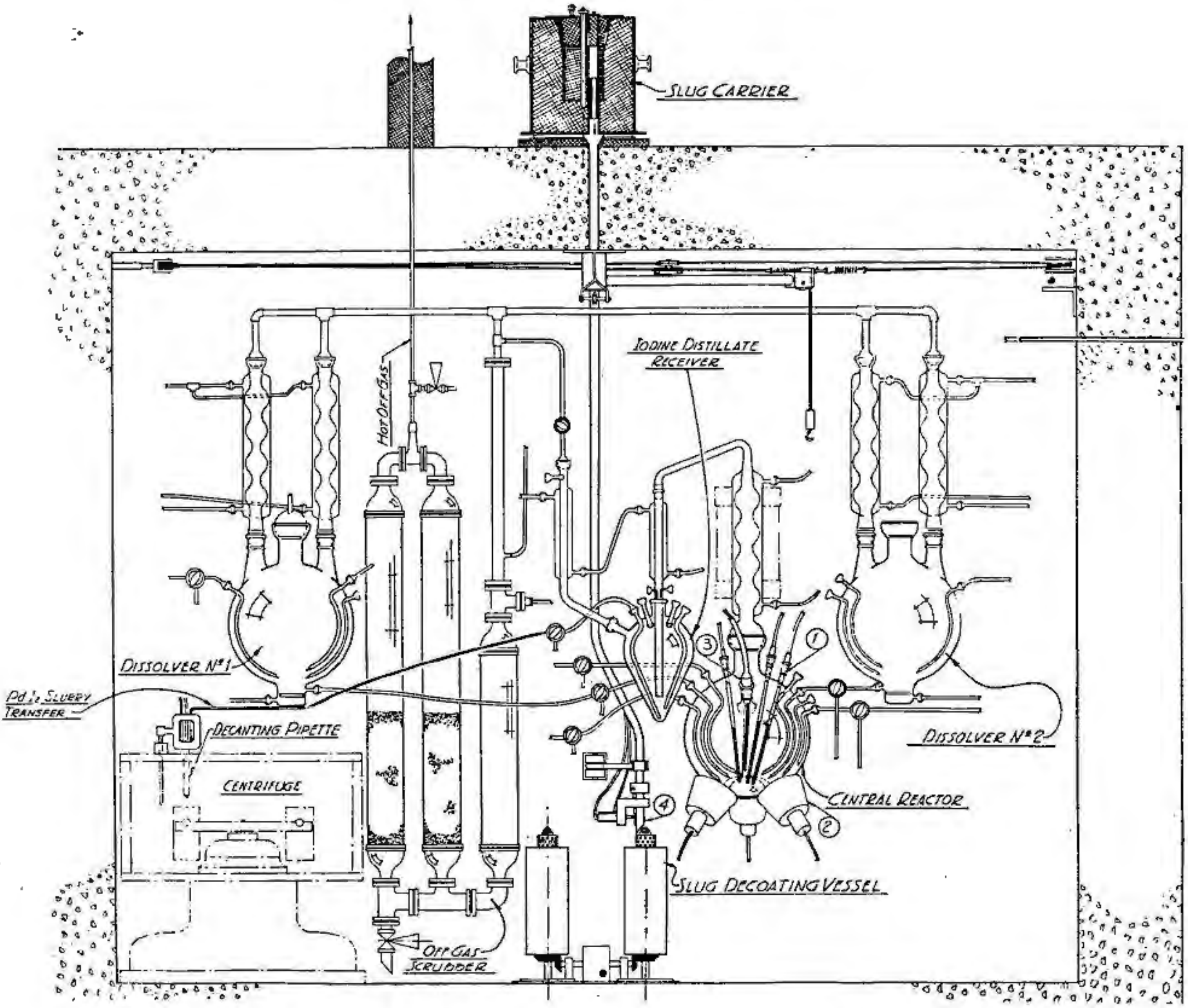

Figure 1. Hot cell equipment for small scale production of 6-hr I^{135}

SAMPLE LOADING AND ASSAY

By means of some appropriate design, and a small electric manipulator, the centrifuge assembly (Fig. 3) was dismantled and placed in clamps for inserting the tapered aluminum plugs. The sample was then leak tested for gaseous xenon and finally withdrawn in a cask and assayed. Brosi and Zeldes were able to calibrate an ion chamber in conjunction with suitable absorber material for assay of the gross sample through the 2 Mev gamma rays of I^{135}.

It is of interest that the processing time for the entire preparation was approximately one half-life, of the 6.7-hr iodine. At the scale undertaken with the handicap of limited space in the hot cell, the time saving was considered significant. Table I summarized the series of preparations with some remarks about each sample.

TABLE I

Run No.	Relative strength in curies		Transmission range (per cent)	Remarks
	By transmission	By ion chamber		
1	—	0.25	—	—
2	1.8	1.8	—	Dry sample
3	4.2	4.5	—	Dry sample
4	Not used	—	—	H_2O content high
5	3.2	—	—	Dry sample
6	8.0	—	16–33	H_2O content
7	8.3	9.7	41–85	Dry sample
8	4.7	—	62–96	Dry sample
9	8.5	9.8	44–85	Dry sample

PREPARATION OF KILOCURIE QUANTITIES OF XENON

The value of the xenon resonance cross section obtained by Bernstein, *et al.*, has been reliable; however, only a very limited range in neutron energy (0.03 to 0.20 ev) was measured. Therefore with the available higher beam intensity from the ORNL Low Intensity Test Reactor and the simultaneous

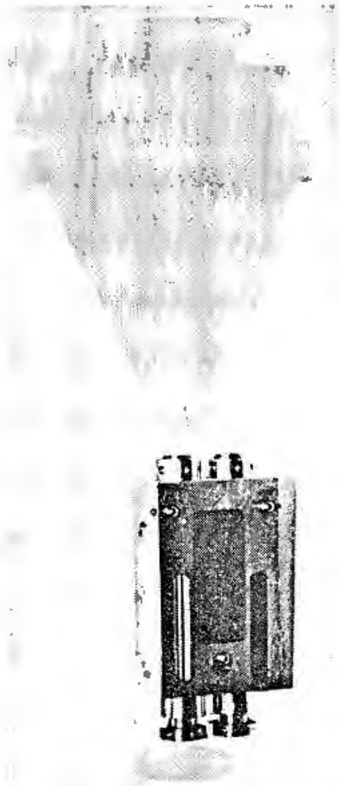

Figure 2. Transmission tube assembly and centrifuge funnel for PdI_2

operation of an 84 channel time-of-flight spectrometer, it became of interest to expand measurements along the energy range, both downward and upward, in a cooperative program between the chemists and the physicists.[5] For this purpose it was fortunate that the Homogeneous Reactor Experiment (HRE) was in operation at the same time since it was capable of producing up to 40,000 curies of 9.2-hr Xe^{135} per day (1000 kw operating power). Other, less attractive, methods of xenon production, including the destructive heating of an MTR type of fuel unit, were investigated as alternate sources.

In its terminal operation, four runs were specially scheduled at the HRE for the collection of kilocurie amounts of the 9.2-hr xenon. The first two runs were required for developing the rapid processing techniques and for providing the usual amount of equipment adjustment. The two latter runs were successfully processed and used for physical measurements and were initially evaluated at 500 to 1000 curies, on the basis of transmission values alone. In order to assign absolute values to the cross section data, considerable attention has been directed toward a precision assay of the intensity of each sample.

THE HRE GAS SYSTEM

The HRE off-gas, as produced, was essentially a low pressure oxygen stream from which large amounts of recombined hydrogen and oxygen had been stripped. The blanket oxygen flowed through a ½ inch pipe into a 600 foot continuous charcoal bed and a vacuum pump; the rate of flow was usually about ½ to 1 liter per minute. In order to divert gas into a trap system, it was necessary to provide only an additional pressure drop over that existing in the off-gas line. Normally, the off-gas was dry but contained in-leakage air as well as the fission products Rb, Cs, and Sr resulting from decay of Xe and Kr; however, it contained no halogens.

GAS COLLECTION AT THE HRE

In Fig. 4 some consideration is taken of the production of relative amounts of the noble-gas fission products. Since some 20 to 30% of the total fission pruducts originate as a gaseous precursor or pass through such a stage, it is immediately evident that in order to collect a predetermined 5 to 10 kilocurie xenon fraction, 10 or 20 times as much krypton and rubidium (the rubidium isotopes are short lived) must be taken (Fig. 5). Disregarding the Xe^{135}, handling of the other activities represented a formidable task.

Since every reasonable precaution had to be observed to prevent out-leakage of even trace amounts of the gas into the working area and very slightly larger amounts into the fan stack, a procedure was devised which permitted collection, over a long period, of a measured flow of gas into a previously evacuated holdup tank (see Fig. 6). In this manner much of the intense radiation was already spent during the collecting time. Then, by outgassing the holding tank with a laminar flow of helium, a further "cooling" period was obtained for the fresher portion of the gas in the tank.

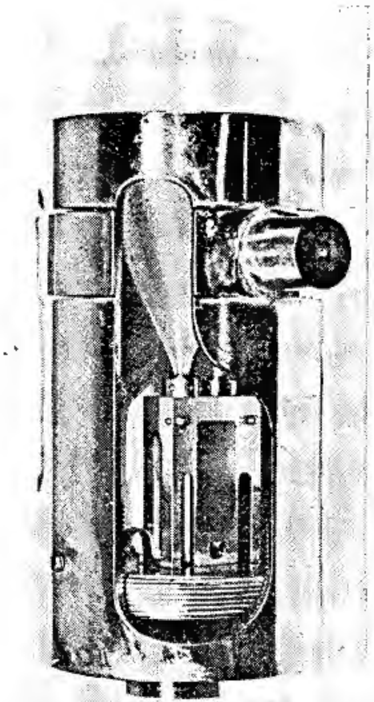

Figure 3. Complete centrifuge assembly for mounting PdI_2

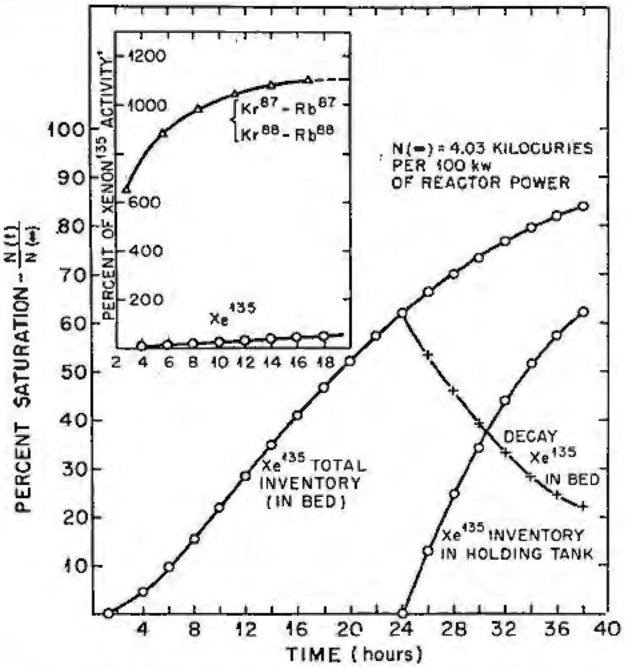

Figure 4. Production of short-lived gaseous fission activities

In making a collection, the off-gas was cooled in a cold trap to liquid oxygen temperature to remove water vapor and then was passed through a flowmeter into the holding tank at such a rate that the tank was filled in 4 to 5 hours. The collected gas was allowed a 2-hr decay time to reduce the intense irradiation due to 78-minute Kr^{87} and 2.7-hr Kr^{88}. After "cooling," the xenon and krypton were displaced with helium and adsorbed on a 2 inch diameter by 36 inch long silica-gel bed (trap no. 1) maintained at liquid oxygen temperature. The gas stream passed through a backup silica gel bed (trap no. 2) and then through a controlled vacuum pump to the HRE stack.

Separation of the krypton fraction was made on trap no. 1 by removing the liquid oxygen coolant from the Dewar and raising the trap temperature to —60°C. The krypton was eluted from the silica gel by means of helium flow at 1000 cm^3 per minute and adsorbed on the backup silica gel bed (trap no. 2). A typical Kr-Xe elution curve is shown in Fig. 7.

After removal of the contaminant krypton, the silica gel bed was heated to 100°C and the xenon was eluted with helium and adsorbed on a movable, shielded adsorber bed which could be used for transporting the xenon to the hot laboratory building (Fig. 8).

XENON PURIFICATION AND MOUNTING

Transfer of the xenon from the movable shield to a 100 milliliter volume liquid oxygen cooled silica gel bed (Fig. 9) was made by heating the adsorbent bed with steam and eluting with a hydrogen flow of 50 cm^3 per minute. After the transfer was completed, helium and hydrogen were removed from the cooled silica gel bed by means of a pump. Pumping was discontinued when the xenon showed an appreciable break-through.

The liquid oxygen coolant was removed and the trap was heated to 100°C, allowing the xenon to expand into an evacuated purification system consisting of a heated barium getter furnace which removed oxygen, nitrogen, water vapors, etc. From the barium furnace the gas stream was passed through a hot palladium valve (Fig. 10), which effectively removed hydrogen. The stream was then circulated through the barium furnace-palladium valve system by means of a Toepler pump until sufficiently purified (as indicated by the residual pressure) and was then expanded into a 1 liter evacuated glass bulb.

The xenon sample bomb, holding a 1 milliliter gas volume (Fig. 11), was fabricated from a copper or steel block having an inside opening measuring 0.900 × 1.125 × 0.060 inches and matching the general beam dimensions. The xenon was pushed into the evacuated holder through 0.023 inch ID stainless steel tubing by means of a mercury leveling bulb and was condensed at liquid nitrogen temperature. Any uncondensed gas was released back into the barium furnace-palladium valve system for recycling.

After loading, the sample holder was raised from the coolant, and the inlet and outlet tubes were crimped, sheared, and soldered by remote manipulation. The sealed sample was allowed to warm up and was checked for leakage by drawing air over the holder and onto a monitored charcoal bed.

ABSOLUTE ASSAY OF Xe^{135}

In order to establish the absolute number of xenon atoms present in the bomb at the time transmission measurements are made, a number of methods of assay were considered. These methods can be divided

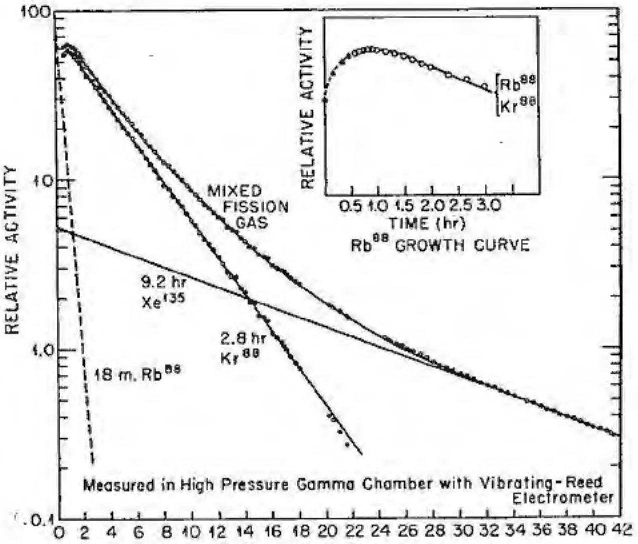

Figure 5. Decay curve of short-lived fission krypton and xenon

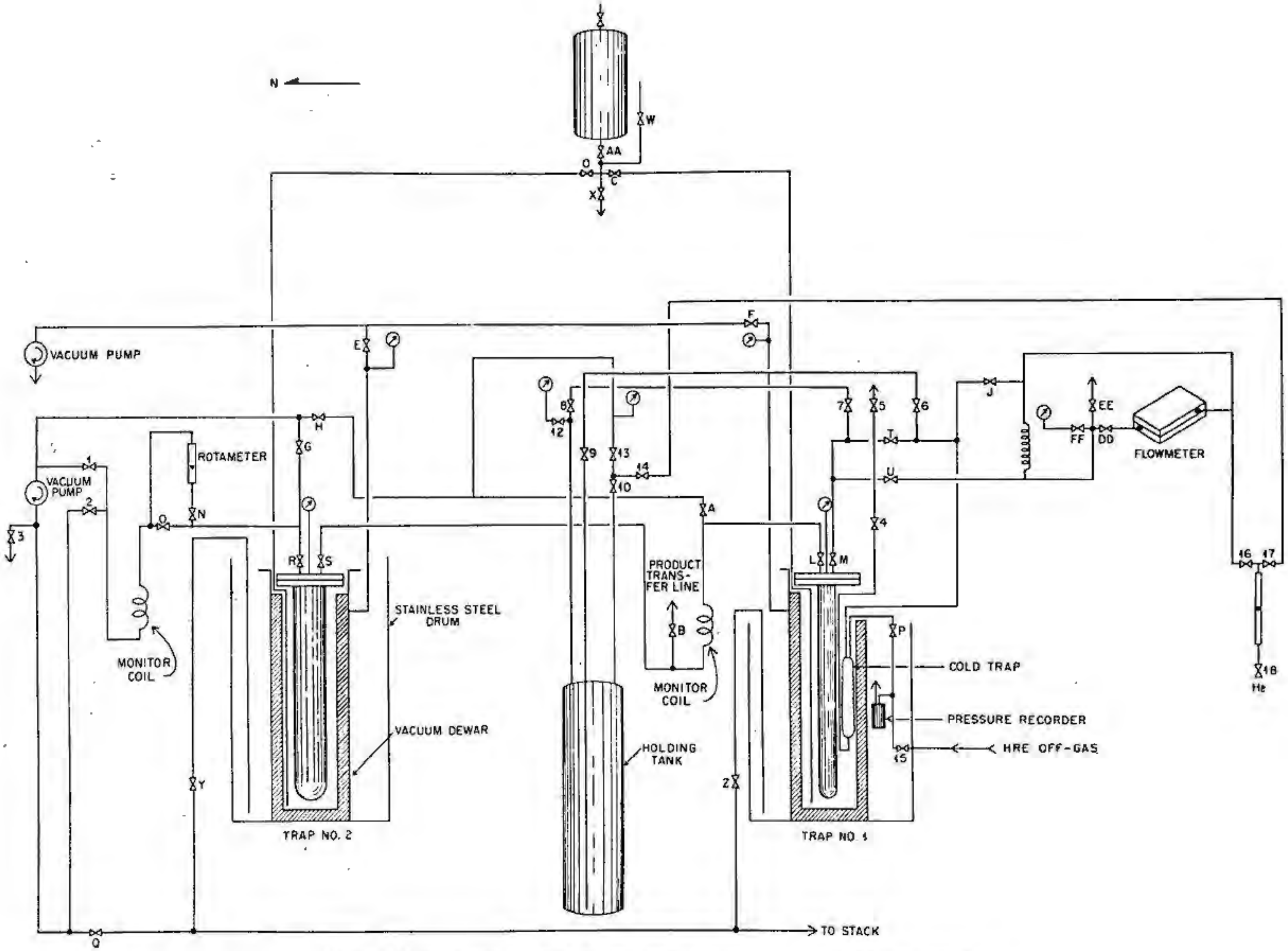

Figure 6. Collecting and fractionating equipment for large-scale production of xenon-135

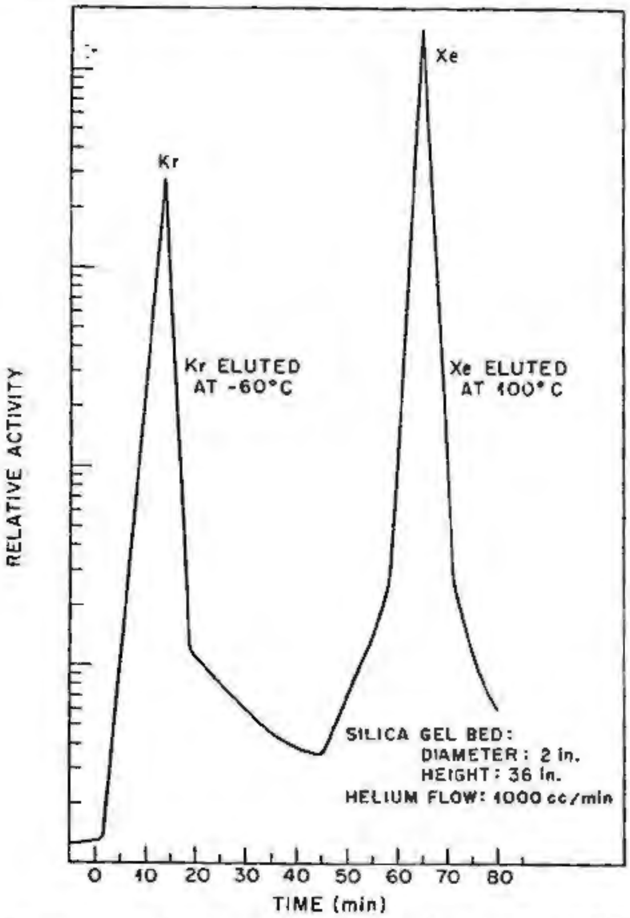

Figure 7. Typical elution curves of large-scale separation of Kr-Xe

into two classes, physical measurements and radiochemical methods, and include absolute gas counting, radiation calorimetry, gamma coincidence counting, xenon and cesium mass spectrometry, and cesium absolute beta counting. For this work, three somewhat independent measurements were selected and have been applied to the 500 curie samples.

ABSOLUTE GAS COUNTNG

It was felt that by no reasonable method could a representative aliquot be made of the gas actually contained in the bomb at the time of closure; therefore an indirect method involving an arbitrary sample taken at the sealing time was used to establish only the ratio between 9.2-hr Xe^{135} and 5.2-day Xe^{133}. For this purpose and for the remainder of the gas counting, a proportional, internal gas counter was used which had a volume of about 300 milliliters, contained argon-methane, and operated at about 2000 volts. The sensitive volume as determined by Macklin and Banta[6] was 85%, and the energy dependence was negligible. The procedure required that the gas contained in the bomb after a reasonable period of decay should be accurately diluted in a calibrated system and that the disintegration rate obtained should then be extrapolated on the 5.2-day decay curve to sealing time.

In practice, some difficulty was experienced with this method because the samples were not available for opening and counting within a reasonable number of half-lives of the 5.2-day activity (more than 100 days elapsed), and this resulted in counting a predominance of 12-day Xe^{131m}, whose X-rays have a high counting efficiency. The gamma spectra in Fig. 12 indicate a more nearly correct proportion of the two activities.

ABSOLUTE BETA COUNTING OF RADIOGENIC Cs^{135}

The second method of assay was based on the pure beta activity of the cesium daughter of the Xe^{135}. A strict requirement with this method is an accurate value for the half-life of the cesium, which unfortunately, was not available. The values reported by Sugarman ($2.1 \pm 0.7 \times 10^6$ years) and by Zeldes *et al.* ($2.95 \pm 0.3 \times 10^6$ years) were obtained under somewhat unfavorable conditions, and because of their uncertainty the assay would have been of little value. However, in this case, by comparing these values with those acquired by other methods of assay, it was possible to obtain an improved half-life value for cesium.

The procedure for quantitatively recovering cesium from the gas bomb was as follows. Before the xenon was released into the aliquoting system, Tygon capillary tubes were sealed over the 0.035 inch stainless steel tubes, a glass wool plug was inserted, and, with the gas container cooled to liquid nitrogen temperature, the outlet tube was flexed until it was work-hardened and broken. The bomb was then warmed to room temperature, and the inlet tube was also broken so that counter gas could be passed through the bomb to sweep out all the xenon. The bomb was then detached along with the glass filter and both were washed with water until the rate of the beta activity being removed had decreased to zero. Finally, a dilute acid wash was used to make sure that no trace of cesium had been left. This material was all made up to exactly 10 milliliters, from which many aliquots were counted. The mass

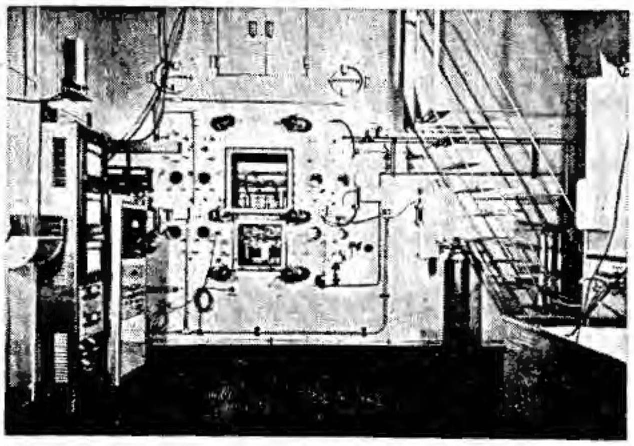

Figure 8. Hot cell operating face, high-level radiochemistry building

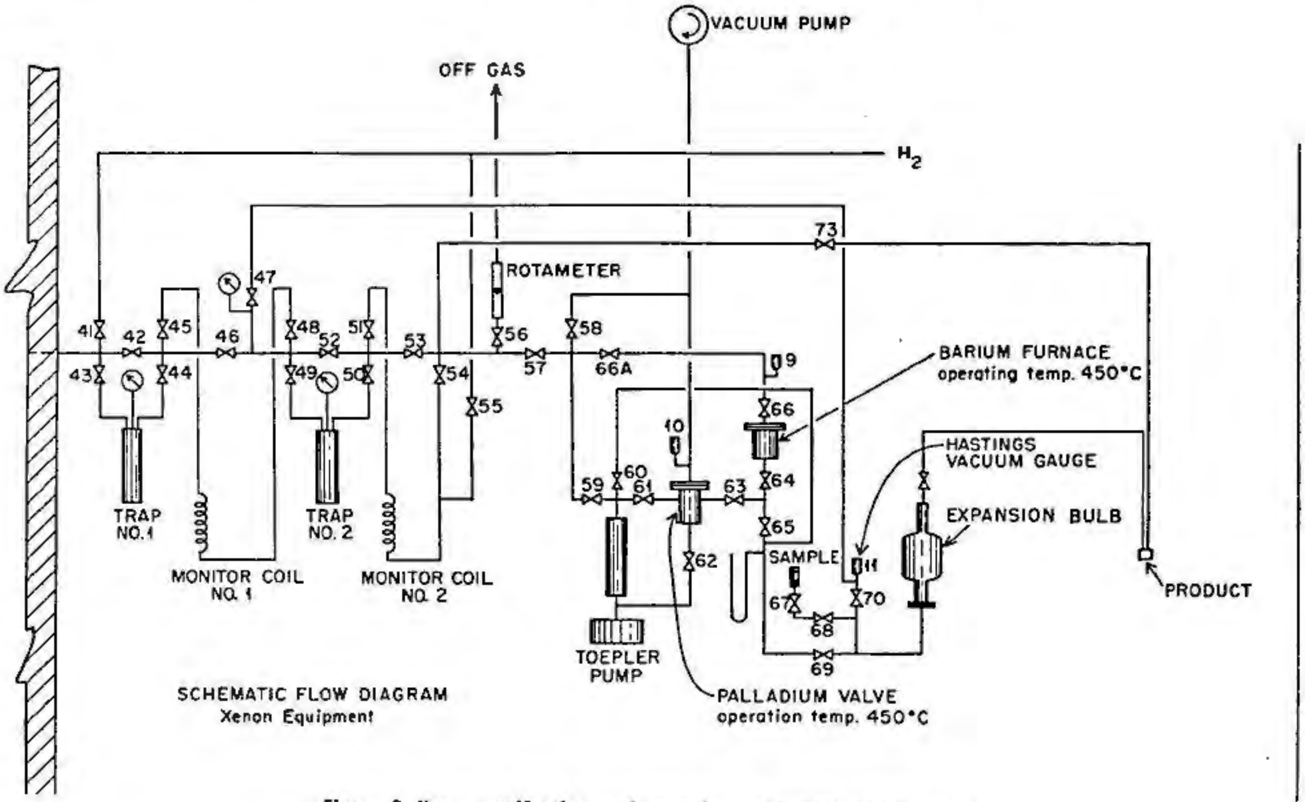

Figure 9. Xenon purification and mounting equipment arrangement

of the mounted samples was determined by direct weighing, and correction was applied for self-absorption. For 100 microliters (1 per cent of the total sample) the observed average counting rate was 2650 counts per minute. By correcting for the mass of the sample (60 micrograms on an area of 0.3 cm^2) by a plot (Fig. 13) of successively decreasing sample sizes from (400 to 20 micrograms) and normalizing each to the equivalent of the 100 microliter count, an extrapolation to zero sample thickness gave a corrected count of 3100 counts per minute. Samples were mounted on 75 micrograms per cm^2 Zapon film and counted in a flow type proportional counter at 50% geometry. The slide chamber was provided with a 50 cm^3 gas pocket below the sample to reduce back-scatter. Based on the use of the number 279,000 (310,000 minus 10 per cent for back-scattering) for the total disintegration rate at 50 per cent geometry and the value 185 micrograms for the weight of the cesium (given in the paragraph below), a value of 2.0 × 10^6 years ± 10 per cent is proposed for the half-life of Cs^{135}.

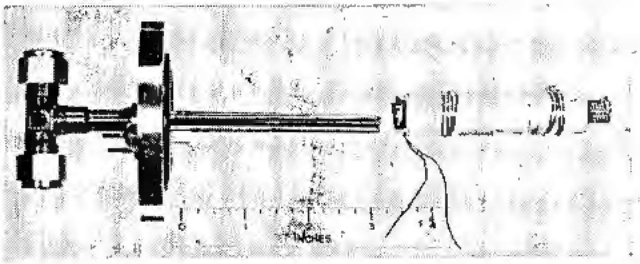

Figure 10. Palladium valve and heater

CESIUM ASSAY BY ISOTOPIC DILUTION

One of the unique properties of cesium is its high thermal ionization efficiency in a mass spectrometer. As a means of assay the isotopic dilution analysis of the extracted cesium was obtained through the assistance of R. Baldock and J. R. Sites of the Mass Spectrometry Group.

In order to obtain the concentration of the dissolved cesium-135 in a carefully aliquoted amount of solution, a standard reference solution, or spike, was prepared with an appropriate concentration of pure natural Cs^{133}; and, in definite ratios, for example 1:1 or 2:1 the two solutions were mixed and the resulting new ratios of Cs^{133} to Cs^{135} determined in

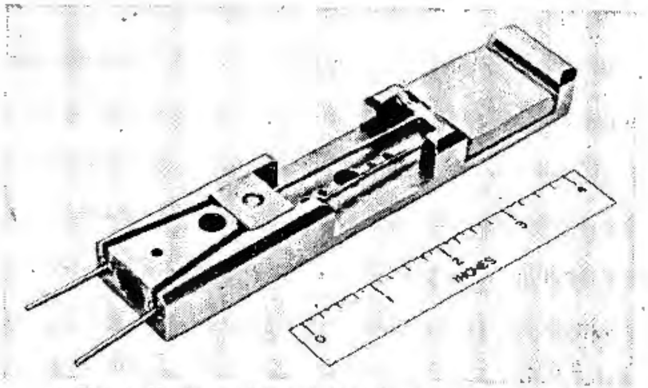

Figure 11. Xenon transmission changer and holder

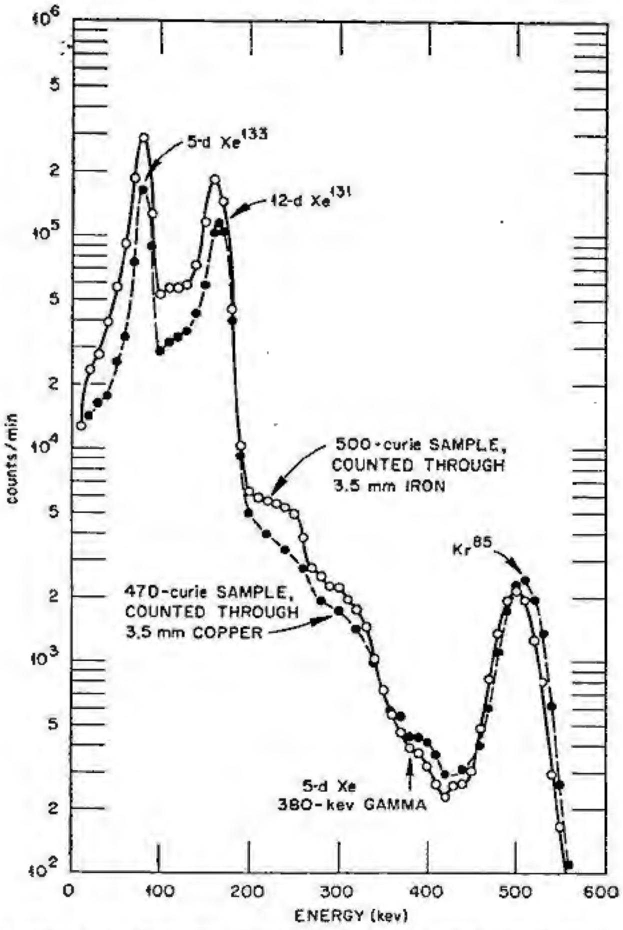

Figure 12. Gamma ray spectrograms of multi-curie xenon samples after cooling

the mass spectrometer. The method of calculation was then set up which depends only upon the measured intensities of the ion peaks. This value is used in the following expression:

$$W_x = W_s \frac{\frac{A_a{}^s}{M_s} - \frac{A_b{}^s}{M_s}\cdot R}{\frac{A_b{}^x}{M_x}\cdot R - \frac{A_a{}^x}{M_x}}$$

where: x = unknown (Cs^{135}); s = spike (given, equal to 66.5 μg/ml); w = weight (or concentration); A = relative abundance; M = atomic weight; and R = cesium peak height ratio.

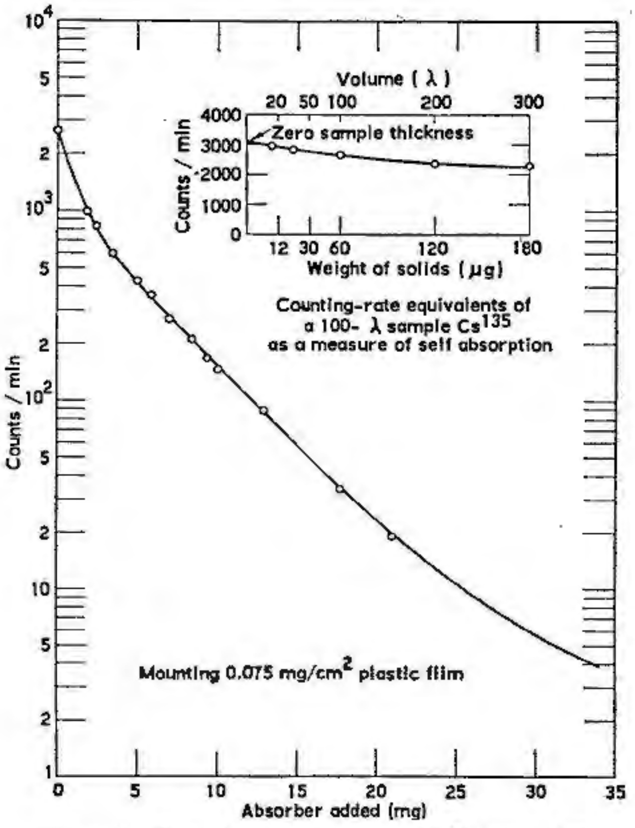

Figure 13. Beta absorption curve of Cs^{135} from Xe^{135}

For the final values used in the absolute cross-section calculations, a total of six determinations on each sample were evaluated by the method of least squares. For the purpose of illustration a mean determination is given in Table II showing how a value in terms of curies of xenon was obtained.

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TABLE II. Isotopic Dilution Analysis of Extracted Cesium

Material	*Mixture**	*Relative abundance found*		*Radio: R*	*Concentration*	
		Cs^{133}	*Cs^{135}*		*Cs^{135} micrograms/ml*	*$Xenon^{135}$ (curies)*
Spike	natural	100	0			
Sample # 1 (hot)		84.12	15.88			
Sample # 1 spiked	2s + 1x	92.58	7.42	12.48	18.5	467
Sample #2 (hot)		78.22	21.78			
Sample # 2 spiked	1s + 2x	70.30	9.70	10.31	19.75	500

* s = spike (normal cesium, 665 μg/ml) aliquoted in 100 micro-liter portions. x = unknown (aliquoted in 100 micro-liter portions) from 10 milliliter total sample.

Design and Operation of a Pilot Plant to Produce Kilocurie Fission Product Sources

By R. H. Simon and J. A. Consiglio,* USA

The industrial utilization of the gamma radiation from fission products requires the use of compact sources of high specific activity. The starting point for the preparation of such sources will be the process waste streams from fuel reprocessing plants. These streams will usually be solutions of gross fission products mixed with inactive chemicals. The problem of preparing a source of a specific fission product from such solutions will involve the development of chemical separation processes and equipment to handle the material safely. Although the chemical process will vary depending on which fission product is to be concentrated, many of the design and equipment problems will be common to all processes. This paper presents the design concepts, the layout, and a description of the major components of a pilot plant for the recovery of a pure fission product from process waste streams. The pilot plant produced kilocurie sources of solid carrier-free material.

THE PROCESS

The process which was developed for the separation of the specific fission product was divided into two phases:

1. The isolation of the desired fission product from other fission products and from nonradioactive chemicals.
2. The concentration and conversion of the isolated fission product to a solid.

The isolation was accomplished by adsorption from a prepared feed in a packed column. The isolated fission product was eluted from the column and concentrated by precipitation in the presence of a cellulose filter-aid. The precipitate was then filtered, washed, and dried. The filter-aid was removed by combustion, after which the temperature was raised to about 650°C to calcine the product.

DESIGN CONCEPTS

Five concepts guided the design:

1. Compartmentalization. Equipment which might require maintenance or which would be difficult to decontaminate, such as valves and pumps, was isolated.
2. Gravity flow. To minimize the possibility of backup of radioactive solutions into operating areas, all radioactive equipment was located in a lower cell called the process cell, and all operations were performed from above. To avoid holdup of solution, there were no loops in lines within the process cell.
3. Clean layout. To simplify maintenance, equipment was laid out in rows and the piping was installed parallel to the cell walls.
4. Contamination confinement. To minimize the spread of contamination in the event of leakage of radioactive solution, major components were confined in sheet metal boxes.
5. Minimum movement of solid. To minimize the material handling problem, several of the steps of the concentration process were carried out within a product vessel contained in a shielded cask.

LAYOUT

The pilot plant was located in an existing two-floor cell containing obsolete but uncontaminated equipment. As shown in Fig. 1, this cell was divided into three areas: an operating room, a sample room, and a process cell.

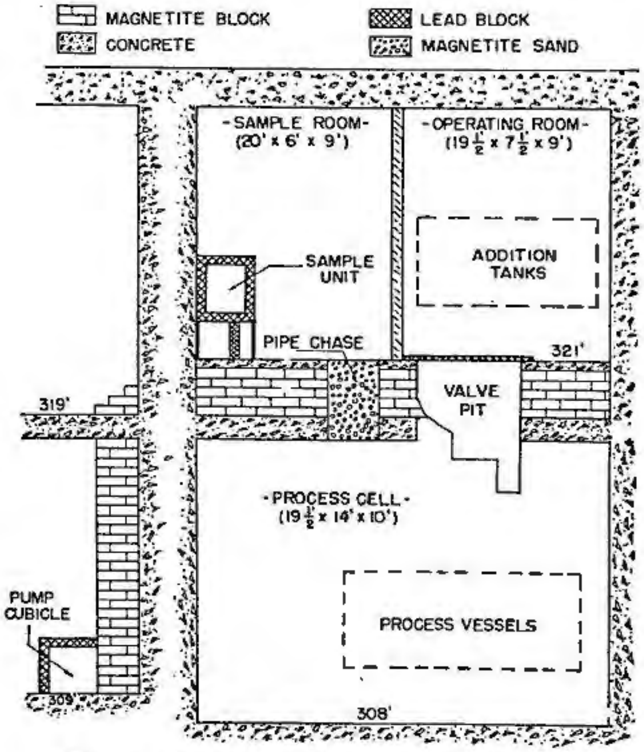

Figure 1. Cross section of fission product pilot plant

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* Knolls Atomic Power Laboratory, Schenectady, New York.

The operating and sample rooms were separated by a control panel which also acted as a contamination barrier. Instruments which might become contaminated due to a backup of radioactive solution were located on the sample room side of the panel board, and were viewed from the operating room through sliding plate glass windows. The floor of the sample room was lower than that of the operating room and drained to a sump in the lower cell. The floor was sheathed with stainless steel sheet for ease of decontamination.

The process cell contains all the process vessels. These were located in an open-top stainless steel box to prevent the spread of contamination in the event of leakage; sprays and steam were provided for decontamination. Also located within the process cell was the product handling equipment. This included a narrow gauge track on which an electrically driven transfer cart could be moved into and out of the cell.

Magnetite concrete blocks were added to the original concrete walls to provide sufficient shielding. Since the upper level was to be used for operations, it was necessary to shield the floor of this area from the lower level. Magnetite blocks were laid on the existing floor to a depth of twenty inches; a concrete cap was added providing a total floor thickness of thirty-five inches.

EQUIPMENT

Chemical process pilot plants, by their nature, require valves, pumps, and sampling equipment. Where the final product is a solid, material handling equipment must be included. The high levels of radioactivity associated with fission products is the major consideration in designing these components. Equipment must be as nearly maintenance-free as possible, and where this cannot be guaranteed, the particular component must be made reasonably accessible. In order to achieve this accessibility, the various components were compartmentalized as described below.

Valve Pit

Valves usually require maintenance. If possible they should be accessible and easily replaced. By the use of lead shielding it was possible to create what was essentially a cell within the floor of the operating room. In this cell called the valve pit, all valves were located through which passed radioactive fluids and all valves carrying nonradioactive solutions to the process vessels. To prevent the spread of contamination in case of leakage, the valve pit container, as shown in Fig. 2, was an open-top water-tight vessel.

There were three levels of valves in the valve pit. The valves in the upper level were used to feed nonradioactive solutions from the operating room into tanks in the process cell, and the flow through them was downward. These valves were installed with regular tubing and fittings. Two inches of lead shielding in the form of lead blocks, which covered the top of the valve pit, protected the operators against the slight possibility of accidental backup of radioactive solutions through the lines to these valves. In the middle level were located valves and lines through which ran process solutions, or through which the process tanks were connected to the pilot plant vent system. Flow through these valves was from below into the valves and down into the process cell. These valves were shielded by an additional two inches of lead in the form of lead blocks. The process valves which required gravity flow from one piece of process equipment to another were located in the lower level. As seen in Fig. 2 the lower level valves were located below the ceiling of the bottom cell. These valves were to carry the most highly radioactive solutions and were shielded by an additional four-inch thickness of individually removable lead blocks.

The valves in the middle and lower levels were stainless steel gate valves of slightly modified standard design. For quick removal each valve of the lower two levels was installed between two smooth-faced stainless steel blocks, one fixed and one movable, as shown in Fig. 3. Teflon gaskets were inserted into recesses in the end faces of these valves. The valve seal was made by forcing the movable block closer to the fixed block, thus compressing the gaskets. This was accomplished by means of a single large diameter screw.

The use of comparatively small pieces of lead shielding eliminated the necessity of decontaminating the entire valve system. To replace a valve it was necessary to decontaminate only one row of valves and remove the lead shielding blocks over the row of valves and the row of jack screws. A faulty valve could be released by backing off the jack screw by means of a long-handled ratchet wrench; the valve was then removed with a hook. A new valve could be installed by reversing the above procedure. To avoid radiation from the tanks in the lower cell when part of the lead was removed from the valve pit, a two-to-four-inch-thick shadow shield of lead was located on a platform in the lower cell directly under the valve pit.

All the valves were operated by means of extension handles protruding through the shielding to the operating floor. Universal joints were used in these extension handles to take care of any misalignment of holes in the lead shielding. No radiation leakage through these holes was detected.

Process Pump and Pump Cubicle

A diaphragm pump was used to feed the adsorption column at controlled flow rates. This pump had a hydraulically driven diaphragm which in turn was coupled hydraulically to two remote diaphragm heads, either one of which could be operated independently. The heads and the process lines leading to and from them were contained in a lead-shielded stainless steel box (the pump cubicle) located outside the wall of the process cell. To avoid holdup of liquid, care was taken in designing the pump cubicle piping in order that the flow from the feed makeup tank was down-

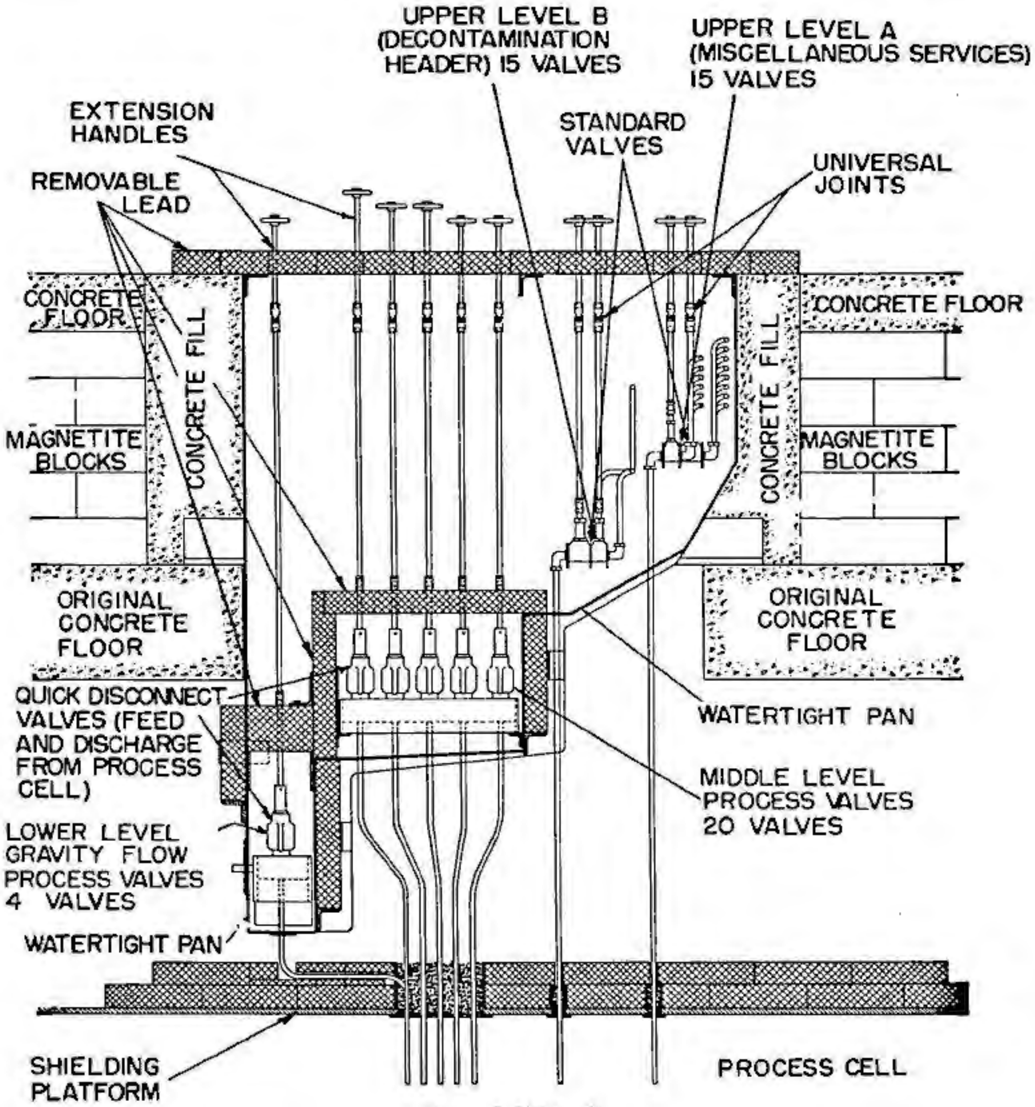

Figure 2. Valve pit

ward to the bottom of the pump and upward from there to the column. The valves for isolating the remote heads from each other were of the same type as those used in the valve pit and were installed in a similar way.

Concentration Equipment

For mechanical simplicity and to minimize the movement of solids, the concentration operations were carried out in a product vessel which was located within the product cask during the entire sequence of operations. These operations included filtration, drying, washing, combustion, and calcination. As shown in Fig. 4, the product cask consisted of two basic parts, a body and a cover. The cover was constructed so that a large hole was left in the body when the cover was removed. The product vessel was keyed to a pedestal welded to the cask at the bottom of the hole. The cask body was positioned by dowel pins on the transfer cart on which it was moved into the process cell.

A cover removal station and a processing station were located within the process cell as shown in Fig. 5. At each of these stations there was a hydraulic jack which raised the cask from the cart. After the cask cover was removed at the first station, the cart was moved to the processing station where the hydraulic jack raised the cask and sealed the product vessel against a fixed flange. As shown in Fig. 6, the product vessel was a double-walled vessel, with the inner wall made of a sintered stainless steel filter. There were two openings in the top of the product vessel: (*a*) through one, the slurry of precipitate and filter-aid entered the filter element, and (*b*) through the other, the filtrate was discharged by way of a tube connected to the annulus between the filter element and the outside wall of the vessel. A spiral-wound asbestos and stainless steel gasket selected for its ability to withstand high temperatures was centered over each hole and tack-welded to the top of the product vessel. These gaskets were used to seal the product vessel against the fixed flange and also to seal the product vessel cover to the body of the product vessel after operations were completed.

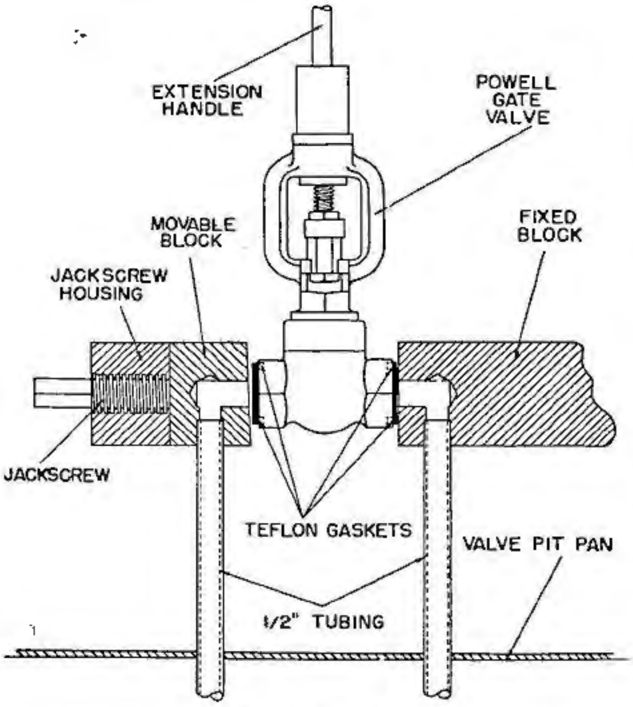

Figure 3. Removable valve

Suspended from the fixed flange was a cylindrical electric resistance open-end furnace. This furnace was used to dry the filter cake, to burn the cellulose filter-aid, and to calcine the product. Surrounding the furnace was a stainless steel can, containing thermal insulation, which fitted inside the hole of the cask body.

After the operations were completed, the cask containing the product vessel was lowered from the fixed flange and returned to the cover station where the cover was replaced on the cask. The product vessel cover was locked and sealed in position after the cask was brought outside the process cell. The locking was accomplished by means of a key which extended through the cask cover.

Process Vent System

The off-gases from the combustion operation passed through the filter element into a filtrate receiver, and from there into the process vent system. The purposes of this vent system were (*a*) to prevent the spread of contaminatinn; (*b*) to remove particulate matter which might be present in the off-gases not only from the filtrate vessel but also from all the other process vessels; and (*c*) to maintain a slight vacuum in the process vessels. The blower for the vent system discharged into the building ventilation exhaust system, providing a large dilution before the exhaust gases passed to the building stack. An asbestos-paper filter was located in the process vent line and was installed between two spring-loaded flanges which could be sprung apart to permit quick replacement.

Sampling

It was desired to make sampling simple, safe, and reproducible. All radioactive solutions associated with the sampling operations were contained within a shielded stainless steel box called the sample unit. This box was divided into two cells, a sampling cell in which samples were drawn from the process tanks and a sample handling cell in which the samples were prepared for transfer to the analytical laboratory.

Both recirculating and noncirculating types of samplers were considered. The recirculating type gives good samples but results in the atomization and subsequent entrainment of appreciable quantities of radioactive solution. The problem with the noncirculating type is to obtain reproducible samples. It was decided that for the small volume of highly radioactive solution in the pilot plant, the entrainment would be the more serious problem. As a result, the noncirculating type was selected.

The samples were drawn by means of a hand-operated piston. As shown in Fig. 7, the vacuum created by pulling out the piston caused the liquid to flow from the tank below up into the sample unit. A surge pot was inserted in the line to prevent drawing of the sample solution into the unshielded piston chamber. Eight tanks could be sampled, each having its own piston and surge pot. The components of the sampling system and tanks were elevated with respect to each other in such a way that there was no holdup of solution in lines or equipment. This was a major factor in obtaining reproducible samples. Pro-

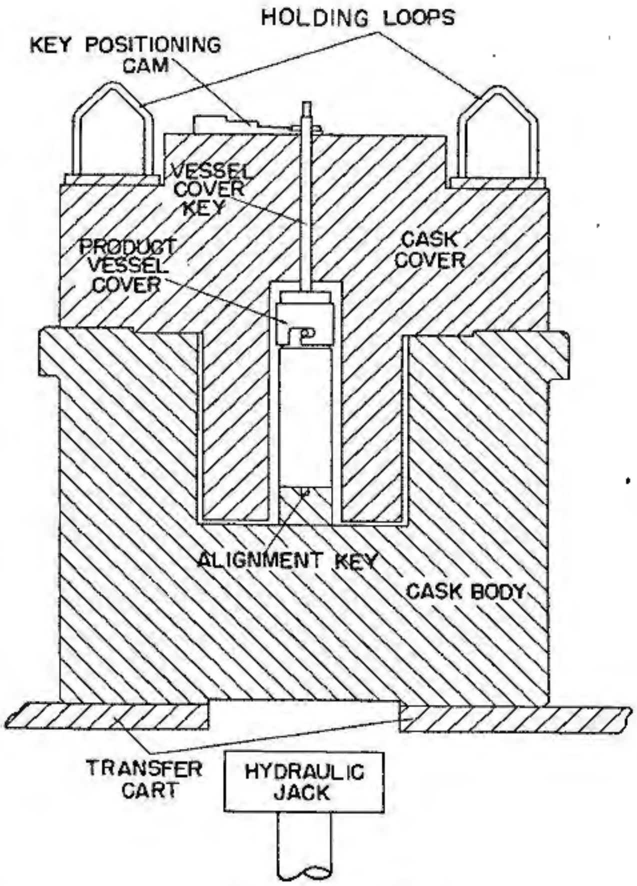

Figure 4. Product cask

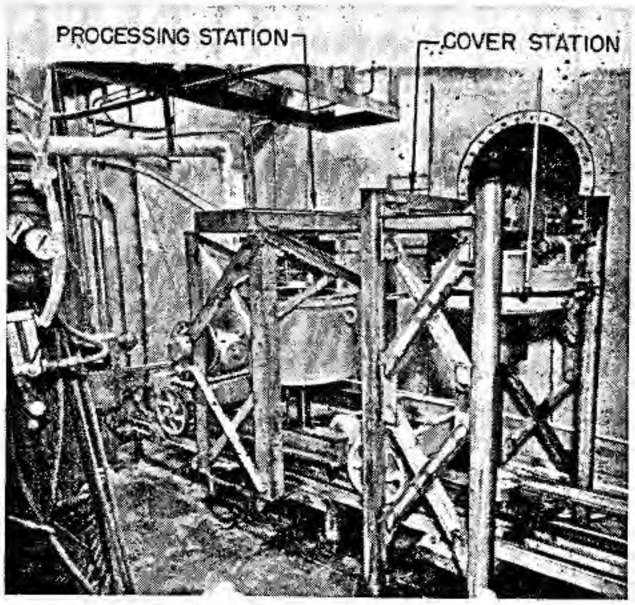

Figure 5. Concentration equipment

vision for flushing out the surge pots and sample lines was by funnels entering the surge pots.

With the sample line full of solution up to and partly into the surge pot, a sample was removed by means of a two-way valve located in the sample line between the tank and the surge pot. This valve was a stainless steel plug valve with a Teflon liner. The body of the valve had four openings, and the plug had a hole with a volume of ½ milliliter. After the sample was drawn, the plug was rotated 90 degrees and the sample was dropped into a small glass cone. The cone was held in a slide assembly on a monorail and was moved into the handling cell.

In the handling cell primary dilution for the analytical laboratory was made, if necessary. In addition, unused portions of samples were removed through a suction line connected by way of a trap and vacuum pump to a waste tank in the cell below. Decontamination sprays were provided in each of the two cells of the sample unit. The interior of the sample unit was observed through lead glass windows.

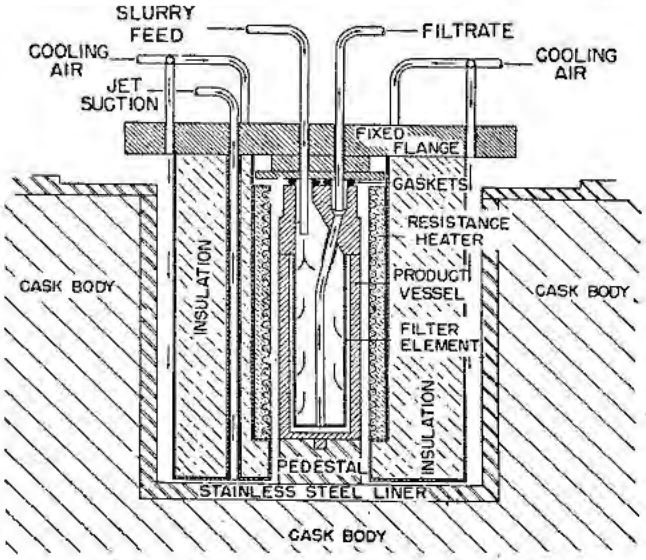

Figure 6. Product vessel at processing station

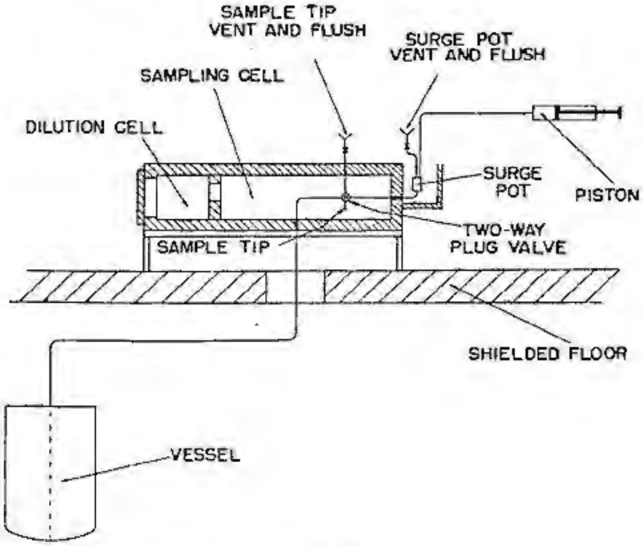

Figure 7. Sampling system

SUMMARY

A review of the actual design shows how the design concepts were followed.

1. Compartmentalization. The chief example of this was the division of the pilot plant into three areas for processing, sampling, and operating. In addition, the major components, the process valves, the process pump, and the sample unit were further compartmentalized.

2. Gravity flow. The location of the operating and sampling rooms above the process cell greatly reduced the possibility of radioactive solutions accidentally appearing in operating areas. Installation of piping without loops simplified decontamination and aided in obtaining reproducible samples.

3. Clean layout. The installation of process cell equipment and piping in an accessible manner provided for ease of maintenance.

4. Contamination confinement. The valve pit, pump cubicle, process vessel box, and the separation of the sample room from the operating room, are all examples of the application of the principle of confining contamination.

5. Minimum movement of solids. The use of the product vessel for several processing steps while it was positioned within the product cask was an application of this concept.

In practice, the design concepts proved sound. The pilot plant was operated for a period of six months during which a number of successful kilocurie runs were made. Because of the short period of operation, some features designed into the pilot plant to aid in decontamination and maintenance were either not tested or tested only to a limited extent.

There was no apparent leakage through valve gaskets or packing or across valve seats. During subsequent modifications of the pilot plant for a different process, some of the valves in the valve pit were removed and replaced without difficulty.

The feed pump operated satisfactorily except during the last run when the two remote heads failed and a temporary feed system was used to complete the run. Because of the imminent termination of the project, the pump was not dismantled to determine the exact cause of the failure.

The concentration equipment gave no difficulty. The design of the product vessel proved adequate for the multiple processing steps and in the prevention of carry-over of activity to the process vent filter during combustion and calcination. The process vent filter was replaced once per run when the dose rate reached 100 milliroentgens per hour.

The sample unit provided a safe means for obtaining reproducible samples. The safety of this operation and the operation of the pilot plant as a whole is indicated by the fact that radiation exposures to personnel did not exceed the maximum allowable daily radiation dose of 50 milliroentgens per day, at any time.

It is believed that the concepts followed in the design of this pilot plant are applicable to most high-level radiochemical pilot plants.

ACKNOWLEDGEMENTS

The authors wish to thank W. E. Unger of Oak Ridge National Laboratory for his valuable suggestions during the preliminary design stages. The authors wish to acknowledge the work done by S. Bigelow, S. C. Furman, D. Mohler, B. F. Rider, and F. N. Schell.

Current Techniques in the Handling and Distribution of Cobalt-60 Radiation Sources

By A. B. Lillie,[*] Canada

One of the most important early uses of gamma-ray sources was in therapy treatment with radium. With the advent of nuclear reactors and pile-produced isotopes it was quickly apparent that a major step in radiation therapy could be made by replacing the radium sources with much more intense sources artificially produced. And it is not surprising that some general techniques in producing and handling large gamma-ray sources should develop around the special requirements of the first of these high-intensity multicurie sources—the therapy sources.

The characteristics of a source suitable for radiation therapy are high intensity, small size, reasonably long half-life, and sufficiently high energy gamma-ray emission.[1] Of the various gamma-ray emitters which can be produced in a reactor, cobalt-60 with gamma-ray energies of 1.17 and 1.33 Mev has appeared best suited and has been used the most.

The high intensity required for a therapy source together with the need of a small geometrical size infer the use of high specific activity material. This high intensity allows therapy treatment from a distance large enough for good depth dose characteristics and the small geometrical size causes a desirably small penumbra. Specific activity is proportional to the neutron activation cross-section and to the neutron flux, and for cobalt this cross-section is indeed relatively large—34 barns. But the maximum specific activity is still limited by the neutron flux available, and not until high-flux reactors such as Canada's heavy-water reactor, NRX, began operating was it possible to produce the high specific activity and thus the high intensity sources suitable for radiotherapy. In a standard production position in NRX, for example, it is possible to produce specific activities of 35 curies per gram in less than 18 months. Using this specific activity, a source containing approximately 2000 curies of cobalt-60 can be made which will deliver a dose rate of 50 roentgens per minute in air at a treatment distance of 80 cm.

The absorption of neutrons in the target material—here cobalt metal—causes other effects which must be taken into account. In a reactor a mass of neutron-absorbing material will lower the neutron flux in the vicinity of the mass and, in addition, the outer layers of the mass will shield the inner layers from the neutrons. Each of these two phenomena, "flux depression" and "self-shielding," will lower the effective neutron flux and thus the specific activity that will be produced.[1] When producing cobalt-60 in large quantities in NRX, it is estimated that the specific activity is lowered by 50 per cent, due primarily to flux depression.

To reduce the self-shielding, the Canadian approach has been to use the cobalt in the form of small cylindrical pellets 1 mm in diameter by 1 mm long. These pellets can be held, during neutron irradiation, in thin (approximately 3 mm) cylindrical shells, and then after removal from the reactor be transferred into a source container suitable for use in therapy machines. In this latter container, the pellets form a right circular cylinder of maximum diameter 3 cm and maximum length 3 cm. This versatility of form can take advantage of the best geometry for pile irradiation as well as the best geometry for therapy use. One disadvantage of using a pellet source is that the bulk density of the cobalt pellets (4.8 gm/cm^3) is approximately one-half that of solid cobalt (8.9 gm/cm^3) and as a result fewer curies activity can be contained within a given volume than with the use of a solid cobalt source.

Since cobalt-60 has a half-life of only 5.3 years, corrections for its decay must be made periodically during its use. Eventually—probably within one half-life—the intensity of the source will have decreased to the point that the source must be replaced. Even the decayed source does represent, however, a considerable neutron irradiation time and cannot be lightly discarded. One possibility is for the source to be used where a lower intensity source is adequate. Another possibility—and the one which has been adopted by Atomic Energy of Canada, Limited, the Canadian supplier of radioactive isotopes—is for this active material to be returned to the supplier for reactivation. This choice is based on estimates of the economic factors involved rather than on the technical aspects of the operations.

PROCEDURE AND EQUIPMENT

Upon these general considerations are based the steps which are involved in manufacturing and delivering a source of the therapy type. These steps are:

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[*] Commercial Products Division, Atomic Energy of Canada, Limited, Ottawa, Ontario, Canada.

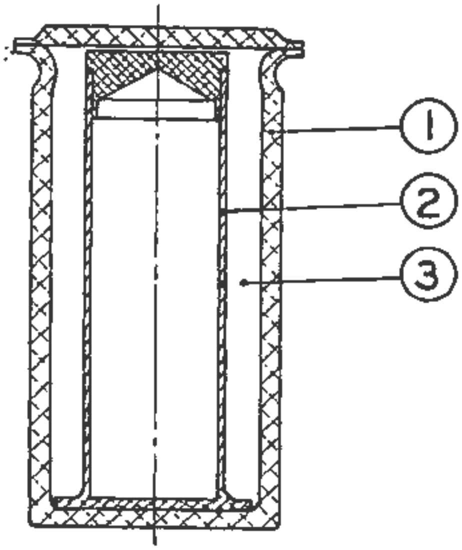

Figure 1. (1) Irradiation capsule, (2) liner, (3) annular shell

(1) the loading of inactive or active cobalt pellets into irradiation capsules; (2) the loading of irradiation capsules into irradiation rods and the rods into the reactor; (3) the neutron irradiation of the cobalt in the reactor; (4) the removal of the cobalt from the reactor to the source preparation facilities; (5) the source preparation; (6) the shipping of the source; and (7) the loading of the cobalt source into the facilities where it is to be used.

In the first step we will disregard the loading of inactive cobalt which is a straightforward operation, and we will disregard step 3, the neutron irradiation, which is beyond the scope of this paper. All remaining steps involve transferring radioactive material. In steps 1 and 5 this material is in the open while being transferred from one container to another, but in steps 2, 4, 6 and 7, it is inside sealed containers while being transferred.

Steps 1 and 5

Since steps 1 and 5 are concerned with transferring active material from one sealed container to another, it will be helpful to describe these containers. Fig. 1, item 1, shows the irradiation capsule—the sealed container which holds the cobalt while it is being irradiated. It is a circular cylinder approximately 19 mm in diameter by 4.5 cm long, made of 1-S (high purity) aluminum, with a flange on one end and closed on the other end. To this flange an aluminum disc or lid is sealed by cold welding (700 kg/cm^2 are required). In order that the cobalt pellets will shield each other as little as possible from the neutron flux, as discussed above, a closed aluminum tube or liner (Fig. 1, item 2), 12.5 mm in diameter by 44 mm long, is inserted in the capsule to form an annular shell (Fig. 1, item 3) into which 30 grams of the pellets are loaded.

Fig. 2 shows a therapy source container the design of which was developed and adopted as a standard by interested manufacturers of therapy equipment and by the source producers of the United States of America and Canada.[2] A thin, 0.5 mm, stainless steel cup (Fig. 2, item 1) which fits in the tungsten alloy body (Fig. 2, item 2) holds the radioactive material but permits the gamma radiation to pass through the end with only a 2 per cent attenuation. A tungsten alloy plug (Fig. 2, item 3) holds the source and steel cup in position—sealed by two lead wire gaskets (Fig. 2, item 4) to prevent the escape of any dust. The outer size and shape of this container have been standardized to allow its use in different designs of therapy machines, but the internal size may be altered by a change of plug to accommodate cobalt pellets, cylinders or discs, or to accommodate the various specific activity materials available. For small diameter sources, tungsten sleeves can be inserted to reduce the cavity size.

The source preparation facilities have been designed, then, to handle the loading and the unloading of these two containers although other containers of similar size and shape can be handled. These facilities—or hot cell—require first, equipment to prevent exposure or contamination of operating personnel and second, equipment to load and seal or to open and empty irradiation capsules and therapy source containers.

The first of these two requirements is concerned primarily with shielding. The hot cell shown in use in Fig. 3 is a cavity 84 cm wide by 65 cm high by 56 cm deep surrounded by poured lead encased in steel. A front window 25 cm wide by 16.5 cm high made of a high-density glass (density of 6.2 gm/cm^3) is provided for viewing all operations. This window consists of four plates of the high-density glass each 10 cm thick, and one plate, the innermost, which is a darkening-resistant cerium glass 5 cm thick. The amount of shielding used, 30 cm of lead on the front wall and 25 cm of lead elsewhere, will allow sources of several thousands of curies of cobalt to be prepared without raising the external field above the working health tolerance (7.5 mr/hr). Operators of the hot cell can thus work normal hours without undue ex-

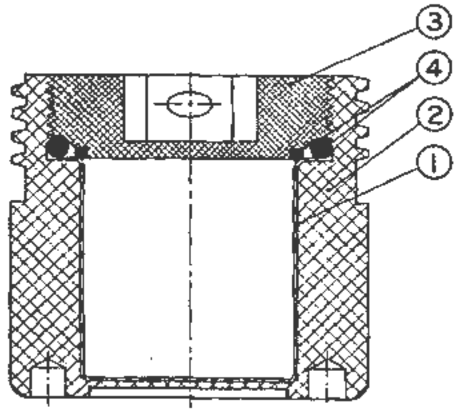

Figure 2. Therapy source container: (1) cup, (2) body, (3) plug, (4) gaskets

posure. Also required for safety is a vacuum system which maintains a negative pressure within the cell ensuring that no radioactive dust, if produced, could find its way out. The three intakes of the vacuum system are arranged near those points of operation where dusting could occur, and the exhaust of the system is passed through a filter before being released to the atmosphere.* Due to the very high specific activity involved, even the slightest amount of dust escaping from the cell could produce a serious contamination problem. Even though the shielding is greater than actually required for most of the sources prepared, film badges and pocket dosimeters are worn by the operators and a health surveyor is on hand to monitor the radiation field. This monitoring is particularly important as a safeguard during transfers of sealed containers from one shielded case to another, as in Fig. 3.

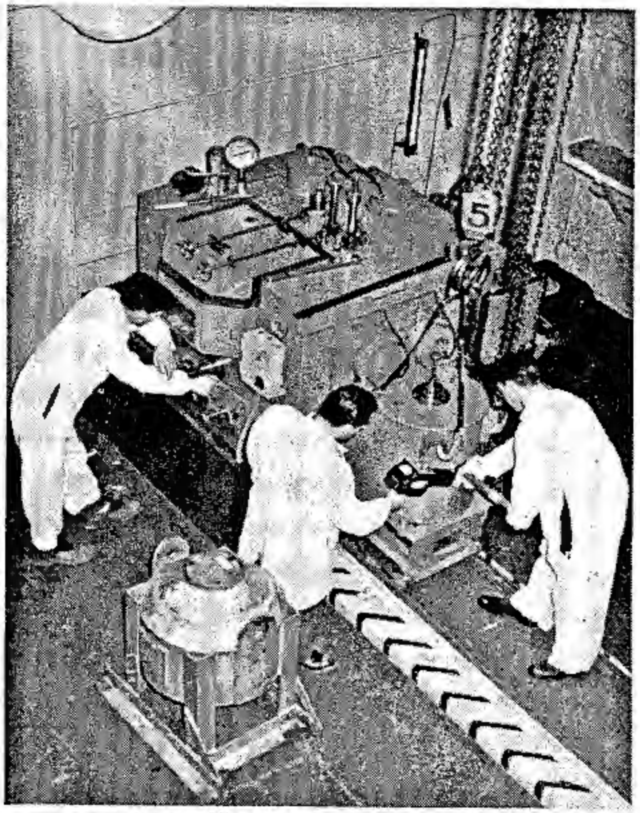

Figure 3. "Hot cell" in use for the preparation of sources

The second part of the equipment required for the hot cell is chiefly that used for actually handling the active material. Some of this equipment is used with irradiation capsule operations, some with therapy source container operations and some with both. First, all tools and accessories are mounted on stepped plugs in the walls or ceiling and are removable for cleaning, repair or exchange. This last allows a greater flexibility in the operations which can be performed in a cell of this size. Where possible all tools are arranged with preset stops to facilitate the operations. All the movements within the cell are mechanically operated through the shield by hand, except for a press and a conveyor which are hydraulically and electrically operated respectively. If necessary, the cell can be cleaned of pellets or active dust through the ports made by removing stepped plugs. The interior surface is stainless steel with the joints filled with a metallic filler to permit easier cleaning.

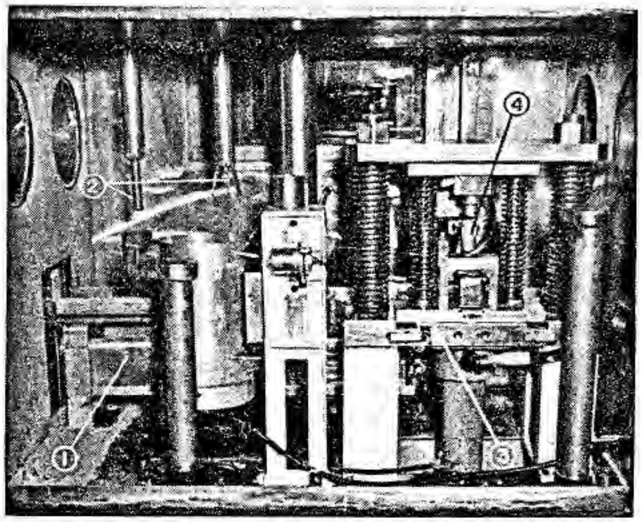

Figure 4. "Hot cell," rear view. (1) Entry drawerway, (2) tongs, (3) hydraulic press, and (4) capsule being placed in press

The operations can best be described with reference to Figs. 4 and 5 showing the hot cell from the rear and top respectively at different stages of construction. The sealed container of source material which is brought to the hot cell in a transfer-case drawer is moved into the cell as shown in Fig. 3. This entry drawerway is item 1 in Fig. 4. Directly above this drawerway is another slide on which a tray carrying three inserts can be moved completely across the cell or to the outside. The slide and the tray with

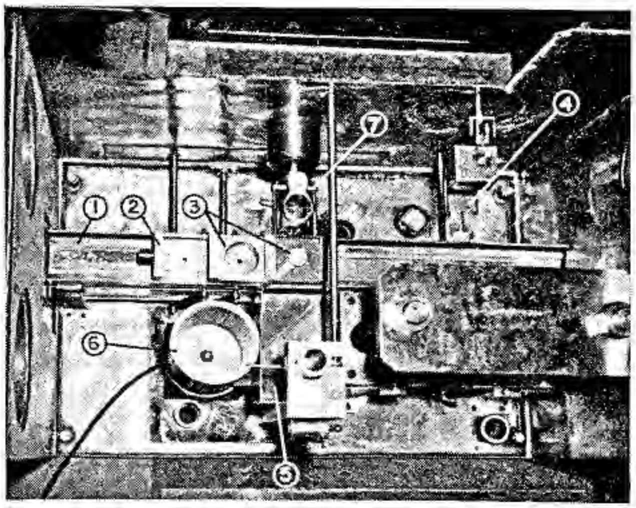

Figure 5. "Hot cell," top view. (1) Slide, (2) tray, (3) inserts, (4) tongs, (5) pellet reservoir, (6) vibrating spiral conveyor, and (7) loading tongs

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* Absolute Filter, made by Cambridge Filter Corporation, Syracuse, New York, USA.

two inserts in place are best seen in Fig. 5, items 1, 2 and 3. This tray with the appropriate inserts can carry irradiation capsules or source containers. Two vertical tongs (Fig. 4, item 2) are mounted 180 degrees apart on a plug in the ceiling. By rotation of plug either one of these tongs can be located directly above the drawerway and slide and can lift the container from drawer to tray insert or vice versa. The tray carries the container of active material to one of two positions. At one position, used primarily with irradiation capsules, is the hydraulically-operated press (Fig. 4, item 3). The press, which can be removed through a port to the outside of the cell, can be used either for cold-weld sealing of the loaded irradiation capsules, or for shearing off the capsule lids for unloading, merely by changing inserts. The tongs at the press position (Fig. 5, item 4) have longitudinal and vertical movements. In Fig. 4, item 4, an irradiation capsule has been placed in the tongs to indicate the capsule position just as it is put into the press. At the other of these operating positions is a reservoir for the cobalt pellets from which, by a vibrating spiral conveyor, the pellets can be fed, with a fine control, into the final container—either therapy source container or irradiation capsule (Fig. 5, items 5 and 6). At this loading station the tongs (Fig. 5, item 7) have longitudinal and rotary movements.

A square end wrench through the ceiling can be inserted into a socket in the plug of the therapy source container to unscrew, lift, and hold the plug during loading and subsequently to replace it into the container.

A simple spring balance made to hold an irradiation capsule and used for weighing out the amount of cobalt, can also be placed in the cell through a port. This balance, for convenience, is normally preset so that the desired amount of cobalt causes it to balance at the index.

Two other operations which have been used in loading inactive pellets are in the process of being incorporated into the loading of active pellets. It has been found necessary to outgas the pellets before loading, because of the internal pressures which are otherwise built up in the capsule during irradiation causing excessive bulging.[3] This outgassing is accomplished by heating the pellets for approximately 4 hours in a muffle furnace at 300°C, then cooling to room temperature under a vacuum and finally storing in a closed vessel until used. Also, in order that the cobalt, which is at a high temperature during the irradiation, will not oxidize, the capsules are sealed under one-half atmosphere of helium.

Illumination of the interior of the hot cell is provided by four 150 watt incandescent lamps also mounted on stepped plugs—two in the ceiling and two in the walls. Since most glass will darken under intense gamma radiation, the viewing of the operations will become impossible if suitable precautions are not observed. Thus, besides making the innermost layer of the window of a darkening-resistant glass, it has been found desirable to avoid storing active material in the cell except when it is actually being used. In addition, the lamps are left on at all times when source material is in the hot cell, so that the heat will keep the glass bulbs relatively free from darkening.

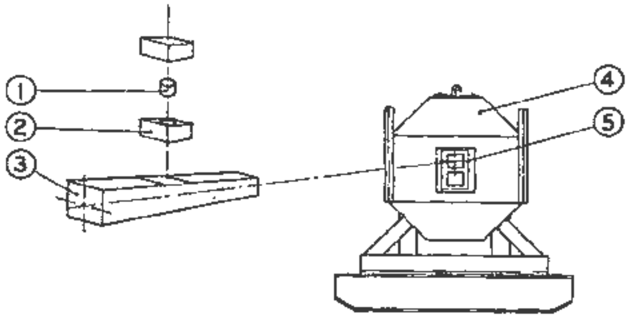

Figure 6. (1) Source, (2) insert, (3) lead drawer, (4) transfer case, (5) drawer holes

Steps 2, 4, 6, and 7

The transfer of active material, whether sealed in irradiation capsule or in final source container, is based on the use of a sliding lead drawer (Fig. 6, item 3) and a lead-shielded case. The source in a lead or tungsten alloy insert (Fig. 6, items 1 and 2) is placed in a cavity in the center of the lead drawer and in subsequent transfers, the drawer plus source move as a unit. This drawer provides a major part of the primary shielding of the lead-shielded transfer case or therapy unit in which it is placed. While there are several lead-shielded cases whose exterior sizes and shapes vary somewhat, depending on their uses, their drawer holes are standardized—as of course is the drawer cross-section (3 × 3 inches or 7.62 cm^2)—to allow the drawers to be moved freely and safely from one container to any other container.

To move or to ship a source from one location to another consists then of (*a*) sliding the source plus drawer unit from its original housing into a transfer case, (*b*) moving this case to the new location, and (*c*) sliding source plus drawer from the transfer case into the new facility or container. During the operation when the drawer is moved from one container into another, the two containers are lined up next to each other so that exposure to personnel is negligible.

To remove the cobalt from the reactor and take it to the source preparation hot cell, two lead-shielded cases are used. The irradiation rod in which the capsules of cobalt are placed in the reactor is withdrawn into the first shielded case—a tall 25-ton steel-encased lead cylinder shown over the reactor in Fig. 7. On the side of this case is a shield (Fig. 8, item 2) into which a drawer slides and through which tongs move. The capsules are then removed with the tongs and dropped into the holes in the drawer, two capsules per drawer. (Operation shown in Fig. 8.) The drawer is then slid into the second transfer case (Fig. 8, item 1), the case used to take the active material to

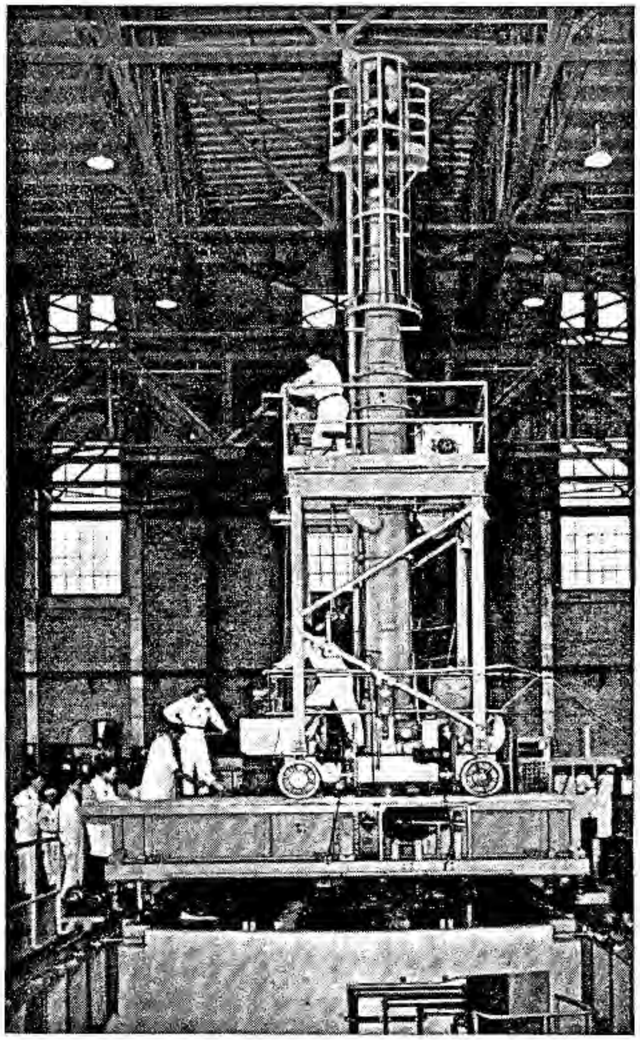

Figure 7. Removal of capsules from reactor

the source preparation cell. To reload active material into the reactor, these operations are reversed.

A two-drawer transfer case[4] in which therapy sources are normally shipped is shown in Fig. 6, item 4 and can be seen in the foreground of Fig. 3. The total weight of this steel-enclosed lead container is about 1800 kg, and the 25 cm of lead provides sufficient shielding that with 10,000 curies of cobalt in the case, the radiation field is approximately 10 mr/hr at one meter. This shielding is adequate to meet shipping regulations, for rail, boat, or truck, in most countries. For shipping, of course, all necessary safeguards such as end plates bolted over the drawer holes, asbestos gaskets, locks, labels, etc., are added. When a source in a therapy unit is to be replaced, the new source is shipped in the transfer case to the therapy unit. The drawer plus old source is drawn from the therapy unit into the transfer case and the new source plus drawer is then moved from the transfer case into the therapy unit. An empty or dummy drawer is used to follow up the full drawer during each transfer to avoid leaving an opening through which a dangerous radiation beam could emerge.

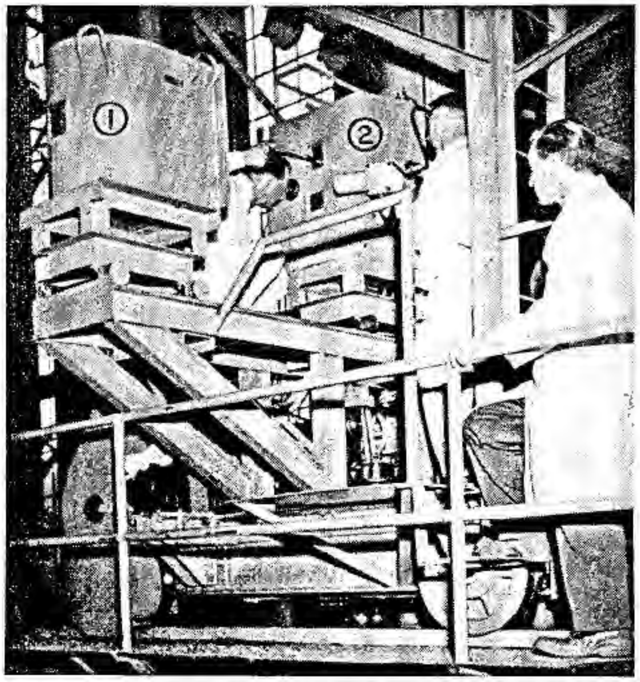

Figure 8. After removal from the reactor capsules are manipulated in the shielded box (2) into drawers which can then be slid into the transfer case (1)

To load these sources into shipping cases not incorporating the sliding drawer technique, special methods are used.

While the development of the above techniques has been based on the production of therapy sources, other types of even higher curie content have been prepared by irradiating cobalt rods or cylinders individually and then assembling them into the larger sources. A description of two such sources will indicate how these can be treated.

In one source the active material was in the form of 100 aluminum-jacketed cobalt cylinders, 6.3 mm in diameter by 25 cm in length. In the irradiation rods they were arranged in circles of 6 rods each. After the irradiation, the rods were removed from the reactor and placed in a water trench. Here with 10 feet of water providing sufficient protection, the irradiation rods were cut apart and the cobalt cylinders transferred with tongs into a cavity in a special shipping case which had been lowered into the trench. After the case was closed, it was lifted from the trench and any water in the cavity drained out through a special offset drain-hole. The total activity of the source was approximately 4000 curies.

Another source about to be fabricated will consist of cobalt slugs 6.3 mm in diameter by 2.5 cm long—also aluminum-jacketed. These slugs are also arranged in circles in the irradiation rods with 7 slugs per circle and will be unloaded in the water trench in the same way as was the source mentioned above. In this case, however, the slugs will be loaded within a lead shield into stainless steel tubes to form a "pencil" source of 9 slugs each. After the lead shield is raised just above the water level, a special heater will bake out any moisture and the pencil will be sealed by shrink fitting a plug into it. This pencil will then be dropped into a special shipping container under water as before. By this method, only a small fraction of the total activity need be handled at any one time. The total source strength in this case will be approximately 10,000 curies.

SUMMARY

It is seen that two approaches to handling high intensity sources have been used. First, for the so-called point sources (therapy type), a small cell well shielded by lead and fitted with a few interchangeable tools has been used. The cell is limited primarily by the geometry of the containers of the activity. The operations are, on the other hand, easily viewed and relatively delicate manipulations can be carried out. Second, for those sources which are geometrically large or distributed, a water-filled trench has been used. The operations here are not as readily visible and precision operations cannot be carried out. The shielding, however, is inexpensive and the shielded volume can be large. In addition, by handling the activity a little at a time, it is sometimes possible to avoid altogether the problems of a very high-intensity source. For transferring or shipping sealed sources, the sliding drawer method is seen to be adaptable and, for this type of operation, easy to carry out.

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Mechanical Arms Incorporating a Sense of Feel for Conducting Experiments with Radioactive Materials

By J. R. Burnett,* R. C. Goertz† and W. M. Thompson,† USA

The problem of carrying out experiments and operations involving radioactive materials has required the development of many remotely controlled devices. When experiments involve gamma-activity levels above a fraction of a curie, shielded enclosures called caves, cells, shielded hoods, etc. are used.[1, 2] Quite often the walls of these enclosures are constructed of thick, heavy materials. The equipment within the enclosure is generally controlled from the outside by mechanical, electrical or fluid linkages. General viewing is usually provided by large, thick, dense liquid or glass windows.[3] Caves used for experimental operations are nearly always provided with one or more general-purpose manipulators. These manipulators are used to perform many of the general operations and handling that would be performed directly if no radioactivity were involved. The greater the dexterity, speed and accuracy of the general-purpose manipulators, the less is the complexity needed in the other specific-purpose, remotely controlled apparatus.

A program to develop improved general-purpose manipulators has been active for several years. This is a report of the latest and best manipulators developed at the Argonne National Laboratory, along with some of the basic objectives and means for achieving them.

SOME DESIRABLE FEATURES FOR A GENERAL PURPOSE MANIPULATOR

When the experimentalist is separated from his apparatus by a thick wall, all operations must be performed by specific, remotely controlled equipment and by general-purpose manipulators. An ideal manipulator might be a remote artificial arm that duplicates all the motions of the human arm and hand and that might reflect back to the operator all the senses in a natural manner. This is impractical at present and for most operations is unnecessary. A more realistic set of requirements, based on the physical functions that must be performed, can be established. The following are considered to be a minimum for miscellaneous handling and manipulation in a research, "hot" laboratory cave facility.[4]

1. The manipulator must have at least seven independent motions: three to move its artificial hand or tongs about in space, three for angular orientation, and one for grasping objects.
2. The manipulator should be easily controlled in all of its motions by only one hand. An obvious and natural means to accomplish this is to have the working arm follow a master handle as a simple function of the motion of the master handle.
3. Load forces on the slave arm should be reflected back to the master arm and handle in a proportional manner. This will allow the operator to apply forces on the object with easily controlled magnitudes and directions.
4. Each motion of the manipulator should be mechanically reversible; that is, forces applied at the slave arm should cause it to move and drive the master arm in a corresponding direction. This condition is necessary because of the many operations that dictate the path of motion a manipulator must take. Turning a crank or assembling mating parts are examples.
5. Low inertia and low friction are needed so that load forces are not masked and so that forces under collision conditions are not excessive.

Additional features may be required to make the manipulator more useful for certain applications. Some of these are: addition of a forearm and elbow, remotely removable tongs, provision for protective booting, indexing of the slave arm with respect to the master arm and additional fingers.

MECHANICALLY CONNECTED MASTER-SLAVE MANIPULATORS

Several mechanically connected manipulators, which fairly well fulfill the above desirable features, have been developed. They have been named master-slave manipulators because all of the motions initiated at the control arm and handle (master) are duplicated at the working arm and tongs (slave). The newest manipulators of this type are the Model 7, for use in junior or intermediate level (up to 100 curies gamma activity) caves, and the Model 8 for use in intermediate and high level (up to several thousand curies gamma activity) caves.

Figure 1 shows the general configuration of the Model 7 manipulator. The master and slave arms are connected by a torque tube which keeps them

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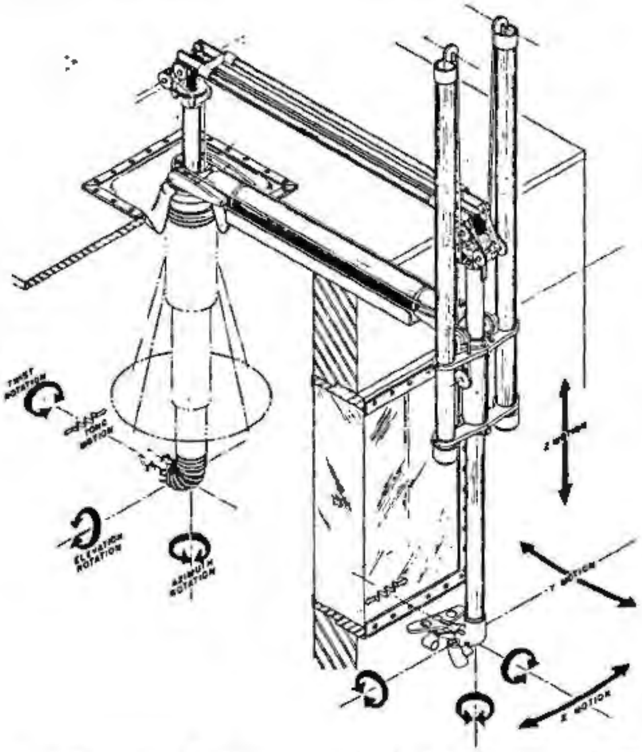

Figure 1. Mechanically connected master-slave manipulator Model 7 shown in a typical installation

in a common plane and by a linkage at the top which completes a parallelogram. The *Y* motion is provided by the parallelogram linkage. The *X* motion is provided by supporting the entire manipulator on rollers which are in contact with the torque tube. The master arm moves up and down in the *Z* direction between the two counterweight tubes and drives the slave arm up and down by 0.005 by $\frac{3}{16}$-inch Elgiloy tapes passing over pulleys and anchored at the extremities of the two arms. (Elgiloy is the trade name for a strong, corrosion-resistant alloy.)

The three wrist-joint motions and the squeeze motion are transmitted from master to slave by $\frac{1}{16}$-inch diameter stainless steel cables. The elevation and twist motions are obtained from a gear differential, Fig. 2, having the two side gears equipped with cable drums. The cable drums at the master wrist joint are connected by cables to corresponding drums on the slave side. Azimuth rotation is transmitted by cables attached to drums at the top of the inner tubes of the master and slave arms. The tong squeeze cable passes through the center of the wrist joint over small pulleys located so that there is a minimum change in its length as a function of the elevation angle.

The counterweights have the same mass as the manipulator components moving in the *Z* direction. They are located in tubes on each side of the master pivot and their center of gravity and the center of gravity of the parts moving in the *Z* motion are equal distances above and below the pivot. Since they are guided in tubes parallel to the master and slave arms, a balanced condition is maintained for all positions of the manipulator.

The manipulator is designed to have low inertia in all its motions. Most of the structural parts are made of aluminum alloys and are designed to have large strength-to-weight ratios. Friction is kept small by using ball bearings for all moving parts with the exception of some of the joints in the handle and tongs. The manipulator can safely exert a force of 10 pounds in any direction.

The slave arm of the manipulator enters a hole in the roof of the shielded enclosure and can be completely covered with a synthetic rubber boot. The boot prevents radioactive contamination and chemical corrosion of the slave arm. The tong assembly may be remotely attached to or removed from the booted wrist joint, Fig. 3, by using a special jig.

The Model 8 manipulator, Fig. 4, has the same seven basic motions as the Model 7, but is considerably larger and is designed to pass through the vertical front wall of a shielded enclosure.[5] The master and slave arms are connected to the ends of an 8-inch diameter horizontal tube which is supported on rollers attached to the cave at about 120 inches above floor level. The slave arm extends in a downward direction with no parts protruding above the horizontal mounting tube. The lower tube of the slave arm telescopes into a single upper tube to give a *Z* motion of about 36 inches. The *Y* motion of the master and slave arms is coupled together by tie rods passing through the 8-inch tube. These

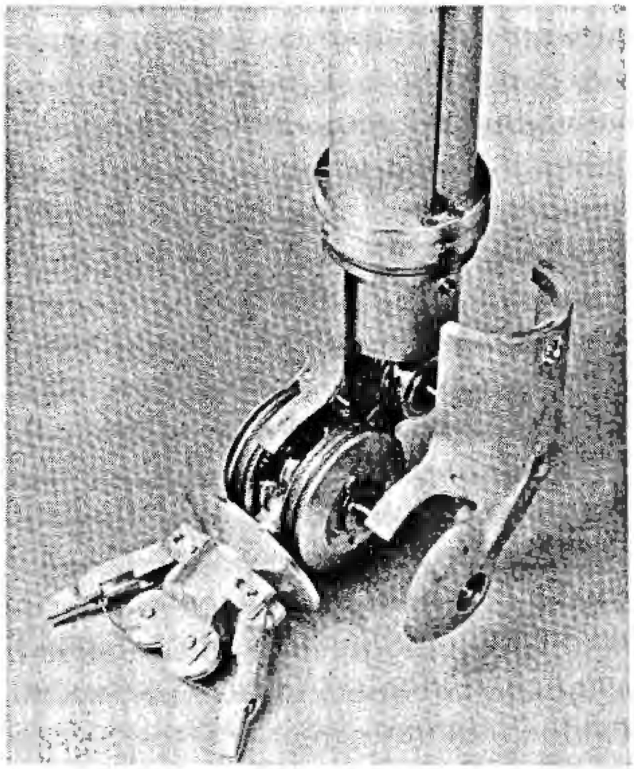

Figure 2. Wrist joint, less boot and tongs, of mechanically connected Model 7 and electrically connected Model 2 manipulators

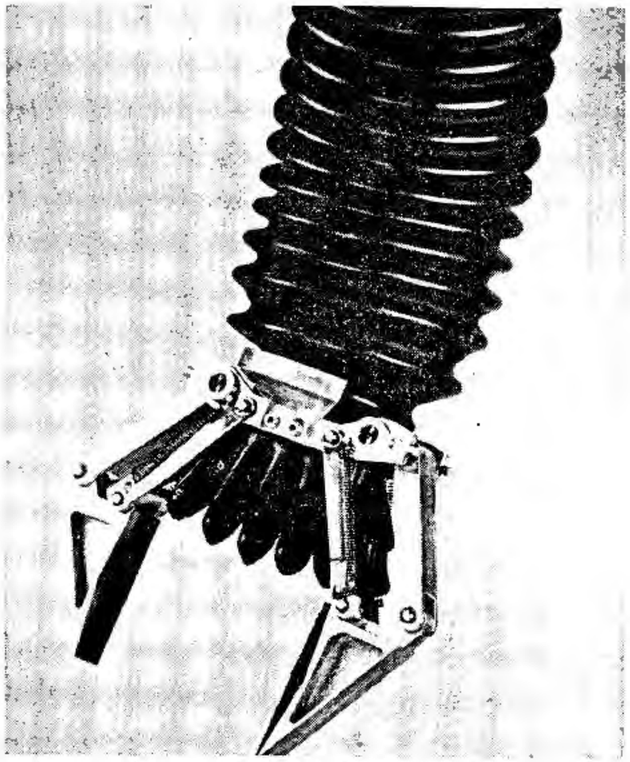

Figure 3. Booted wrist joint and tongs

rods terminate at an auxiliary parallelogram linkage on the master side which, in turn, is attached to the master arm assembly through an electric linear actuator. This actuator can be energized, causing the slave arm to be indexed in the Y direction while the master arm remains vertical. Extra volume coverage of the manipulator is provided by this indexing, or the slave arm may be driven all the way to the horizontal position. In this position, the entire manipulator may be easily installed or removed from a hole or slot in the cave wall.

Two counterweights within tubes on either side of the lower master-arm tube provide the balance for the master and slave arms as they move in the Z direction. An auxiliary counterweight is located above the horizontal pivot on the master side and is coupled to move in synchronism with the slave arm. This helps to keep the manipulator in balance while the slave arm is indexed with respect to the master arm. This counterweight provides an acceptable balance for the manipulator when the slave arm is as much as 35 degrees out of parallelism.

The elevation, twist, squeeze and Z motions are transmitted from master to slave by means of 0.005 by $\frac{3}{16}$-inch Elgiloy tapes passing over suitable banks of pulleys. When the Z motion is actuated, all of these pulleys as well as guide rollers must move; consequently, friction in these parts becomes very important. After a series of tests on various sizes and shapes of pulleys for tapes and cables, crowned pulleys with auxiliary flanges were selected because of their much lower friction. The crown on the pulley acts to keep the tape from rubbing against the flanges. The frictional drag in the Z motion of the manipulator is 8 to 10 ounces with the tapes tensioned at 30 pounds.

The wrist joint, Fig. 5, of this manipulator has been designed in a compact arrangement to accommodate the same boot and tongs used on the Model 7. The tape drums are geared to the differential side gears, increasing the effective lever arm. This reduces the compliance and increases the strength of the wrist joint so that it is capable of carrying a load of at least 20 pounds in its tongs.

Mechanically connected master-slave manipulators having moderately low inertia and low friction in all their motions inherently provide a moderately good sense of force reflection or feel, as well as a duplication of the motions of the master arm at the slave arm. Although the mechanically connected master-slave manipulators are a direct means of achieving many of the desired results, they are limited in several respects due to the mechanical linkages between the two arms. The volume that the slave arm can cover is somewhat limited and the cave wall must have a hole through which the linkages pass. Also, the work produced at the slave arm must be supplied entirely by the operator. The load capacity is thus limited by the effective strength of the operator.

ELECTRICALLY CONNECTED MASTER-SLAVE MANIPULATORS

The need for a manipulator to operate at greater distances, over a larger volume, and inside a sealed

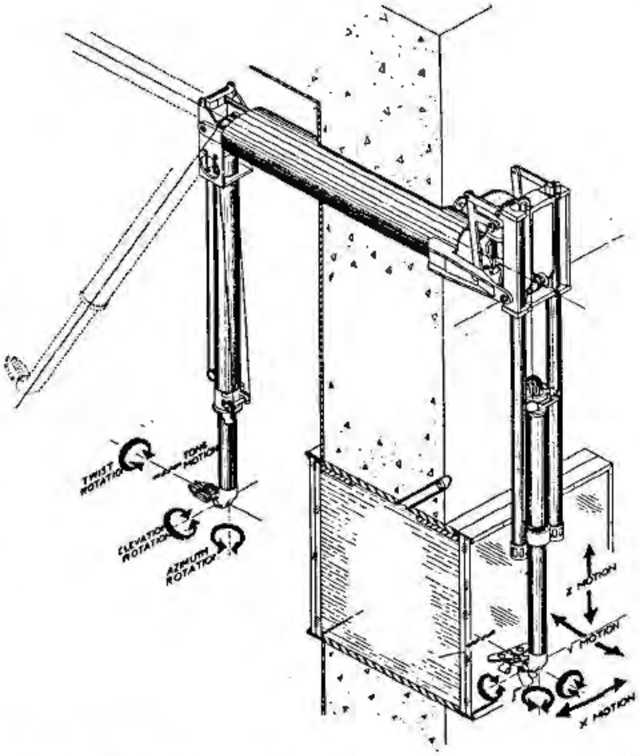

Figure 4. Mechanically connected master-slave manipulator Model 8 shown in a typical installation

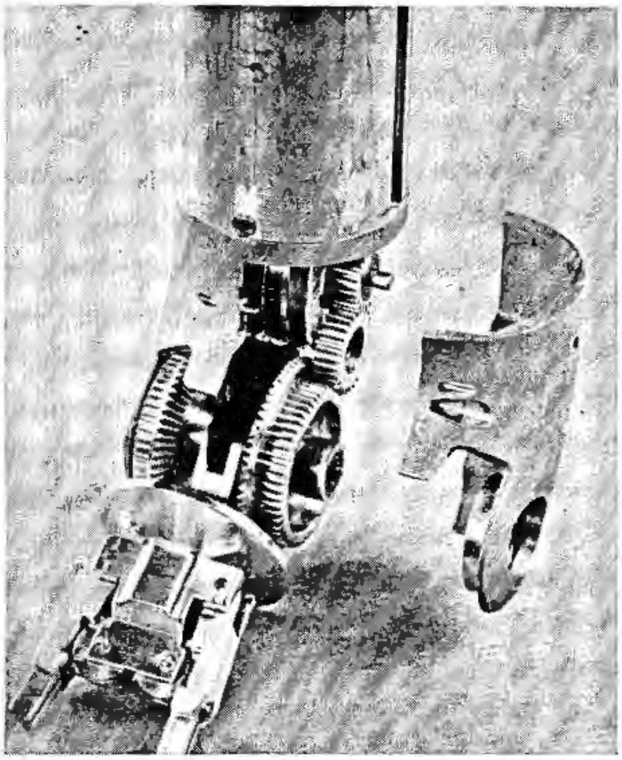

Figure 5. Wrist joint of Model 8 manipulator

enclosure has stimulated the development of electrically connected master-slave manipulators.[9] Figure 6 is an isometric illustration of the newly developed master-slave servo-manipulator, Model 2, which requires only electrical connections between the master and slave arms. It consists of master and slave arms having the same seven independent motions as the mechanical manipulators. However, since only electric wires connect the two, the slave arm can be mounted on a bridge-type crane or other mobile device.

In this manipulator the characteristics of a mechanical linkage have been duplicated by a bilateral, force-reflecting positional servo-mechanism or "feel servo."[7, 8] One complete servo is used for each of the seven motions. A symbolic diagram of the feel servo is shown in Fig. 7. The components in the servo assembly at each arm consists of a two-phase, 60-cycle ac servo motor with a directly coupled ac tachometer generator, a position data transducer (synchro transmitter on slave, synchro control transformer on master), and an efficient gear train with an output shaft. A console contains the amplifiers and limiters for each servo as well as the common power supplies. Steel cables, gears, or linkages are attached to the output shafts of the gear boxes to provide the arm motions. The servos for each motion are the same except for different gear ratios and gain constants.

The operation of the servo can be described briefly and qualitatively by examining the torques produced by the two servo motors. The motor on the slave arm produces the torque which is applied to the load. The motor on the master arm produces the torque which the operator feels. The two motors are supplied by the same current, I, producing nearly equal torques. This current is proportional to position error, E and velocity error, dE/dt. The direction of these torques is such that they tend to reduce the positional and velocity error between the two arms. This torque equality is fairly well maintained whether the system is in motion or applying a static force. The servo system is completely symmetrical and may have the input and output interchanged without affecting its operation. The system is therefore mechanically reversible and bilateral.

The tachometer generators, connected so that their output voltages cancel each other for equal speeds in the same direction, provide excellent damping for the servo system. Error and error rate signals are set so that the servo is more than critically damped for all useful loads and stable with an infinite load at the slave. The high stability is desirable so that there will be no unwanted oscillations or overshoots during operation of the manipulator.

Slow synchronization of the master and slave arms is provided to avoid damage to apparatus or the operator. Synchronization from a considerable error may occur when the equipment is turned on, after a power interruption or after an overload at the slave arm. Fast synchronization is prevented by the voltage limiter in the positional error signal channel. It is adjusted to limit just above the maximum

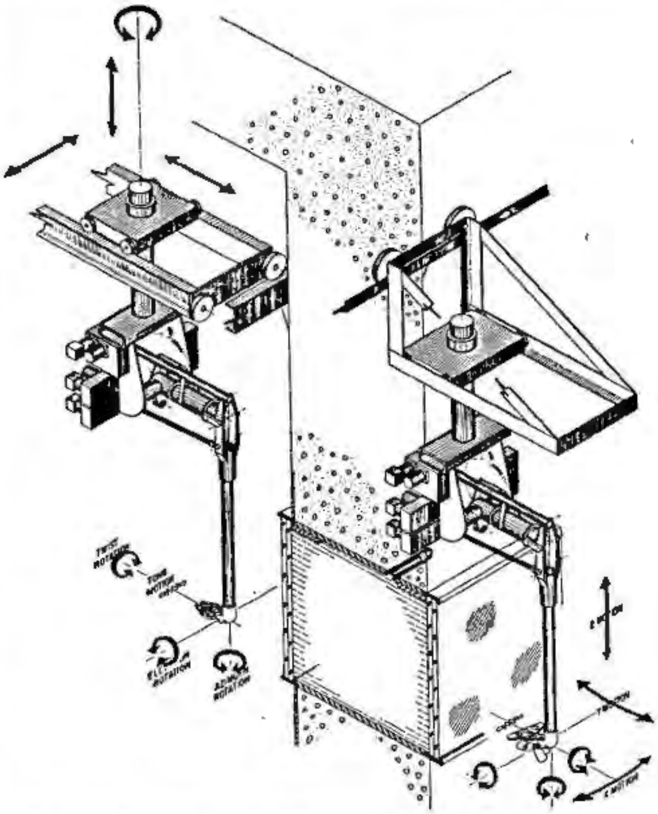

Figure 6. Master-slave servo-manipulator Model 2 shown in a typical installation

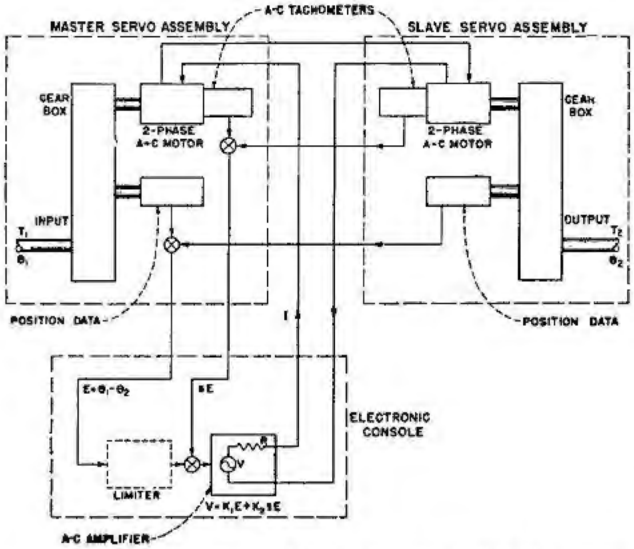

Figure 7. Symbolic diagram of feel servo used in Model 2 manipulator

power signal level. Since the error signal cannot exceed this limit for large differences in the position of the arms, the speed difference of the motors is limited by the high damping signal from the tachometers.

Each of the 60-cycle electronic amplifiers delivers about 100 watts of controlled power to the two servo motors connected in series. The two-pole, two-phase motors have a stall torque of 16 oz-in. and a rotor inertia of 0.2 oz-in.[3] The gearing and other mechanical connections give a resultant load capacity of about 10 pounds and a maximum linear speed of 3 feet/sec.

Direct gearing from the servo gear boxes to the manipulator upper arm is used as the mechanical coupling for the *X* and *Z* motions. The *Y* motion has an additional linkage. The wrist joint of this manipulator is the same as the one used on the Model 7 mechanical manipulator, Fig. 2. The elevation, twist and azimuth rotations and the tong squeeze motion are coupled with $\frac{1}{32}$-inch diameter stainless steel cables. The cables pass up inside the lower and upper arm tubes and out to drums on the gear boxes.

A single counterweight is used on each of the master and slave arms of the manipulator. This counterweight is attached by simple parallelogram linkages to the upper and lower arm tubes so that, except for wrist rotations, it gives complete balance for all positions of the manipulator. To keep the mass of the moving parts low, six of the servo assemblies are mounted to the frame structure. The *X* motion servo assembly is attached to the upper arm assembly and moves with it.

The manipulator is designed so that its various sub-assemblies, such as the servo assemblies, can be quicky and easily removed. This is important so that these units may be repaired or replaced in a minimum of time and sometimes under conditions of radioactive contamination.

ANALYTICAL PROCEDURES AND SYSTEMS UNDER INVESTIGATION

The bilateral servo used in the Model 2 manipulator is probably the least complex of a large variety of possible configurations. It is essentially two unilateral, positional servos connected back to back. The servo motors are geared directly to the input and output and therefore their inertia and friction is felt directly. This system can be extended to manipulators having two or possibly three times the load capacity of the Model 2. The high inertia of larger motors places a limit on this extrapolation.

Experimental studies have been made on more complicated configurations using both electrical and hydraulic servo units. These employ strain gages at the input and output of the servo to provide signals which are fed back to reduce the effective inertia and friction. These systems are much more difficult to stabilize over the wide range of load conditions imposed on the slave side.

The difficulty of analyzing bilateral servos using the conventional transfer function servo theory has stimulated the investigation of another approach using the fundamentals of modern network theory.[9] It is well known that a passive 4-terminal network is stable for all values of loads. Also its characteristics can be completely defined by its admittance parameters. If a servo can be represented by an analogous 4-terminal network whose admittance parameters represent a passive network, then this servo is stable for all passive loads. This is true even though there are active elements within the servo. A particular servo configuration may be studied by finding its admittance parameters and synthesizing a network from these parameters.

Admittance parameters are usually defined in terms of an electrical network. They can be redefined in terms of a mechanical network by using the torque-current, velocity-voltage analogy. Figure 8 is a mechanical network which could represent a force-reflecting servo. Its admittance parameters are:

$$Y_{11} = \frac{T_1}{s\theta_1} \bigg|\; s\theta_2 = 0 \qquad (1)$$

$$Y_{22} = \frac{T_2}{s\theta_2} \bigg|\; s\theta_1 = 0 \qquad (2)$$

$$Y_{12} = \frac{T_2}{s\theta_1} \bigg|\; s\theta_2 = 0 \qquad (3)$$

$$Y_{21} = \frac{T_1}{s\theta_2} \bigg|\; s\theta_1 = 0 \qquad (4)$$

The servo used in the Model 2 manipulator can be used to illustrate the technique of obtaining the admittance parameters for bilateral servos. Referring to the symbolic diagram of the servo, Fig. 7, the following equations can be written in transformed form:

$$T_1 - K_T I = (Js + F)\, s\theta_1 \qquad (5)$$

$$T_2 + K_T I = (Js + F)\, s\theta_2 \qquad (6)$$

$$V = K_1(\theta_1 - \theta_2) + K_2 s(\theta_1 - \theta_2) \qquad (7)$$

$$IR = V + K_b s\theta_1 - K_b s\theta_2 \qquad (8)$$

Equations 5 and 6 are torque equations where J is the motor and gear-train inertia, F the mechanical viscous friction, s the differential operator d/dt, and K_T the torque constant. T_1 and T_2 are externally applied torques. K_b is the back emf constant. All values are referred to the input and output shafts of the servos. The equations assume a completely symmetrical and linear system.

Using these equations and the defining Equations 1 through 4 the admittance parameters of the servo are:

$$Y_{11} = Y_{22} = Js + F + \frac{K_T}{R}\left(K_b + K_2 + \frac{K_1}{s}\right) \qquad (9)$$

$$Y_{12} = Y_{21} = -\frac{K_T}{R}\left(K_b + K_2 + \frac{K_1}{s}\right) \qquad (10)$$

Figure 9 is a mechanical network which can be synthesized from these parameters. Its electrical analogue is also shown. The shunt inertia and damper at each end of the mechanical equivalent circuit have the same value as the effective inertia and damping of the physical components of the servo. Only the damper and spring in parallel can be adjusted by varying the design parameter of the servomechanism. Since this is a passive 4-terminal network which was realized from the admittance parameters of the servo, the servo is stable for all passive loads.

The analysis of servos by means of admittance parameters can be extended to synthesis procedures.

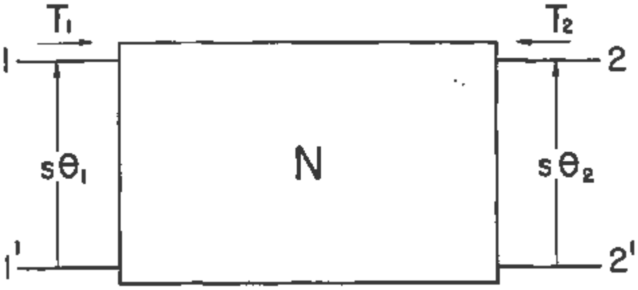

Figure 8. Four terminal mechanical network

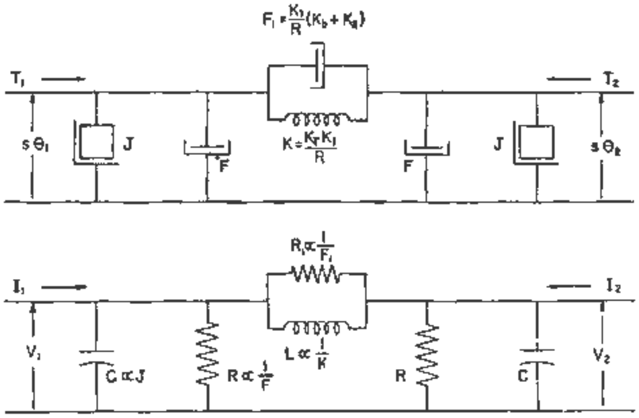

Figure 9. Mechanical and electrical network representing the Model 2 bilateral servo

It is possible to choose a passive network with certain specified admittance parameters. These parameters can be formulated from the desired performance specifications of a manipulator but, of course, are limited by the physical characteristics of the servo components. These parameters may be used to synthesize a servo system which is stable under all load conditions and meets the performance specifications.

The research and development program on both mechanically connected and electrically connected manipulators is continuing. Although mechanically connected manipulators have certain limitations, as pointed out earlier, they are being used extensively. Due to their relative simplicity, dependability and modest cost it is expected they will continue to be specified in many new facilities. Emphasis is being placed on servo-manipulators having load capacities ranging from 25 to several hundred pounds. These manipulators are needed for both research and plant operations in the nuclear power reactor program.

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Design of Radiation Analytical Facilities at Oak Ridge National Laboratory

By H. E. Goeller and E. J. Frederick,* USA

In the last few years emphasis in radiochemical research and processing has been on shorter-cooled reactor-irradiated materials with generally higher orders of specific activities. Furthermore, the increased use of enriched U^{235} for reactor fuels has made the handling of even small quantities of such materials particularly hazardous. In order to cope with this increasing hazard in analytical work at Oak Ridge National Laboratory, a new remotely operated analytical facility is being constructed. This paper reviews the basic philosophies on which the new facility was designed and describes the various design features of the actual installation.

The primary aim of the design is to provide a facility in which gram quantities of highly radioactive materials can be analyzed by trained personnel safely, rapidly, and efficiently. Thus, the first step of design is to determine the specific and total activity of the materials to be handled, and from this to determine the amount of shielding required. For α-emitting materials, such as thorium and natural uranium, regular laboratory hoods are adequate; for more hazardous α-emitters, such as plutonium and highly enriched U^{235}, dry boxes are required for protection of personnel. For β-emitters the same general assumptions hold. For γ-emission, where attenuation is exponential rather than linear, much more shielding is required, its thickness being dependent on the amount and energy of the γ-radiation and the shield material. The more commonly used materials for γ-shielding, arranged in order of increasing cost but of decreasing space required are: regular concrete, high-density concrete, iron and lead. Where large amounts of γ-radiation are involved, apertures or large unshielded openings are not permissible because of the danger of overexposure to operating personnel from scattered radiation. This factor is most serious for low-energy γ-radiations.

The laboratory under consideration in this discussion is committed to analyze samples from (1) pilot plant and semi-production reactor fuel recovery operations, (2) homogeneous reactor experimental and operating fuels, and (3) radioactive material obtained in research reactor experiments. The radioactive materials are primarily high-level β- and γ-emitting fission products. The shielding thicknesses required for fission products from fuel irradiated for more than a few days and allowed to decay for up to 120 days can be calculated adequately by considering only its La^{140} γ-activity over 1.5 Mev. The shielding for active material with decay periods in excess of 120 days can be safely calculated by considering all γ-radioactivities averaged to 0.8 Mev. In the present installation γ attenuation to 1 mr/hr outside the shield is required. Since no special space limitations were imposed on this design, regular concrete was used except in the few instances where the shield was locally thinned as in the service chase. Here, full shielding was obtained by using barytes ($BaSO_4$) aggregate for the concrete.

The analytical facility currently under construction at Oak Ridge National Laboratory is 84 ft long, 34 ft wide, and 17.5 ft high, and it abuts on an existing chemical processing pilot plant facility.

The plan of the new laboratory (see Fig. 1) covers an over-all floor area of 3665 ft^2. The laboratory is subdivided into a nonradioactive and a radioactive area by a partitioning wall extending the length of the building along the front face of the cell bank. The nonradioactive, or operating, area contains office space and normal laboratory facilities, and instrument tables are conveniently located to form a protective bay against the flow of traffic in the working area along the cell bank.

In the radioactive area are two facilities, the cell bank and a decontamination pit, which will be located in a corner of the building. Change facilities, a counting room, and supporting laboratory facilities are located in the adjacent building. Many arrangements of groups of cells are possible; however, for analytical work the in-line arrangement is preferred for the following reasons:

1. It permits use of a heavily shielded, centralized storage cell which localizes the bulk of the radioactivity and reduces the shielding required in the analytical cells.
2. It uses common shielding walls and reduces the over-all area covered by the facility.
3. It provides for economical expansion of the facility.
4. It permits subdividing the area surrounding the cells into two separate areas, a nonradioactive or operating area and a radioactive or maintenance area, by extending a wall along the front face and closing-in the area around the top and sides of the cell bank.

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* Oak Ridge National Laboratory.

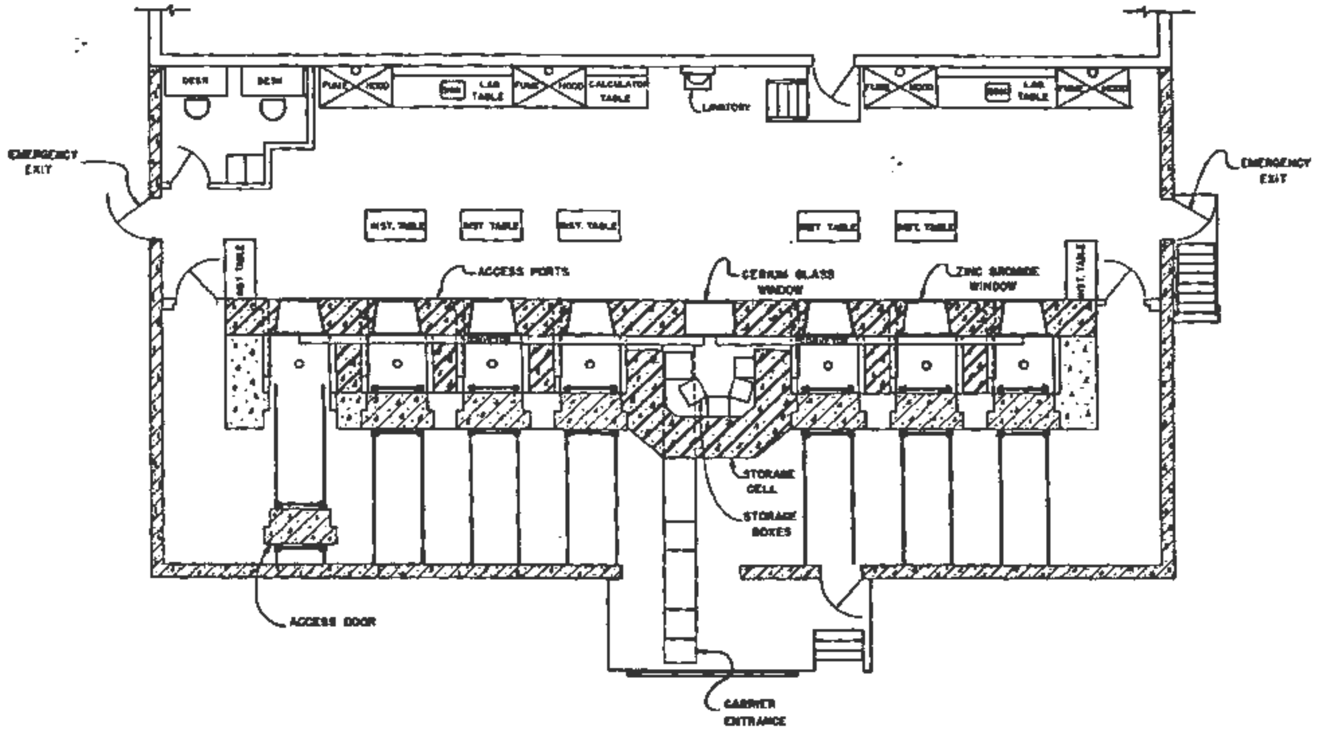

Figure 1. Plan view of high level analytical laboratory

In the present design the cell bank consists of a centralized storage cell flanked by three and four in-line analytical cells, respectively. Biological shielding is provided by 3-ft-thick concrete walls; normal-density concrete is used for the analytical cell and high-density for the storage cell.

Good viewing is essential in remote analytical work. The number of tedious and exacting operations make direct viewing preferable to operation with such indirect-viewing devices as mirrors, periscopes, or television. In order to keep a uniform shield thickness, direct-viewing windows are usually of the same density as the biological shield material. The most popular and economical is the liquid zinc bromide window. This medium has a specific gravity of 2.5, is colorless, has excellent light-transmittancy, and will not darken upon exposure to radiation. The disadvantage of accidentally losing the shielding through breakage of either cover glass can be greatly reduced by using a combination window of liquid zinc bromide and glass of the same specific gravity. The liquid windows are angled out to give a natural, wide-angled view into the working area.

Where radiation fields of higher intensity are encountered, lead glass windows of specific gravity 3.27 in combination with high-density concrete and lead glass windows of specific gravity 6.2 in combination with steel are used. The windows are assembled from 7-in.-thick glass plates to obtain the required thickness. A $\frac{1}{16}$-in. void is left between the glass plates and filled with liquid to minimize reflection. Glass with specific gravity of either 3.27 or 6.2 are commercially available as packaged units that can be inserted directly into the shield during construction. At this writing there is no viewing medium with a density comparable to that of lead.

It is imperative, where work is generally non-routine, as in the present installation, that the cell design stress flexibility and versatility. Added utility can be gained by sizing the cell for multiple operations and by selecting a manipulator of high versatility for these operations. The Argonne Model 8 master-slave manipulator is the latest development in a series of light-duty general-purpose manipulators for radioactive cell work. It has seven independent degrees of motion and has excellent feel-back qualities when action is transmitted by tapes from the master to the slave arm. This unit resembles a lower-case letter "h" in appearance, is installed through an 8-in.-diameter hole in the front face of the biological shield, and can be removed easily from the cell for maintenance. Two such units, a right- and left-hand model, are installed in each analytical cell. Only a right-hand unit is used in the storage cell. Under normal operating conditions the slave arm of the manipulator is completely encased in a plastic boot in such a manner that, when the manipulator is removed from the cell, the boot remains inside, thus minimizing spread of contamination into the operating area. This feature permits interchange of manipulators with essentially no interruption of cell operation. The initial investment for each Model 8 manipulator is approximately $4000 per arm; however, considerable savings in the cost of cell equipment can be obtained by their use since they can operate a large amount of standard laboratory equipment with little or no alteration. Specially designed analytical gadgets are required with many other types of manipulators.

One of the prime requisites of the present facility is good accessibility, both from the external areas and between cells. Intercell accessibility is required for the transfer of radioactive samples from the storage cell to the working cells and between working cells. Three types of access from the outside are required: (1) for introduction of samples on which work is to be done; (2) for introduction of reagents and small tools into the radioactive working areas; and (3) for admittance of personnel when required for maintenance or replacement of equipment in the cells.

Intercell transfer can be accomplished by sliding trays, cantilevered arms that move through an opening from cell to cell, or a continuous belt-type conveyor. The last type is best suited for analytical work because it permits continuous contact between the storage cell and any of the analytical cells as well as between analytical cells, and is used in the present design. It can be driven either electrically or manually. The possibility of overexposure of personnel to radiation during maintenance in any of the analytical cells by a high-activity sample passing through the cell on the conveyor can be prevented by inserting the sample in a small lead-shielded carrier during transit.

In the present installation radioactive samples will originate in the chemical processing pilot plant, on which the new laboratory abuts, or in other buildings. Samples from the adjacent building will be transported overhead in a steel-shielded chain conveyor and will enter the storage cell through the roof at a point above one of the storage boxes. Each sample will be unloaded by an automatic pick-off device. Sufficient capacity is designed into the pick-off arm to permit a buildup of samples which can be unloaded at the operator's convenience. When samples arrive at the facility from other buildings, the large shielded carrier containing the sample will be unloaded from a truck bed by a 2-ton monorail hoist and lowered into a carrier trench onto a motorized cart. Access for loading into the trench is provided both at the loading platform outside the building and at a point inside the building. The electrically driven cart is actuated through the tunnel to a predetermined fixed stop beneath the storage cell. A nonrotating automotive-type hydraulic cylinder raises the carrier and cart pallet through an opening in the floor into the storage cell; a ¾-ton hoist is used to remove the carrier lid, and the sample is removed from the carrier with the manipulator. A 6-in. viewing plug located in the front shield of the storage cell is used to double-check alignment of the carrier with the lift before the hydraulic lift is operated. The locations of these devices are shown in Fig. 2.

A small transfer drawer, constructed of the same material and built as part of the biological shield, is

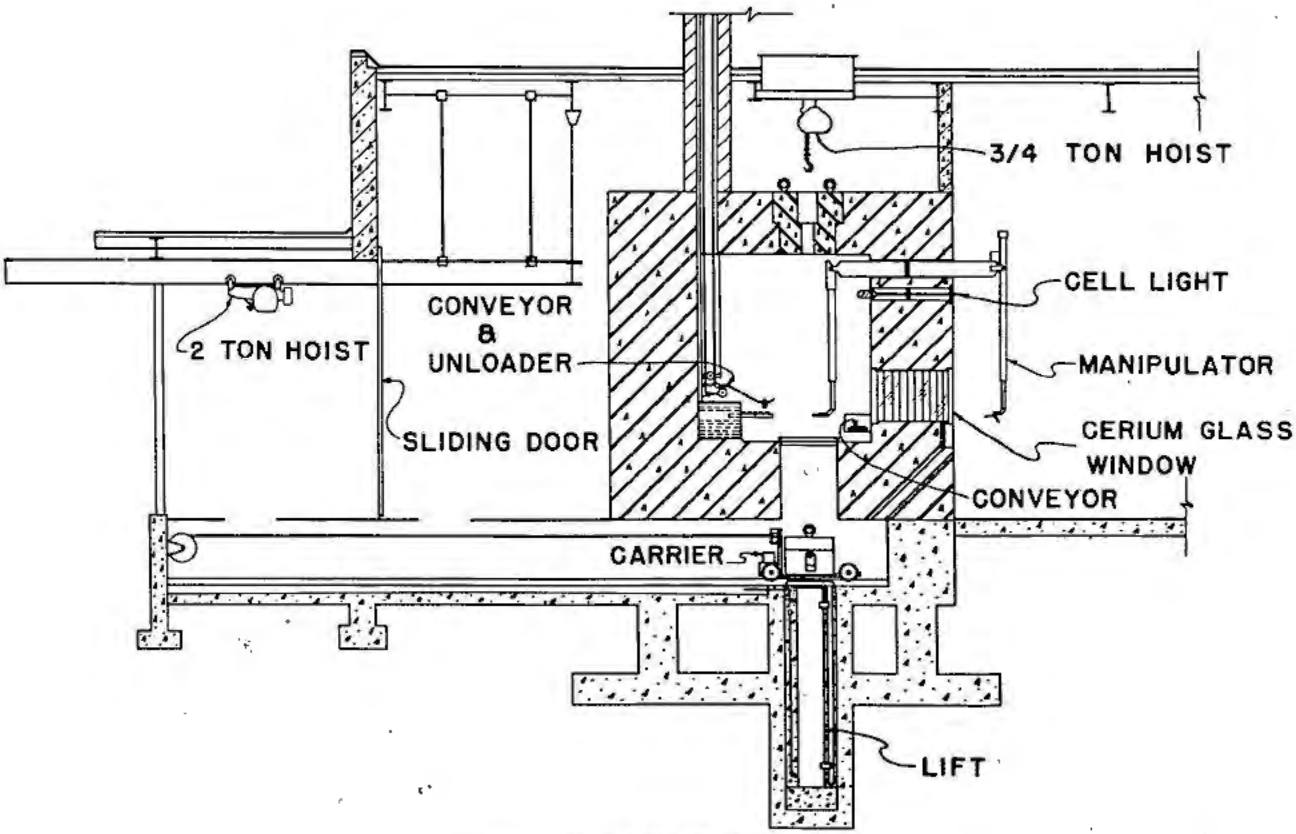

Figure 2. Section through storage cell

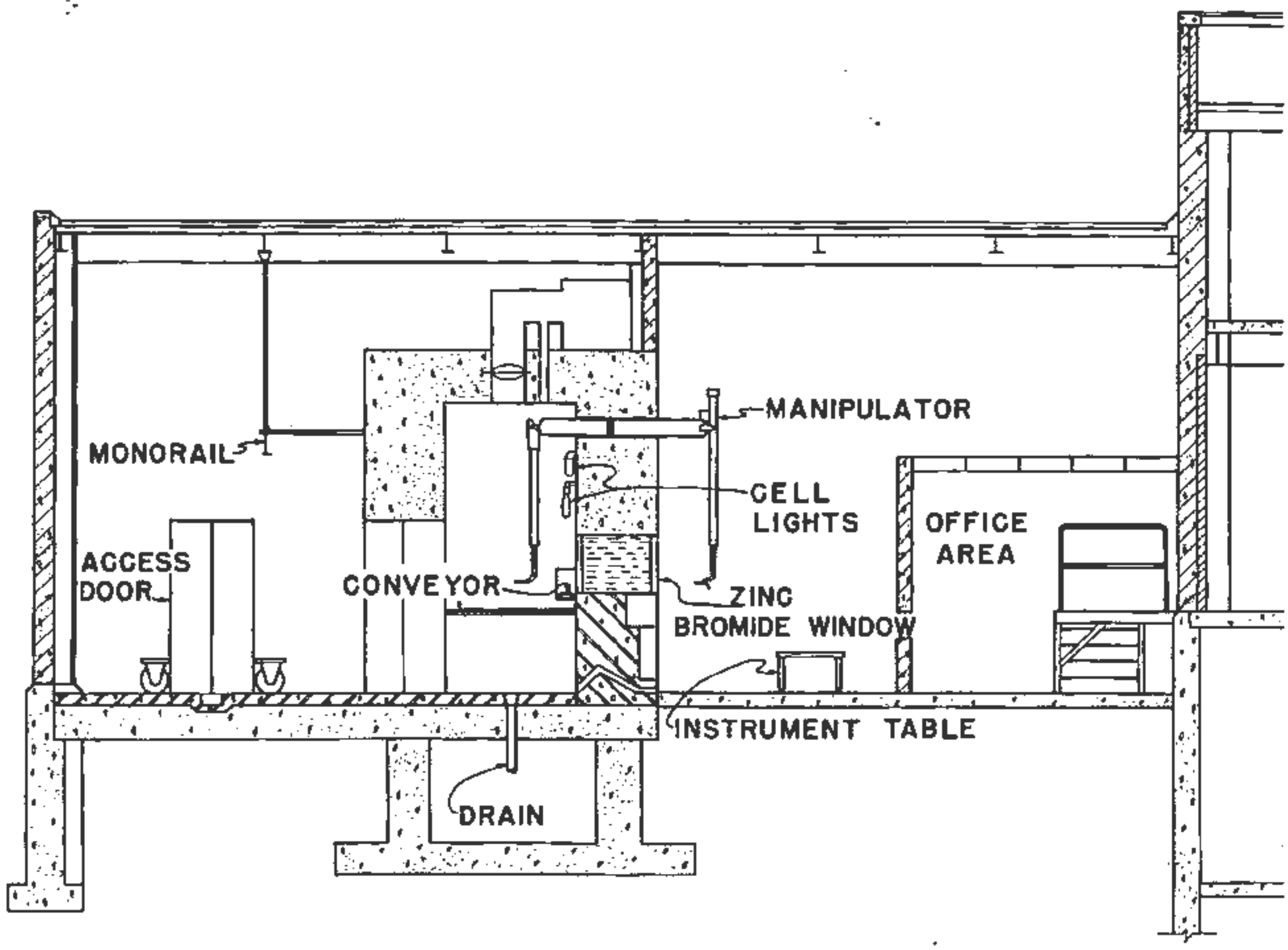

Figure 3. Section through work cell

very effective as a means of admitting reagents and tools into the cell. This type design is much more economical than either lead or steel doors, which require special fabrication with offsets in door and cell shield to prevent leakage of radiation through weak points in the shield. The transfer drawer is fabricated from an 8-in. channel, 5 ft long. It is filled with 8 in. of high-density concrete with a cross section 8 in. by 9 in. In the center of the drawer is a 10-in.-long cavity in which items to be introduced into the cell are placed. The drawer moves on standard conveyor rollers through a hole in the cell front face.

Periodically, access into one of the analytical cells is required in order to maintain or to change the equipment. Generally, large doors or removable roof slabs are used. In the in-line philosophy of the present design, where the rear area is designated as the radioactive area, a large removable door at each cell opening into this area is recommended. With this system equipment in this area can be repaired with a minimum amount of decontamination and loss of operating time. This arrangement is shown in Figs. 1 and 3. This door is mounted on a dolly and is 6.5 ft high, stepped from 5 ft to 4 ft inside width, and is 3 ft thick. The shield is of stacked concrete blocks and weighs 18,000 lb. The dolly moves on rails and is operated by a ¾-in. reversible ratchet wrench to overcome inertia.

For cell lighting, sodium vapor lamps have been the accepted standard for years. Tests made recently at this site show that, in analytical work where all operations are viewed perpendicularly through a cell window, monochromatic aberrations are practically nonexistent. Therefore, lower cost fluorescent-type mercury vapor lamps are being used in the present installation. In addition to having a higher luminary output and being less expensive, the grade of light is superior to sodium vapor lighting in that it permits better perception and differentiation of colors than do sodium vapor lamps. Each cell is provided with two sets of two 400-watt fluorescent lights and two incandescent maintenance lamps.

In the new facility air conditioning is employed to control both temperature and humidity. In order to control contamination, the air flow pattern is from the operating area into both the rear maintenance area and to the cells and from the maintenance area only to the cells. A continuous flow of air is forced from the operating area through a 3-in. offset duct into the cell, from which it exhausts to a blower suction serving all cells via a 3-in. duct into an exhaust manifold. When in use the cells are maintained at about 1 in. of water vacuum relative to the operating area and at 0.5 in. relative to the maintenance area.

When the large rear access door is opened, an automatic electrically controlled damper is opened in a larger duct to assure an airflow of 100 ft^3/min. This is used to prevent back flow of air from cells in use to the cell being maintained. The exhaust air is normally unfiltered and ties into a radioactive off-gas header which services this area. However, air filters will be provided for use under special operating conditions.

A section through the storage cell is shown in Fig. 2. The inside dimensions are 7.5 ft wide, 6.75 ft deep, and 7.5 ft high. The thickness of high-density concrete shielding is 3 ft on the sides and 2.5 ft in the ceiling. The lead glass window is 3 ft wide, 2 ft high, and 3 ft thick.

The storage cell is considered a nonaccess area. Therefore, each piece of equipment is designed for mounting on plugs or for removal from outside the cell. In the event that either of the sample-unloading devices breaks down and cannot be repaired immediately, single samples may be introduced into one of the analytical cells through the rear maintenance doorways. The lights are mounted on removable wall plugs for easy replacement.

The storage boxes for samples are simply 20-in. cubes fabricated from ¾-in. plywood and painted with Amercoat. Each box has five trays with a capacity of 100 bottles per tray. The trays are dowel-mounted on ball-bearing file-drawer slides and can be operated very easily with the manipulator. The whole unit is considered expendable. The storage boxes are mounted on a small ledge in the cell to prevent spills from running underneath the boxes.

A section through one of the seven identical analytical cells is shown in Fig. 3. The inside dimensions of the cells are 6 ft wide, 5 ft deep, and 11 ft high. Biological shielding on the front face, 3-ft-thick barytes concrete, to the 6-ft level, is used to permit recessing of services, valve handles, and small instru-

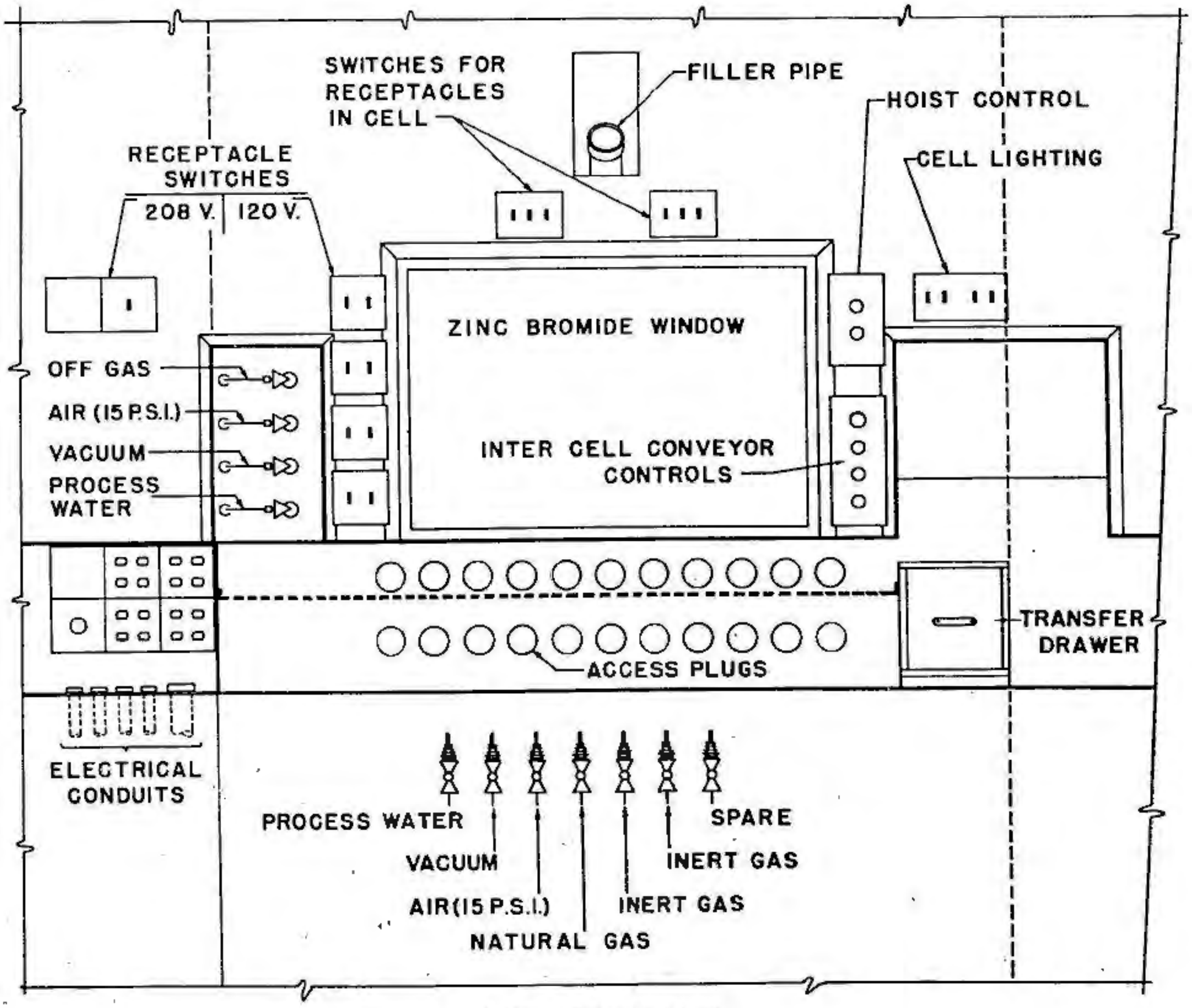

Figure 4. Typical cell front

ment controls in the shield. The upper front section and the back sections are 3-ft-thick normal concrete, and the rear access door is 3-ft-thick stacked concrete blocks. The roof section is 2-ft-thick normal concrete. The intercell partitions are 3-ft-thick poured normal concrete, 38 in. high, and 2-ft-thick stacked barytes blocks from 38 in. to the ceiling. The work surface is approximately 40 in. from the floor and consists of either a loose-fitting plywood or stainless steel pan supported by a welded steel grating suspended on the 6-in. concrete ledge at the 38-in. level. The cell interior is painted with white Amercoat 33 and, when required, will be covered with a removable strip coat for easier decontamination.

Standard laboratory services, including air, distilled water, vacuum, natural gas and electricity, are supplied in duplicate to the analytical cells (see Figs. 4 and 5). One set, excluding natural gas, is located inside the cells and is controlled by valve extension handles to the outside of the cell. The other set is run in the lower service chase of Fig. 4, and lines are run through the access plugs into the cell when required.

Electrical outlets are located outside the cell in a recessed chase beneath the cell window. This chase also provides space for small control boxes and instruments, thereby keeping the cell face free from obstruction. Larger instruments and control boxes are located on an instrument table behind the analyst. Conduits for instrument cables run from these tables under the area floor into the cells.

Each analytical cell is equipped with four types of drains: a nonradioactive drain, a cell floor drain, a radioactive waste drain, and a radioactive salvage drain. Except for the cell floor drain, these drains are shown in Fig. 5. The cell floor drain is indicated in Fig. 3. It should be noted that all cell radioactive drains are located on the opposite side of the cell from the transfer drawer. All service utilities and drains are constructed from stainless steel pipe.

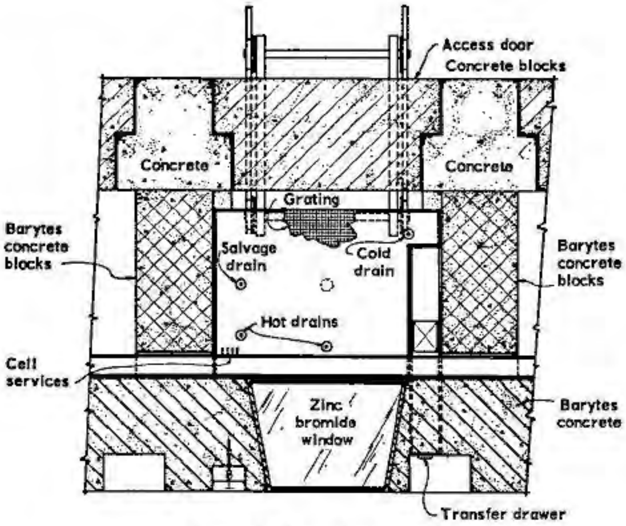

Figure 5. Plan of work cell

The laboratory will normally utilize standard analytical routines and techniques. Some of the more common operations that will be performed include specific gravity determinations by the falling drop method, pipetting and dilution of highly radioactive samples, polarography, colorimetry, volumetric and gravimetric analyses. Specialized work will include spectrographic analysis of highly radioactive solutions and solids and the study of radiation damage to solids, solutions and slurries.

Methods of Handling Multikilocurie Quantities of Radioactive Materials

By A. F. Rupp,* USA

The handling of discharged fuel elements, irradiated target materials, processed waste solutions and gases, and the kilocurie amounts of dry fission product chemicals is an important part of all radiochemical processing operations particularly at high-radiation levels. In addition to the actual handling operations, it is often necessary to perform mechanical operations, such as cutting, sawing, welding, soldering, and the like, on highly-radioactive materials or highly-contaminated equipment. While fairly simple means of handling can be employed with modest amounts of radioactivity, in working at the kilocurie level, complete precautions must be taken for safety; since, for example, 1000 curies of 1-Mev gamma emitter would produce a radiation level of about 6000 r/hr at a distance of one foot, or enough radiation to give a lethal dose in a relatively short period of time. In handling solids, one of the oldest methods was to handle the material at a distance in air, perhaps as much as 10 or 20 feet, using long tongs, hooks, or other similar devices. Solids, such as fuel pieces, highly-radioactive cobalt-60, are also handled under water and with manipulators inside of suitably-designed cells. Dried fission products, on the other hand, would be very difficult to handle in anything but a cell equipped with the proper type of remote control manipulators. Handling radioactivity in the form of fluids is perhaps the easiest method, and the most common. In this case, the material is transferred through pipelines which may either be three to six feet underground for shielding purposes or within cells; or in some cases, under water for shielding. Fluids, such as water containing radioactive materials, or chemicals, such as nitric acid with radioactive chemicals in solution, can be transferred by using steam jets, air jets, pumps, pressure vacuum pots, and other devices in order to move the liquid from one place to another. Remotely-controlled valves can be used to divert the liquids from one vessel or location to another. Almost all handling of wastes in the tank farm areas is done by jets, although the more recently-developed types of pumps, particularly those that are completely sealed in order to prevent leakage, allow a greater usage of pumps than was formerly the case.

DISTANCE FOR ATTENUATION OF RADIATION

When handling large amounts of radioactivity exposed in the air, the two principles which are adhered to are shortness of exposure and maximum distance compatible with the operation. Other factors to be considered are contamination which may be spread into the air, scattering of radiation from surrounding objects, and the necessity for isolating the area to avoid accidental exposure of personnel. Fairly cheap means of handling solid radioactive materials at a distance behind earthen barricades using wires and pulleys can be devised. For making measurements on highly radioactive materials, towers equipped with cables and pulleys for hoisting radioactive materials into the air can also be used. Remote equipment hydraulically operated, and pneumatic transfer devices, such as tubes containing projectiles which are transferred by air pressure, are useful. Pneumatic tubes of this type are used to insert small cans containing materials to be irradiated into the reactor while it is in operation; the cans may be removed to just outside the reactor shielding or the pneumatic tubes may be extended to nearby laboratories. This is particularly useful when working with short-lived radioactivities.

UNDERWATER HANDLING

The ORNL graphite reactor has a large canal at the back of the structure through which the irradiated uranium slugs can be discharged for storage and handling. It is constructed of concrete and formed in the shape of an "L," with water ranging from 9 feet deep to 20 feet deep at various parts. For the most part, the canal walls are unpainted; a special part known as the "deep pit" has bottom and sides of white glazed tile. Fuel elements which are completely enclosed in aluminum, and other pieces of material from which activity is not easily leached, can be handled with long tongs under water and stored in bins or buckets for long periods of time, where they may be easily identified and used as needed. Illumination in the 9-foot sections is provided by powerful floodlights above the water, but submarine-type lights such as those that are used for marine salvage and diving operations are required in the deep parts, or for special work. Other materials that are handled

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* Oak Ridge National Laboratory.

are slugs containing as much as 10,000 curies of cobalt-60, pieces of contaminated equipment, and other objects which are being temporarily stored to allow the radioactivity to decay before final disposal.

The long tongs that are used for manipulations under water are especially designed, and for deep operations often have a float along the tong handle to support part of the weight. An improvement in a canal of this kind would be to have the walls painted or covered to prevent leaching of calcium and magnesium into the water and allow easier cleaning of radioactive accumulations from the walls. Iron hydroxide (which adsorbs radioactivities from the water) is scraped from the walls and sucked up from the floor with a device similar to that used for cleaning swimming pools. The best coating for canals or other pits for underwater handling would be liners of glazed tile, aluminum, or stainless steel, which have smooth surfaces that can be easily cleaned and decontaminated.

The purity of the water in the canal should be high, preferably demineralized water, and the system should be so arranged that the water can be pumped from the canal through a filter and ion exchange equipment for continuous re-purification. Materials stored under water, such as cobalt-60, sometimes allow activity to leach out; or uranium slugs have imperfections in the jackets that allow fission products to dissolve in the water. It is therefore necessary to have a filtration and ion exchange system to keep the amount of radioactivity in the water low, as well as to maintain the chemical purity of the water. Water of high clarity is required for good visibility in the canal. Careful pH control of the water is required, since it has been observed that if the water is too acidic more material is dissolved from submerged objects; if the pH is too high, a haze is produced in the water, probably from the accumulation of very finely divided hydroxides.

Certain operations using tools can be performed under water with relative ease; for example, fuel pieces, slugs, and pieces of contaminated equipment can be held in the jaws of a vise under water and the pieces sawed off with an ordinary hack-saw attached to the end of a long special pole. Other tools such as wrenches, screwdrivers, pliers, and the like, can also be used under water. Many ingenious adaptations of tools of this type have been successfully made. Some power tools can be used under water, as, for example, a grinding wheel that is used to cut fuel pieces or slugs into small slices for experimental purposes. A fully-enclosed electric motor is used to power the wheel, and the grinding wheel section is separately enclosed and has a circulating water system which passes the water through a fine filter to remove the radioactive particles generated by the grinding operation and prevent it from gaining entrance to the canal water. Other power tools, such as impact wrenches, drills and hack-saws, can be used under water. There is considerable precedent and technology built up for underwater work of this kind in marine salvage operations.

While handling under water is one of the older methods used for very large quantities of radioactive materials, there is renewed interest in performing some selected types of radiochemical and related operations under water, such as the operation of pumps, submerged valve pits, canals containing multiple pipelines, submerged tanks containing radioactive materials, etc. Here again the type of operation selected must be one in which there is little hazard of spreading contamination throughout the pool of water and thereby producing large quantities of highly-contaminated water to be disposed of.

Extremely large or heavy objects can be easily handled under water because of the ease with which the extensions of lines from overhead cranes and hoists may be manipulated under water. Very often underwater handling is the only means available for handling large, heavy objects or pieces of equipment.

Finally, it should be mentioned that underwater handling need not be confined to cells or zones that are permanently flooded with water. It is possible to have equipment located in pools which can be suitably drained, or spaces where equipment can be operated part of the time suitably shielded, but flooded with water when the shielding cover to the pit or cell is taken off. It is probable that increasing numbers of designs will be seen of equipment for handling radioactivity under water or in locations that can be flooded with water for certain types of maintenance operations.

REMOTE MANIPULATORS

The most versatile of all handling arrangements is the heavily shielded cell having manipulators—mechanically, hydraulically, or electrically controlled—which can be operated as in a master-slave fashion by an operator on the outside of the cell who makes observations through a thick, high-density, zinc bromide or glass window to guide his work. Many smaller variations of this particular type of operation are used, ranging from open lead barricades into which indirect observations are made through mirrors using over-the-barricade tongs or tongs operating through ball joints—to the most advanced cells containing master-slave manipulators of the type developed by the Remote Control Section at Argonne National Laboratory.[1] Several of these cells have been used in radioisotope operations at Oak Ridge National Laboratory for handling of very large quantities of cobalt-60 and fission products. In handling bulk dry fission products or similar radioactive solids, a cell of this kind is essential.

The first cell constructed at ORNL for handling cobalt-60 had walls of barytes concrete (specific gravity, 3.6) 2 ft thick and a 2 ft × 3 ft window of similar thickness, containing pieces of special 4 in.-thick plates of Corning cerium-barium high density glass with the spaces between them filled with min-

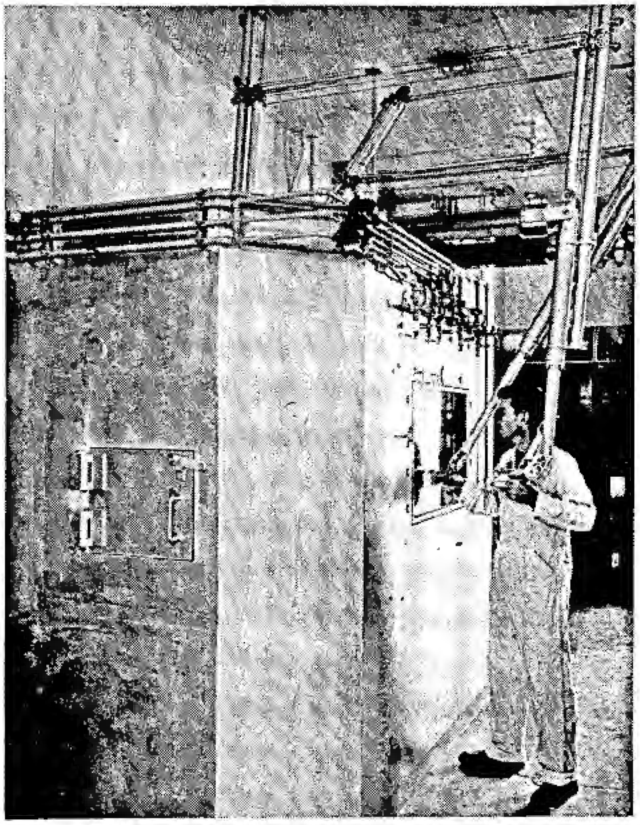


Figure 1. Remote manipulator cell for 500 curies of cobalt-60

eral oil. Type 4 master-slave manipulators were used. The radioactive material capacity of this cell is approximately 500 curies of cobalt-60 (see Fig. 1).

Of particular interest is the fabrication of a 1540-curie cesium-137 teletherapy source which was done in this cell.[2] Cesium was received from associated process equipment as the purified chloride solution and dried in a Vycor beaker in the cell where the operation could be closely observed. The cesium salt was thoroughly dried at just below its melting point (600°C), and then scraped from the vessel with an ordinary spatula, the mechanical hands being invaluable for this operation. A hydraulic press was put inside the cell and with suitable dies several pellets of the pure cesium chloride were pressed out at 20,000 psi pressure. The pellets were then weighed on an ordinary balance and the height measured by making observations through the window with a cathetometer and the density calculated. The actual weight and known specific activity of the material gave an accurate measurement of the amount of cesium in each pellet. The pellets were then placed inside a stainless steel container and a top plug secured in place, which then was soldered with soft solder. This container was then slipped into another stainless steel container and another plug was silver-soldered into place, using the manipulators to perform the soldering operation with an ordinary torch, with the help of a mechanical device to rotate the source slowly. During the operation of this particular cell, almost every type of mechanical manipulation has been done, including the use of screwdrivers, wrenches, pliers, presses, vises, special hammers, tongs, forceps, and all types of chemical operations, using special tools to handle glassware (see Figs. 2 and 3). For high-radiation-level operations involving many complex operations, especially those requiring special tools, the manipulator cell is practically a necessity.

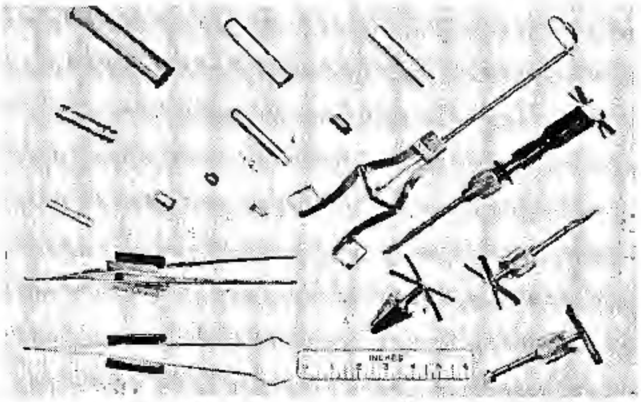


Figure 2. Tools used with remote manipulators

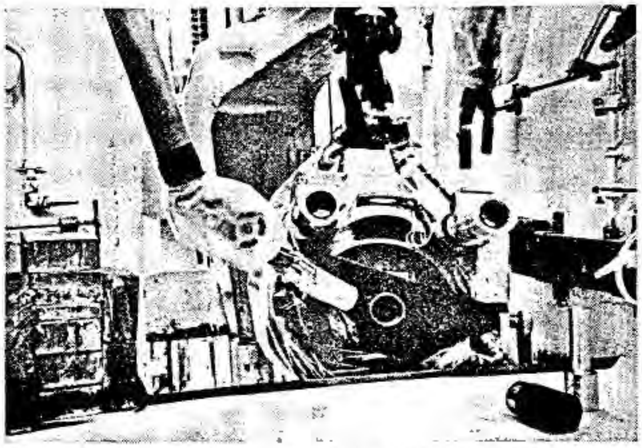


Figure 3. Loading 1500-curie source into teletherapy shield

TRANSPORTING HEAVY CONTAINERS

The particular manipulator cell described has heavy lead doors in the back, opening out into a road outside of the building. The radioisotope area is equipped with a number of portable gasoline lift trucks that can handle containers weighing up to 7000 pounds. Other similar equipment around this laboratory of course can handle even larger containers, weighing as much as 10 tons, if necessary. A great variety of lead containers has been developed for handling various kinds of radioactive materials which all follow a somewhat general design, being fabricated of a lead-filled outside casing of stainless steel (for easy decontamination) and an inner hole for holding the radioactive material, also lined with stainless steel. A lead plug, completely enclosed in stainless steel, suitably stepped to prevent radiation leakage, is provided as a closure. The walls and plug are often as much as 12 in. thick and weigh several tons (Fig. 4). The containers are designed so that they can be picked up with suitable yokes or cables and lifted by hoisting equipment. The gasoline lift trucks are especially useful for this purpose and will easily handle and transport heavy lead containers and place them into small areas with considerable precision.[3] High-level cells are designed so that a lift truck can place heavy containers into the back portion of the cell, after which the doors are closed and a small hoist on the inside of the cell can be used to remove the plug. The radioactive material can then be taken out either by the hoist or by use of the manipulator hands.

CELL FOR HANDLING 10,000 CURIES OF COBALT-60

The increasing size of the teletherapy irradiation source program has increased the need for facilities to handle cobalt-60 in very large amounts and led to the construction of an extremely high radiation level manipulator cell, shown in Figs. 5 and 6. This huge cell has walls 3 feet thick made of barytes high-density concrete, and a window of Corning high-density glass of the same thickness, composed of 4-inch-thick plates with the spaces between filled with mineral oil. Argonne Type 8 through-the-wall manipulators are used so that the cell is fully en-

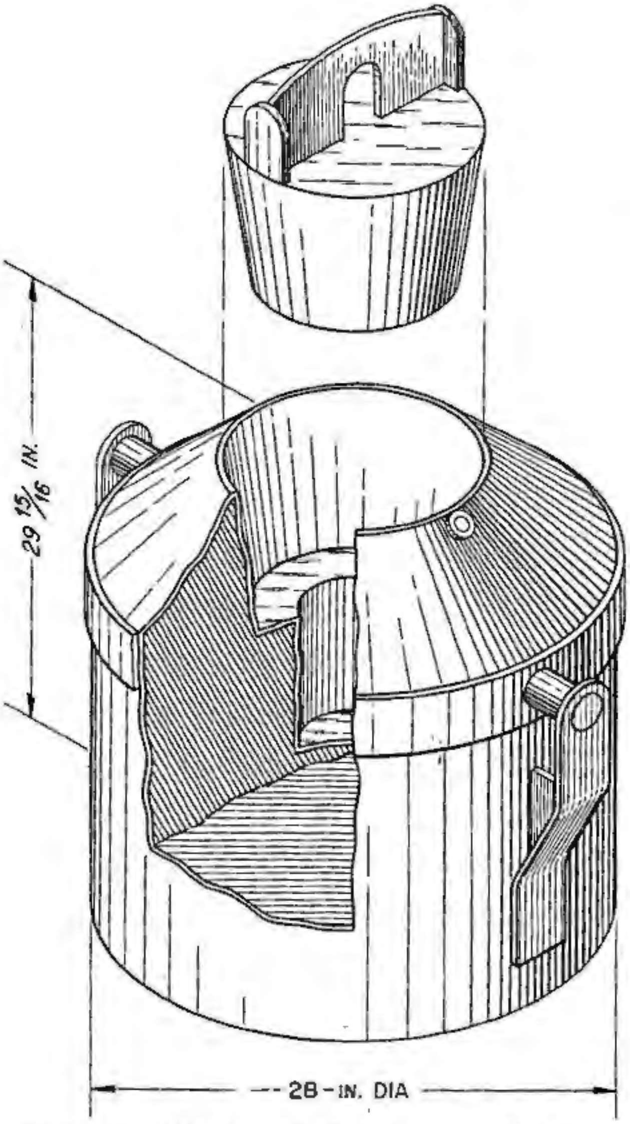


Figure 4. Heavy shielded carrier for large amounts of radioactivity

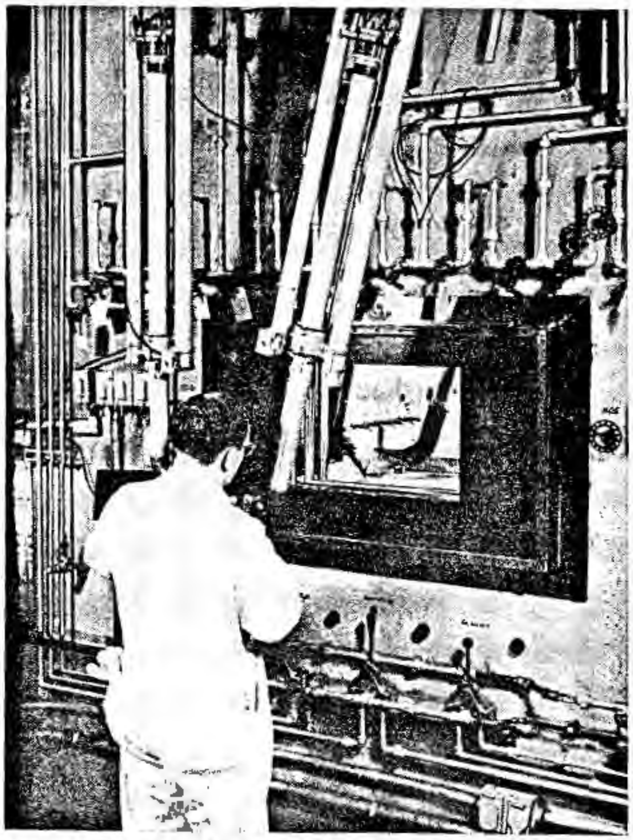


Figure 5. View through window of cell for 10,000-curie amounts of cobalt-60

Figure 6. View of back of cell for 10,000-curie amounts of cobalt-60

closed, eliminating the problem of scattered radiation. The entire back end of the cell can be opened by means of two mechanically-operated, full-height doors so that the entire vertical cross section of the cell, 4½ feet wide by 8 feet high, can be completely exposed to permit very large or tall objects to be placed inside. An air-motor-operated one-ton hoist with full rectilinear motion on the inside of the cell provides for internal heavy-weight lifting. A movable stainless steel tray is provided for most of the work to be performed on, which can be decontaminated, and removed if necessary. The entire cell is lined with stainless steel for easy decontamination. The sides are pierced by a number of holes, closed with removable steel plugs. A number of access holes of this type have been found necessary to take care of contingencies that may arise during operations, such as placing a long special tool through the side of the wall to perform some manipulations that cannot be done with the manipulators or hoists. Special services and lines can also be run into the cell through these access holes. The radioactive material rating of the cell is 10,000 curies of cobalt-60, which would be equivalent in radiation intensity to several thousand curies of one-year-old fission products. Irradiated aluminum cans containing up to 10,000 curies of cobalt-60 each have formerly been opened under water in the canal, but this operation can now be done faster and more easily inside this cell. Such operations can be done by sawing off the end of the slug with a remotely-controlled power hack-saw. The cobalt pieces can be removed dry, sorted, and the radioactivity measured, and the pieces placed in special containers for underground storage.

UNDERGROUND STORAGE

Storage of intensely radioactive materials such as cobalt-60 is done by placing the objects in long vertical tubes 8 feet deep in the ground. These stainless steel tubes are embedded in concrete or crushed rock and have a suitable lead or concrete plug at the top to shield off the radiation and seal against the weather (see Fig. 7). Material can be put down into the hole from a special shielded container which has a drawer at the bottom which can be pulled out and the especially-designed cup or container on the inside lowered down into the hole. This is a very inexpensive and satisfactory type of storage for highly-radioactive materials. Similar types of storage arrangement can be made by putting horizontal holes into a hill or an area covered with crushed rock or concrete. There are certain advantages to both the horizontal and vertical types of storage holes, depending upon the use to which they are to be put.

IODINE-131 PLANT DESIGN

Radiochemical handling in a relatively small plant can be illustrated by the recently-completed fission product iodine-131 processing plant at Oak Ridge National Laboratory. The layout for this plant is shown in Fig. 8. The main feature to be noted about the layout of this plant is that separate small cells are provided for each of the main functions, such as the dissolvers, the main distillation equipment, and the final purification glass distillation equipment. It is necessary for a radiochemical plant of this type to remain in daily operation for years with very little time out for repair of equipment. For this reason, duplicate dissolvers have been provided and all the cells can be quickly and easily decontaminated, having stainless steel linings and stainless steel equipment throughout; each cell is equipped with sprays to help in the decontamination. The two main chemical processing cells have removable concrete plugs through which access can be gained from the top; access to each of the dissolver cells can be had separately by removing stacked concrete blocks from a side opening. By having the important equipment in separate cubicles, it is possible to proceed with repair work on one particular piece of equipment without interfering with, or requiring decontamination of the rest of the equipment in the plant. This is very important for economical and continued operation on a strict schedule. The dissolvers and other equipment in this plant that require heating and cooling are controlled automatically, and all controls are passed to shielded transmitter cubicles; so there is no direct connection between the vessels and the outside operating area, thus preventing any possible back-up of radioactive solutions through the lines. The transmitters change the air pressure signals from liquid-level and specific-gravity tubes to electrical signals, which are transmitted to the instruments on the panel board. Inside the main distillation cell, the most important pieces of equipment, such as scrubbers, distillation columns, and tanks, are secured in place with special fastenings which can be quickly removed so that each piece of equipment can be replaced without delay if necessary. All equipment is attached to an off-gas line; and although the process gas is completely freed of iodine by the iodine process equipment itself, it is finally passed through a precipitron and filters before

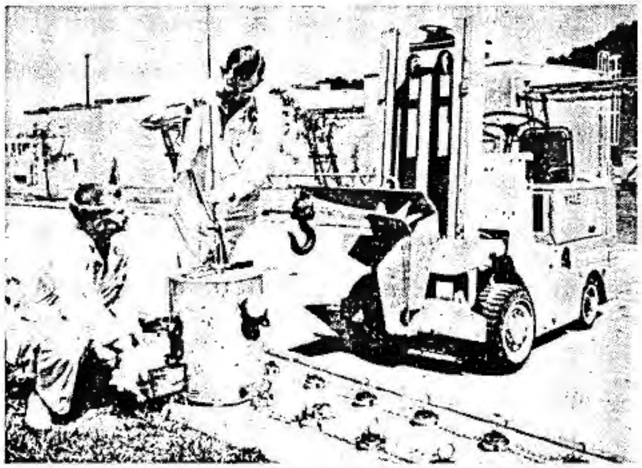


Figure 7. Underground storage operations

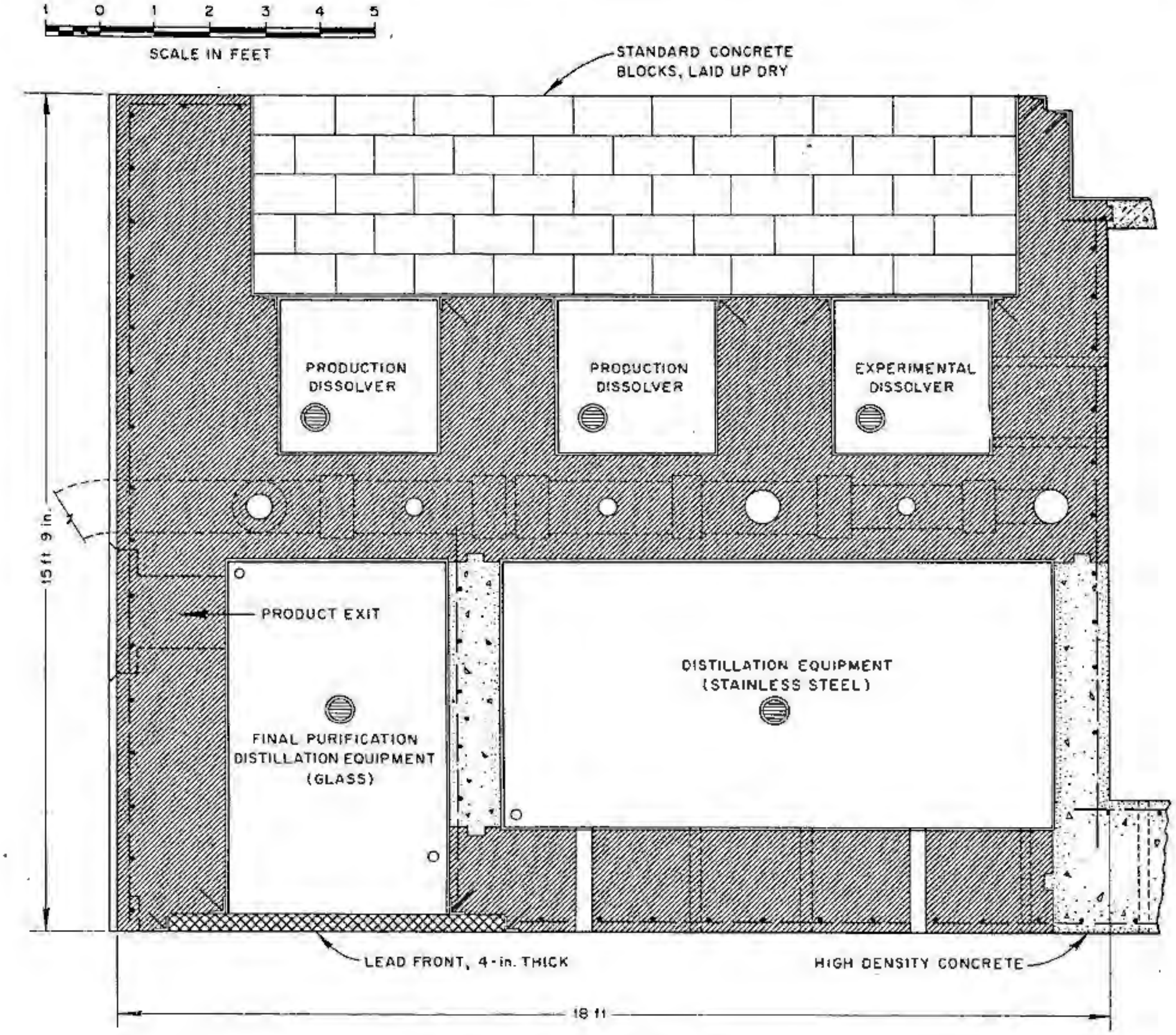


Figure 8. Iodine-131 plant layout

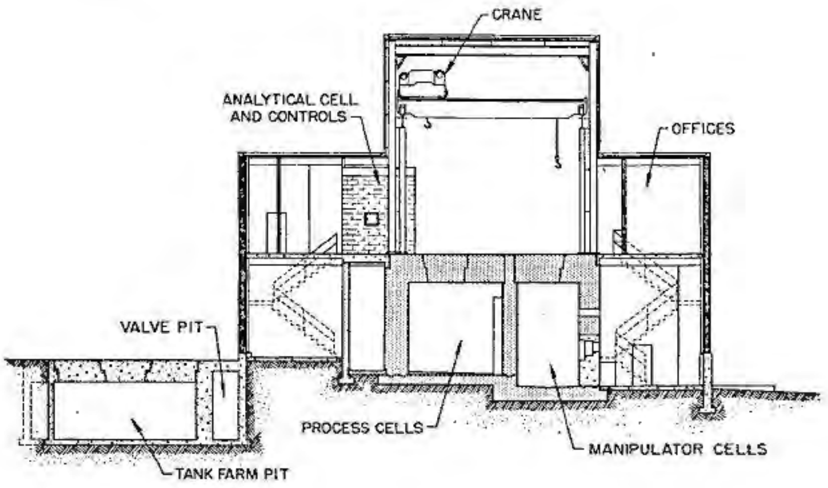
being discharged to the atmosphere. All air that is drawn from the cells is passed through fine filters which remove all radioactive particles down to 0.3 microns in diameter. Cells are kept under slight negative pressure at all times. All transfers of radioactive liquids in the plant are accomplished by means of steam jets or pressure-vacuum systems. Chemical reagents are added to the equipment in the cells, but no direct attachments are made to the vessels. A reagent line from the operating area on top of the cell block is passed into the cell, where the liquid to be put into a process vessel empties into a funnel and valved line which connects to the process vessel itself. In this manner, any back-up of radioactive material to the outside is prevented.

The uranium slugs for the iodine process are discharged from the graphite reactor into the canal and loaded under water into a lead cask with sliding drawer over a bottom opening. The cask is hoisted to a position above the slug chute and the drawer is slowly withdrawn. The drawer is designed with a slanting edge so that one slug at a time will drop into the slug chute. After charging the slugs, the valve on the slug chute is closed and the dissolving process is started.

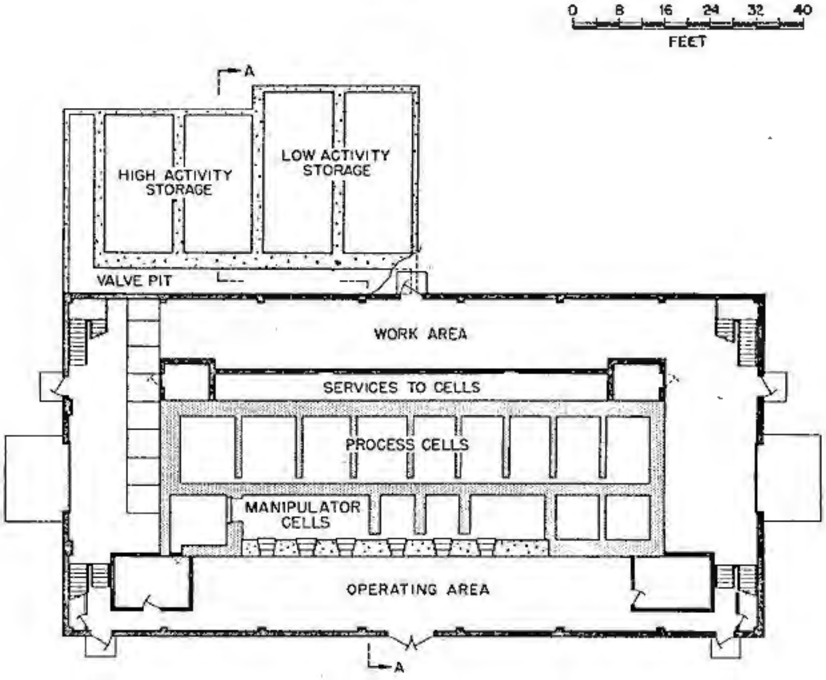
The final product is removed as a solution in a 500-ml bottle. A shielded carrier containing the bottle is shoved into the cell on a track, the iodine is loaded into the bottle, and the carrier closed and removed.

FISSION PRODUCT PLANT DESIGN

A plant for processing 200,000 curies of cesium-137 per year is in the final stages of design, and construction will be started in June, 1955. The layout of this plant is shown in Fig. 9. It is roughly divided into two parts: (*a*) the chemical processing section where the reactor waste is processed to separate the fission products into the various components, cesium,



SECTION A–A



FIRST FLOOR PLAN

Figure 9. Multicurie fission products pilot plant

strontium, rare earths, etc.; (*b*) a group of manipulator cells which fabricate and package kilocurie sources.

The equipment in the processing cells consists mainly of precipitators, crystallizers, and centrifugals, with the inner-connecting lines and steam jets to move liquids from one vessel to another. Associated with the plant is a tank farm underground in a sectionalized concrete pit containing tanks which are connected with various pieces of equipment in the plant by lines that pass through a covered concrete trench; transfers are made back and forth to the plant by means of steam jets. Some of the larger tanks that contain solutions with very high radioactivity concentrations are partly submerged in water for cooling purposes.

The other half of the plant is devoted to final purification of the separated fission product material, fabricating it into various sizes and shapes of radiation sources, testing, and packaging for shipment for use as medical and industrial radiation sources. For the most part, these are manipulator cells containing Type 8 through-the-wall manipulators, the operations being observed through high-density glass windows. The final loading cell is a large one containing two pairs of Type 8 manipulators and one large electrically-controlled General Mills manipulator for heavy handling and the loading of the final materials into the shipping cask.

The shipping casks are loaded through a locking system from a cell adjacent to the loading cell. The containers to be loaded are lowered by the 20-ton travelling crane into this cell and then passed over a small rail system through a hydraulically-operated shielding door into the loading cell. After the source is loaded into the shipping container, the operation is reversed and the loaded cask is thoroughly cleaned on the outside, checked for radioactivity and then placed on a truck for shipment. Sources that will be made in this final operation will involve such operations as pressing powders into pellets with hydraulic presses, pouring molten $Cs^{137}Cl$ into molds, compounding ceramic mixtures using $Sr^{90}O$, $Ce^{144}O_2$, or $Pm^{147}{}_2O_3$, fusing ceramics onto metal plates, encapsulation of both flat and round sources and sealing by welding, and electroplating Ru^{106} onto large copper plates. Other mechanical operations will be required to add various handling connections to the sources and place the finished sources in various kinds of shipping containers. All of these operations can be done by the master-slave and General Mills manipulators, using auxiliary tools and equipment for special jobs, such as welding.

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Transportation of Large Quantities of Radioactive Materials

By H. Blatz,* USA

The transportation of radioactive material involves a number of special considerations which must be taken because of certain unique properties of such materials. This problem first presented itself in the United States as early as 1936 at which time undeveloped photographic films being transported were damaged by the radiation from radium being shipped at the same time. As a result of this and other incidents, particularly concerning shipments of sensitive photographic emulsions, certain precautions were recommended in order to prevent injury to personnel or harm to property during such shipments.

In considering the necessary precautions in shipping radioactive material, there are several aspects to the problem which must be dealt with separately. First, the health of transportation workers as well as others concerned with loading, unloading and handling of packages must be protected. Then, photographic materials must be protected against the effects of radiations emitted from the packages. These first two considerations are both concerned with the nature and amount of radiation emitted from the packages themselves. Other aspects are concerned with radioactive contamination, which may result from shipments of such materials. First is the possibility of carrying radioactive contamination on the outside of packages to the extent that it may be transferred to other objects, thereby causing a possible health hazard to handlers or a technical hazard to photographic materials or scientific instruments. Another contamination consideration is that of possible leakage or the rupture of a package during transportation.

All of these problems have been recognized for some time, and in the United States, regulations have been enacted in order to require shippers to observe certain minimum standards of packaging and labeling. Such regulations have been promulgated by the Interstate Commerce Commission, the Civil Aeronautics Board, the Post Office Department, and the United States Coast Guard, each covering shipments within its particular jurisdiction.

Since the advent of the atomic energy age, the US Atomic Energy Commission has been particularly concerned with shipments of large quantities of various types of radioactive materials, much of it in bulk or loose form, such as granulated or powdered dry materials, sludges or liquids. Some of these contain radium from which radon gas is emitted. The handling of large shipments presents problems which are distinctly different than those encountered in the shipment of small quantities which may be readily packaged according to the accepted standards.

As in all studies of radiation protection and contamination control, the ultimate objectives must be balanced against costs. It is invariably possible to plan an activity, in this case a shipment of radioactive material, in such a way as to be completely safe, but the cost is often prohibitive and unjustified. Special arrangements have, therefore, been made for the shipment of large quantities of materials in a manner other than that prescribed for small quantities, but which are believed to involve no greater risk to health or property.

A special provision has been made in the regulations for carload shipments of bulk radioactive materials in which the car is treated in much the same manner as a package is considered in the shipment of smaller quantities of materials. In other words, the car must be loaded and braced in a prescribed manner and the radiation levels on the outside of the car must be within certain limits. The regulation currently used to cover such shipments reads "Radioactive materials such as ores, residues, etc., of low activity packed in strong tight containers are exempt from specification packaging and labeling requirements for shipment in carload lots by rail freight only, provided the gamma radiation or equivalent will not exceed 10 milliroentgens per hour at a distance of twelve feet from any surface of the car and that the gamma radiation or equivalent will not exceed 10 milliroentgens per hour at a distance of 5 feet from either end surface of the car. There must be no loose radioactive material in the car, and the shipment must be braced so as to prevent leakage or shift of lading under conditions normally incident to transportation. The car must be placarded by the shipper." The particular radiation levels stated in this regulation were arrived at after considering two possible difficulties that might arise.

The 10 milliroentgens per hour at the twelve-foot distance represents the maximum calculated risk of radiation exposure damage to photographic material in an adjacent railroad car. Such a calculated risk would, of course, involve the length of time during which such cars would remain in adjacent positions as, for example, in a freight yard.

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* US Atomic Energy Commission.

The lower value of 10 milliroentgens per hour at a distance of five feet from either end of a car was set in order to reduce the exposure of railroad workers engaged in coupling or uncoupling the cars. Additional protection is afforded in the event of any necessary repairs by a statement on a large placard affixed to the side of each carload of radioactive material (except, of course, where such shipments are escorted). These placards which are conspicuously marked with the conventional warning symbol, limit the length of time during which a worker may remain close to or underneath such a car, and a telephone number is given for notification in the event of required emergency repairs. This approach has been quite effective a number of times when it was necessary to make such a repair to the truck of a freight car during the course of its travel. The designated office was notified and a surveyor was dispatched to the location of the car in order to make measurements and direct the repairs in so far as limiting the exposure of the repairmen.

After use, the car must be suitably cleaned if it is then to be used in the shipment of other materials. Ordinarily, such cleaning is done by means of a central type vacuum cleaner which collects any spillage from the floor without ejecting it into the air. Occasionally, scrubbing and even sandblasting has been necessary to clean cars to the prescribed limits. On a few occasions, car floors have been replaced or repaired in order to remove badly contaminated floor boards. In order to avoid repeated cleaning of freight cars, the Atomic Energy Commission has obtained special cars which are used for no other purpose than the transportation of radioactive or contaminated materials.

When bulk shipments of radioactive materials were first made on a large scale, it was decided not to permit them to go by automobile truck on public highways. With favorable experience in the shipment of carload quantities and also based on a good experience record in the shipment of other dangerous material such as explosives by truck, it was decided to permit such shipments under close control. In many cases, railroad routes are such that truck shipment is much more economical. At present, all truckload shipments are made either by special permit or under escort by the Atomic Energy Commission. In spite of the fact that a considerably large volume of material has been shipped by truck in this manner, there have been no major accidents. It is, therefore, planned to consider permitting truckload shipments in much the same manner, as railroad carload shipments are not permitted under the general regulations.

One problem which presents itself in the shipment of large quantities of materials in a closed box car or truck is that much of the material handled contains quantities of radium which, in turn, emanate radon gas. If, after considerable time during which the car doors have been tightly closed, when it is necessary for workers to enter the car in order to unload the material, they are apt to be confronted with a concentration of radon gas which would be excessive for inhalation. For that reason, the special railroad cars assigned to this service have been equipped with large ventilating exhaust fans in order to provide adequate movement of air before the cars are entered. Fig. 1 shows the location of the ventilating fan grille in the end of the car. The usual procedure is to open the car doors which are located at the center of the car and exhaust the air through both ends by means of the fans. In this way, the accumulated radon can be cleared out of each car in a short period of time. In some instances where such mechanically ventilated cars have not been available, cattle cars have been substituted. The cattle cars have walls which are made of wooden slats rather than solid material and thereby provide adequate ventilation during transit. These cars, however, are not ideally suited because they do not protect the materials from the elements during bad weather.

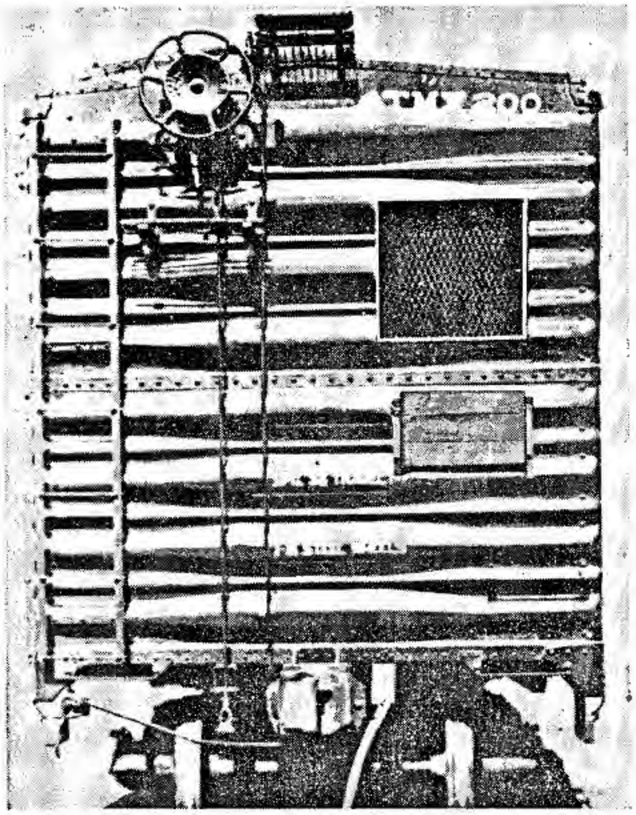


Figure 1

In the case of granulated or powdered dry material and sludges, the material is placed in steel drums, generally of either 30-gallon (113.6 liter) or 55-gallon (208.2 liter) capacity, and in case of moist materials, the covers are provided with gaskets in order to prevent leakage of material. In the case of some relatively low activity materials, they are shipped in fibre cylindrical containers with tight-fitting telescopic covers. Studies have been conducted to consider the feasibility of transporting these materials in hopper cars and tank cars, but the problems of loading and unloading proved so formidable that the advantages provided by such shipments did not compensate for the disadvantages.

The possibility of transporting much of the raw and semi-processed materials from plant to plant within the United States by water has been studied several times but since most inland water ways in the United States are also used for domestic water supply, it was believed that the hazard of contaminating drinking water in case of a major wreck was too great a risk to take to justify the saving in cost. The same consideration has been given to the possibility of a serious train wreck in a watershed area, but except in the case of liquid materials, it is believed that material could be salvaged before it could become dissipated into the ground and watershed. For this reason, special considerations have been given to the shipment of liquid radioactive materials and in most cases, they are shipped in double containers.

Recently, an additional problem in the transportation of large quantities of radioactive materials has been because of the larger and larger units of artificially produced radioactive isotopes which have been made available. The original regulations provided that no more than two curies of radioactive material could be shipped in one package without special arrangements being made to insure safety. It is quite common today to require the transportation of 5 to 10 kilocurie units of cobalt-60 which far exceed the quantities with which we have had experience in the past.

It would be well at this time to review the reasons for limiting the shipment of packaged radioactive material in general to 2 curies. In shipping a package by express or freight in the United States, the exact route of travel is not usually predetermined so that the shipper cannot easily keep track of the location of the package. Most transport workers are not familiar with either the hazards associated with radioactive materials nor the terminology used in their designation to such an extent that it would be possible for them to handle a package in the appropriate manner in the event of an accident. Although each package is provided with a warning sign, it is believed that an accident involving such material could result in the obliteration or the removal of such a warning sign and a value of two curies was selected, quite arbitrarily, as the limit beyond which the risk of harming an unsuspecting salvage worker would be too great to take. Recent modifications to the shipping regulations have permitted the increase of this limit to a much higher level but only under the condition that additional precautions be taken to protect salvage workers in the event of a wreck. For example, for larger amounts the container must be constructed in such a manner that the lead shielding is not likely to melt and run off in the event of a high temperature fire nor can the identification be obliterated by an ordinary fire. This is done by providing an outer container of heavy welded steel construction to withstand high temperatures and also provide engraved warning signs outside the container which will not be removed by fire. The mechanical strength of such a container has also been increased.

Occasionally, it is necessary to ship materials, which cannot be packaged, in a manner which will meet the accepted safety standards. In such cases, the shipments are accompanied by escorts. Whenever a properly designated escort accompanies such a shipment, it is not necessary to comply with any of the regulations. It has been the policy of the Atomic Energy Commission, however, not to use the presence of an escort as an excuse to evade safety regulations. Whenever an escort accompanies such a shipment, his presence is intended to compensate for any exemption from regulations. For example, if the package is such that it would be a hazard to the shipment of photographic emulsions in proximity, it is his duty to see that no such emulsions are loaded into a car near the radioactive material. If the hazard is related to the possibility of the train or truck being wrecked, the escort does not necessarily travel on the same train or truck but keeps in continual touch with it so that in the event of a wreck, he will be in a position to warn salvage workers and take charge of the radioactive material.

In recent years, the transportation of scrap materials and obsolete or worn out machinery contaminated with radioactive material has presented a problem. In addition, considerable quantities of radioactive waste materials have accumulated. It is frequently necessary to transport these materials great distances for the purpose of disposal or for reprocessing to produce new materials. Much of the material is in the form of scrap metal in various shapes that cannot easily be packaged. The concentrations of radioactive material in such scrap usually presents only the problem of contamination control; the direct radiation hazards are insignificant. The principal problem is to transport such materials in the most economical manner and in such a way as to reduce the possibility of contamination spread. Usually, such materials are shipped in gondola cars which are railroad cars in the shape of an open-top box into which the scrap materials are dumped. Usually, the only precaution necessary is to see that in loading and unloading, radioactive dust levels are not raised to such an extent that the air concentrations become an inhalation hazard, and also that the gondola cars be adequately cleaned after use in order to prevent the contamination of other materials which may later be shipped in the same cars.

The history of the transportation of large quantities of radioactive materials has been remarkably free of accidents during the many years of activity of the Atomic Energy Commission. A review of a few accidents which have occurred has served to assist us in formulating transportation policy.

One incident occurred during the war period, during which much obsolete railroad equipment was in use because of shortages. This incident involved a drum of uranium ore which plunged through the floor of an old wooden box during transit. The drum was a 55-gallon drum which, when filled, weighed some 2000 pounds. The drum hit the road bed and its contents were spread over a considerable area. The contents of the drum were shoveled up by railroad workers and placed into a new drum which was then forwarded to its destination. This incident, while very minor, is mentioned because of the fact that it was one of the first transportation accidents involving radioactive material and involved considerable apprehension on the part of some of the workers in cleaning it up. One worker made a claim that whenever he wore a watch, it operated improperly and thereby ascribed this to the fact that he had become radioactive. This accident merely required the education of the person involved since there was, in fact, no damage done.

Another incident involved the accidental dropping of a drum of uranium-bearing ore into a river in the course of its transfer from barge to a dock. The recovery of this drum necessitated the use of a deep sea diver and his equipment, but the drum was recovered intact.

Another accident involved the burning of a truckload of uranium metal on a public highway. The cause of the fire was never adequately explained but the uranium itself was completely burned and the contaminated remnants were removed without incident. This accident pointed out the importance of avoiding heavily populated areas or roads with dense vehicular traffic for the transportation of such pyrophoric radioactive material.

Another truck accident involved the overturning of a truck loaded with low-grade radioactive wastes on its way to a dump. Here again was an incident where virtually all of the material was quickly recovered and transported to its destination, but the accident caused apprehension on the part of the residents in the neighborhood because of an unfounded fear of anything radioactive.

Record of Proceedings of Session 19C

THURSDAY MORNING, 18 AUGUST 1955

Chairman: Mr. W. E. Libby (USA)
Vice-Chairman: Mrs. B. Karlik (Austria)
Scientific Secretaries: Messrs. A. Milojevic and A. Finkelstein

PROGRAMME

19C.1. General aspects

P/985	The use of radioisotopes in Australia	C. E. Eddy
P/801	Report on the use of radioisotopes in the Czechoslovakian Republic	C. Shimane
P/146	Radioisotopes in industrial research	S. E. Eaton

Discussion

19C.2. Production of radioactive isotopes

P/314	Large-scale production of radioisotopes	A. F. Rupp
P/461	The production of beta-ray sources for industrial and clinical uses	P. E. Carter and L. C. Myerscough
P/886	Separation of carrier-free isotopes by diffusion methods	K. Samsahl and K. Taugböl

Discussion

19C.3. Handling of radioactive isotopes

P/13	Current techniques of handling and distributing cobalt-60 radiation sources	A. B. Lillie
P/69	Technical arms incorporating a sense of feel for conducting experiments with radioactive materials	J. R. Burnett *et al.*

Discussion

The Chairman: We gather this morning to continue the discussion of radioactive isotopes and their uses. We have good reason to know that isotopes have many valuable uses, for we can cite them. In the United States, isotopes have proved to be of very material benefit to the economy. We estimate that our industry during the last year has saved over \$100-million in processing costs, and I firmly believe that within five years this figure will be \$1000-million or more—and this, despite the fact that many obvious uses of isotopes have not been made.

I cite in particular the paucity of uses for the isotopes of carbon and hydrogen, elements so important in the chemical industry and in biology and medicine that it is difficult to understand why their radioactive forms have not been more widely used. It is certain that the development of these obvious uses will greatly expand the peace-time benefits derived in this way from the atom.

We have spoken in this Conference at length about atomic power and the promise it holds for mankind. We all believe, I think, that atomic power will one day be realized on a practical scale, and it will indeed constitute a great and substantial benefit to mankind. But, in the case of radioactive isotopes, there is no argument: we do know that these are a great and substantial benefit already. We must therefore constantly bear in mind that, even if power prove impossible of attainment, the atom has already paid in the uses of radioactive isotopes. I myself would not be surprised if isotopes were to lead atomic power for many years in the benefits derived by mankind. It is difficult, of course, to compare the two, for they are so different. But, with all of our preoccupations with the problem of atomic power, let us not forget that isotopes are here.

More important, let us attack some of the problems which isotopes present. We can see that in the case of atomic power there are great and difficult problems. There are problems with isotopes, none of which, I think, are either great or difficult, but which must be solved. I think particularly of the possible applications of isotopes involving small specific radioactivities. I think particularly of the utilization of the soft beta emitters.

Certainly there is no lack of ideas for these applications. One knows that there are many ways in which the isotopes of carbon and hydrogen and of calcium and chlorine, the long-lived isotopes of the ordinary elements, could be used widely in everyday life. Why are they not so used? At the moment, the principal reason is that the instrumentation is not available except in the experimental laboratories. The companies manufacturing instruments have not produced cheap and efficient and reliable devices. This problem must be attacked.

At the moment it is the principal barrier, in my opinion, to the widespread use of these important isotopes.

I would simply emphasize once again that isotopes are a major benefit, that they are well ahead of atomic power and will continue to rival it. We must take care of the problems of producing instruments for the measurement of isotopes and we must study the problems of regulation of isotopes so that we do not over-regulate them but nevertheless protect the public health. With these points in mind, there is no doubt that we will continue to see this exceedingly rich benefit of the atom grow and flourish.

Mr. C. F. EDDY (Australia) presented paper P/985.

Mr. SHIMANE (Czechoslovakia) presented paper P/801 as follows: Mr. Chairman and gentlemen, in my paper I should like briefly to refer to the development of the use of radioisotopes in Czechoslovakia in the last few years. The radioisotopes which we have been importing into Czechoslovakia for a number of years now were originally used, as everywhere else, in work of an exploratory nature directed towards finding methods and measurement techniques. An active part in work on radioisotopes was taken by the physics laboratories of the higher educational institutions and of the Academy of Sciences, as well as by a number of industrial research establishments. From the very beginning, a considerable interest in the use of radioisotopes appeared in approximately fifteen places, in the most diverse scientific fields. The simplest use of radioisotopes, from the point of view of equipment, is in the therapeutic field. This applies chiefly to the use of radiophosphorus, which has been used in our principal hospitals with complete success in approximately 50 cases of treatment of polycythaemia. Treatment by radiophosphorus is considered to be better and longer-lasting in its action than X-ray treatment. Radiophosphorus has also been used in a whole series of cases in the treatment of leukaemia, but in this instance radiophosphorus treatment has no particular advantage over X-ray therapy.

In addition to phosphorus, radioiodine has also been used for therapeutic purposes. There are several hundred cases of hyperthyroidism, under treatment with radioiodine. Treatment has been completed in the cases of approximately 100 patients. Both the consecutive-dose method and the single-dose method are in use. As a result of the development of a complete diagnostic system for the prescription of radioiodine, the proportion of myxoedema cases is approximately two or three per cent. This treatment method has also been used in approximately 15 cases of cancer of the thyroid gland. These were all cases of progressive and inoperable cancer and the life of the patients has so far been prolonged by two years. Cases for treatment with radioiodine are selected on the basis of their accumulation graphs and by means of the method known as the differential accumulation graph method, which is supplemented by gamma-radiography.

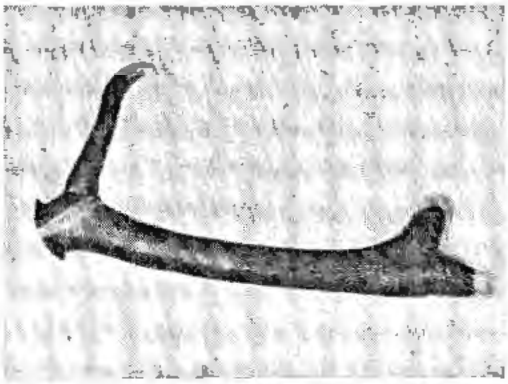
The use of radioisotopes in diagnostic work began to develop at the same time as their use in therapy, but in practice only the diagnosis of thyroid disorders by radioiodine is widespread. The number of diagnoses of thyroid disorders at present exceeds 2000.

The radioisotopes of iodine, sulphur, phosphorus and sodium have been used in studying the dynamics of the circulation of substances in the living organism in a state of shock, from the point of view of their practical use in surgery. This research has produced valuable data on the possibilities of introducing medicaments into the organism of a patient suffering from shock. A whole series of radioisotopes has been used in oncology in the treatment of cancerous tumours. A method of locating brain tumours by radioiodine is being developed. A method using radioiron has been developed to determine the resorption of iron from the digestive organs in patients suffering from various forms of anaemia, and another method uses radioiron to determine the iron in the blood serum or to check the rate of flow of the blood circulation. Nucleic acid metabolism is being systematically studied by means of radiophosphorus.

Let us now turn to the use of radioisotopes outside the field of medicine. In plant physiology, studies are being made of the penetration of mineral salts through the surface of the leaves of plants. These experiments are being carried out in conjunction with experiments on non-root nutrition in plants.

Some interesting research has been carried out in connexion with the study of phosphorus metabolism in deer. Radiophosphorus was given to male deer during their early life and particularly during the antler-forming period. Studies were made of the distribution of phosphorus in various tissues of the killed animal and radiograms were made of the antlers. The radiograms show that the beginning of new tip formation is associated with changes in phosphorus metabolism in the antler. Slide 1 shows that a wedge is forming inside the main antler; this wedge has a high phosphorus content and extends in the direction of the branching part of the antler.

In the field of water conservation, a device based on the principle of gamma-ray absorption to measure the water equivalent of snow will in all probability prove valuable. It is believed that by setting up these devices in the known network of channels it will be possible to predict the strength of the



Slide 1

spring floods and determine the water-level in reservoirs in advance so as to ensure they will hold all excesses of water during the thaw and that the water-level will not fall unduly.

I shall now deal with the use of radioisotopes for industrial inspection purposes. The most widely used in this field are the radioisotopes of cobalt and iridium. Radioiridium is used to check welds of up to 40 millimetres in thickness, to examine boiler-tube welds and to check thin-walled castings, and radiocobalt is used for the same purposes where greater thicknesses are involved. The use of these radioisotopes is of great importance to our industry. With the ordinary X-ray apparatus used in our industry, it is reasonably possible to inspect steel of a thickness up to 80 millimetres. It is well known, however, that the use of such apparatus in assembled plants is extremely inconvenient. By using radioisotopes, on the one hand the thickness of steel which can be inspected is increased to 150 millimetres, and on the other the scope of gamma-ray inspection can be extended and it can be used in places where previously this was impossible owing to the lack of portable apparatus or the physical impossibility of using such apparatus.

In Czechoslovakia all high-pressure tubing welds are checked as a matter of routine by means of radioiridium and radiocobalt and the tubing is also checked in the process of fitting. When any power station is being built in Czechoslovakia all the tube welds are examined by radiation and checked. The methods used for this procedure are described in the paper.

The continuous inspection of tubing by radiation has been found to result in a considerable improvement in the quality of welding work and, consequently, in greater safety and fewer defects. The inspection of seams is of great value in training welders, since they have an opportunity to appraise the quality of their own work objectively and to improve it.

In addition to this widespread use of cobalt and iridium, other industrial applications of isotopes are beginning to develop. For instance, research is proceeding into carbon diffusion in steel during consolidation; radionitrogen, formed in the experimental specimen itself by proton irradiation, is used in these experiments to determine the carbon content in individual layers.

Work on short-lived isotopes is developing slowly in Czechoslovakia because their short half-life does not give us an opportunity of experimenting for very long with isotopes from the same batch.

This situation will improve considerably as soon as it becomes possible to produce isotopes in Czechoslovakia by means of the reactor which we are to receive from the Soviet Union. Widespread applications of radioisotopes may be expected when this reactor goes into operation.

Mr. S. E. Eaton (USA) presented paper P/146 as follows: As my confreres have pointed out, radioactive isotopes are proving extremely potent as research tools for accelerating industrial progress. Developed as by-products of a military programme, they are now creating new knowledge and understanding of nature and of industrial processes and products which will have positive and permanent value to mankind. The purpose of this paper is to discuss the unique and powerful features of radioisotopes which have been found particularly useful for industrial research, and to set forth some typical problems which they are solving in the United States of America.

The four features of radioisotopes on which most American uses are based are as follows: Firstly, they are easy to detect. Simply bringing a Geiger counter near them is sufficient. Secondly, they provide a small, inexpensive, portable source of radiation. Thirdly, they are specific and can be traced in the presence of other chemically identical atoms. Thus specific batches of tagged or radioactive atoms from one source can be differentiated in the presence of other chemically identical atoms from a different source. Lastly, they can be measured in minute quantities with high sensitivity. Someone has calculated, for the benefit of those who like to consume their alcohol undiluted, that it is possible to detect as little as one drop of vermouth in three tank car loads of whisky.

How are these features being applied to problems of industrial research? The fact that radioisotopes are easy to measure is simplifying identification of the constituents of a sample by activation analysis. By irradiation in a reactor or accelerator the sample is made radioactive in a manner characteristic of its constituents. A knowledge of these characteristics then allows interpretation of the sample's radioactivity in terms of its composition. The method is proving of particular value in the States where a non-destructive test for traces of metallic impurities is desired in very pure material.

Ease of measurement also is making it simpler to determine optimum mixing time for such materials as paint, ink, plastic products, powders for powder metallurgy, and the like. One of the components is tagged with radioactivity, either by irradiation in a reactor, simple addition of a physical tracer, or synthesis of an isotope into the component. After predetermined periods of mixing with the tagged material, samples from different parts of the batch can be compared for radioactivity and the batch mixed until activity is uniform or constant. Once the optimum mixing time has been determined, the tagged batches can be disposed of in some safe manner, and it should not be necessary to add isotopes to the process again until some variable is changed.

There have been a number of suggestions for the routine addition of isotopes directly to a manufacturing process for the purpose of controlling the process, but to date these have not been put to practice in America. The reason for this involves both technical and public health considerations.

The next feature of radioisotopes, however, does allow them to be used for industrial control purposes, but in such a way that they are not added directly to the process or product.

An examination of the changes produced in a beam of radiation when it penetrates an unknown material can provide useful information about the material, for example, its thickness. There has been developed, for example, a device employing radiation from strontium-90 to measure the hydrogen content of a pure hydrocarbon. When beta particles from radioactive strontium are passed through a hydrocarbon sample such as gasoline or oil, the attenuation or loss of the particles depends on the number of electrons they encounter in the sample. Since hydrogen has the highest number of electrons per unit weight of any element (in fact, it absorbs just about twice as many betas per unit weight as does carbon), its absorption of the beta particles is greatest. Measurement of betas absorbed by the hydrocarbon sample thus becomes an indication of its hydrogen content.

The tool is being used not only for plant and product control but also for research on such problems as altering the percentage of hydrogen in petroleum fuels to improve their performance.

Examples of the third feature of radiosotopes, that is their great sensitivity, are numerous. Wear measurements have been made on automobile piston rings, gears, floor wax, paint, and cutting tools usually with increased accuracy and in a shorter time because of the high sensitivity of the test.

Another example of the high sensitivity of radioactive materials is an exploratory study my company made for a silverware manufacturing company. The purpose was to learn more about the mechanism by which a thimble full of ammonium thiosulfate added to 100 gallons of electroplating solution makes the resulting silverplate much shinier. Not much is known about the way electroplating brighteners act, but it has been postulated that a minute amount of brightening agent like ammonium thiosulfate might deposit from the plating bath with the silver and interfere with the formation of large, course crystals which produce a dull silver surface. If this were so, sulfur from the ammonium thiosulfate should be present in the silver, but no one has ever been able to find any sulfur there, so the theory could never be substantiated, and further progress toward a more active brightener was slowed down.

Because radioactivity provides an extremely sensitive means of detecting sulphur, we synthesized some of it into ammonium thiosulfate, added it to the plating bath, and analyzed the final silverplated surface for radioactivity. Definite activity was found throughout the plate, and the calibration of radioactivity in terms of sulfur indicated one sulfur atom to every 100,000 silver atoms.

The surprising part of this study was not so much that sulfur was present in the plated silver, but that we found it also present as a surface layer on the other electrode, the silver anode, whose surface was continually dissolving in the plating solution. This discovery led to the finding that sulfur from ammonium thiosulfate deposits on silver by a chemical action without the need for any electric current at all. Such a result completely upsets the previous theory of how this type of brightener acts.

So much for examples of sensitivity of the isotope technique.

A number of examples can be mentioned illustrating the specificity of radioisotopes which allows them to be traced and identified even in the presence of other chemically identical atoms from other sources.

Cattle-feed manufacturers have tagged different chemical forms of vital minerals in order to select the forms having proper solubility for use in salt cake for cattle. Radioactivity measurements on the animal's vitals and excreta could show the amount utilized by the animal.

A large oil company has examined the mechanism by which carbonaceous coke deposits on the solid catalyst beads during catalytic cracking of petroleum. He synthesized various petroleum molecular fractions from radioactive carbon 14 in such a way that different parts of the molecule were tagged. He then observed the radioactivity of the coke which built up on the catalyst beads as the different tagged molecular species were cracked. Within experimental error each sample of coke had the same specific activity as the labelled hydrocarbon from which it was formed. This indicated that all carbon atoms of a given hydrocarbon molecule are equally involved in coke formation. The possibility that part of the molecule forms coke preferentially seems essentially ruled out by the study.

Radioactive tracers sometimes make it possible to do research on a full scale plant. An automobile manufacturer measured the fraction of fine ore dust charged to a blast furnace which is blown out by the high velocity air going through the furnace. By irradiating part of the fine ore and mixing it uniformly with the rest of the furnace charge, then noting the radioactivity of the pig iron, slag, and expelled dust, he found that 60 per cent of the fine tagged ore dust remained in the furnace as pig iron: though surprisingly high, this percentage was not considered high enough to warrant the use of finely divided iron ore powder, which was available from low-grade ores.

The manufacturer concluded that further research is necessary into the way iron particles can adhere together or agglomerate, and that such research might allow finely divided, low-grade ore concentrates to be used in a blast furnace.

My company is currently making another full scale plant study for a client to determine the flow pattern in a continuous glass-making process. Radioactive sodium phosphorus and barium are being added intermittently to specific small portions of the glass feed materials. Radioactivity of the exit glass and of samples taken at certain key spots in the tank are noted as a function of time after the tagged batch is introduced. This work is yielding information on the holdup time of the glass in the tank as well as on the degree of mixing and channelling of the feed materials in flowing through the tank. Final results will be used as a basis for designing an improved glass-melting tank.

Determining the mechanisms of chemical reactions is also possible with radioisotopes.

In the field of metallic corrosion, we have obtained evidence with radioisotopes that helped settle a law suit for one of our clients. The problem, in part, was to determine the mechanism by which carbon disulfide prevents the corrosion of brass fire extinguishers filled with stabilized carbon tetrachloride. By tagging the sulfur of the carbon disulfide molecule, it was possible to follow its reactions and to show that it formed a very thin but tenacious film on the inside surface of the brass fire extinguisher which prevented corrosion. The film could be measured easily by its radioactivity even though it was only about 50 to 80 molecules thick—well below normal limits of detection.

The varied fields from which these examples have been selected illustrate the great versatility of radioisotopes for industrial research.

Though radioisotope techniques are applicable to a wide variety of problems they are not without their limitations. For example, a suitable isotope is sometimes not available or must be used in a quantity too large for safe operation. The cost of using radioisotopes for industrial research is frequently less than for other research techniques, but even with tracers the total research cost may sometimes seem high to those who have thought of the radioisotope technique as a delightfully simple panacea. As in most research, the major direct cost is for the time of the research worker—to plan, carry out, interpret the work and report. We have found that a good research man, familiar with solving industrial research problems, can quite easily become proficient in using radiochemical techniques so that, at least for modest scale operations, a full-time radioactivity specialist is usually not required.

In conclusion, I would like to report that the use of radioisotopes for industrial research in America is increasing steadily. Future growth depends not on cost, but on the industrial researcher himself. This man is in a key position to understand the capabilities of radioisotope techniques and also to recognize the particular problems that they can solve. He can bring the two together.

Radioisotopes are ready to reward the ingenious researcher with new knowledge for the benefit of industry and mankind.

DISCUSSION OF PAPERS P/985, P/801, P/146

Mr. Aebersold (USA): Mr. Eddy's paper has demonstrated very well that a nation a very large distance from a reactor laboratory can make very good use of radioisotopes. Would he comment more on the extent to which a reactor in his own country will meet the needs for radioisotopes, and whether he will still need to import isotopes; and also the extent to which the reactor will assist the extension of isotope uses.

Mr. Eddy (Australia): In reply to Mr. Aebersold, I think there is no doubt that a reactor locally in the country would extend the use of isotopes particularly, I think, potassium, sodium and three or four of those other elements with short lives of under twenty-four hours, for which at present we have a good many requests and no source of supply. Also there would be a question of chemical analysis by activation and possibly also radiation units. Just whether sources of isotopes which are now obtainable in very large quantities, such as cesium, iodine, cobalt-60 and so forth, would be made preferentially in a small country or imported from another, I could not say, but the mere presence of a reactor in the country and the opportunities for irradiating materials and obtaining special isotopes would, I am quite sure, increase the demand for isotopes, and just where that would lead I could not say.

Mrs. P. Cambel (Turkey): I would like to ask Mr. Eddy what are the legal or other requirements in regard to qualifications of the persons who are allowed to use radioisotopes.

Mr. Eddy (Australia): In reply to Mrs. Cambel, in the medical field we have had in Australia a restriction of the use of radium and radon since 1929 to people who had been trained in their use. That was for the treatment of malignancies in which the dose that is required to destroy the malignant cell is so close to the dose required to destroy healthy tissue that very special training was required. In the medical field now, where an isotope is used for therapeutic purposes of the malignant type, approval would only be given to a trained radiotherapist. In the case of more medical treatment, such as using P^{32}

for polycythemia and iodine-131 for thyrotoxicosis, approval would be given to experienced physicians, not necessarily radiotherapists, providing they had full ancillary equipment in conducting the diagnostic tests and full monitoring equipment. In industry it would be necessary for some short training to be given to the people using the isotopes and satisfactory instrumentation would also have to be provided.

Mrs. CAMBEL (Turkey): May I address another question to Mr. Eddy, Mr. Chairman? I would like to know what exactly the certificate would be: would they have to have certificates, or just a statement about their training? Furthermore, who would control the full equipment in your country?

Mr. EDDY (Australia): The Radioisotopes Committee of the Medical Research Council investigates a statement from the intending user in medical cases and is prepared to accept qualifications of a postgraduate type or training in an approved hospital: and if the applicant has not got that training they would suggest he go to a hospital to get it. The object of this committee is not so much to restrict as to provide an ever-increasing circle of people who could be trained.

Mr. S. M. NAUDE (Union of South Africa): South Africa makes use of radioisotopes in all the known fields of application. I will not attempt to give an account of this work, for it is very similar to that being carried out in other countries. I would, however, like to give a brief description of the method of transport which was first applied by South Africa. We obtained most of our radioisotopes from Harwell, and in 1946 to 1948 they were carried by aeroplane in the ordinary lead-lined boxes. In 1948, the South African Council for Scientific and Industrial Research suggested that a great saving could be made by carrying the isotopes in the wing tips of aeroplanes in light aluminium cans instead of making use of the lead-lined cases in the fuselage of the plane. We make use of the inverse square law for the reduction of the intensity of the radiation. In one typical case the cost of transport was reduced from £100 to 27s. per consignment.

Mr. A. F. RUPP (USA) presented paper P/314.

Mr. P. E. CARTER (UK) presented paper P/461.

Mr. E. SAELAND (Norway) presented paper P/886.

DISCUSSION OF PAPERS P/314, P/461 and P/886

Mr. FISCHER (France): Could Mr. Rupp tell me what qualifications are required from commercial firms which handle or redistribute radioactive materials prepared at Oak Ridge, and whether these operations are a commercial enterprise or are subsidized by the United States Atomic Energy Commission?

Mr. RUPP (USA): That question can probably better be answered by Mr. Aebersold.

Mr. AEBERSOLD (USA): The matters put to business concerns relate only to health and safety. We ask for the use to be made, but do not enquire into the feasibility of the use or into the economics. We are concerned with the health and safety aspects only. The sale of isotopes is not at a subsidy for industrial use, and as a matter of fact all the prices are at the full cost of production. If there is any subsidy it is done by research grants rather than in the sale of the isotopes.

The CHAIRMAN: I noted in Mr. Rupp's paper the great disparity between the rate of rise in domestic consumption of isotopes in our country and in foreign countries. In this connexion, I think you would be interested to know that it is the intention to try and simplify the regulations for isotopes sent abroad, and also in some cases to reduce the price. Perhaps, therefore, this foreign shipment curve will rise, at least we hope so.

Mr. RUPP (USA) to Mr. Carter: How much of the carrier-free strontium-90 as the oxide can be incorporated in the powdered silver matrix, per unit volume?

Mr. CARTER (UK): This depends very much on the exact conditions, particularly as regards the extension of the composite billet which has to be made, and also on the exact physical properties of the inclusion compared with the physical properties of the inactive metal. For what it is worth, I can say that in our particular conditions, making silver foil containing strontium carbonate, we can include up to 15 per cent of active material by weight.

Mr. RUPP (USA) to Mr. Saeland: In processing your micro-pulverized sulphur to extract P^{32}, is any difficulty experienced with colloidal sulphur during the filtration?

Mr. SAELAND (Norway): Yes, under some conditions. The colloidal sulphur which might occur in the filtrate can, however, easily be destructed. Under normal conditions, however, no such operation is necessary, and the final solution is free from sulphur.

The CHAIRMAN: Returning to the paper P/146 presented by Mr. Eaton, we have a question by Mr. Bogdanoff of the Soviet Union.

Mr. BOGDANOFF (USSR): If data are available here at the present time I should be interested to learn about the latest achievements in the United States in the use of labelled atoms for the study of ore concentration processes.

Mr. EATON (USA): My paper was directed particularly towards the industrial research uses of tracers. I think this question might be more appropriately put to Mr. Aebersold.

Mr. AEBERSOLD (USA): In the field of metallurgy the tracers are used extensively for metallurgical work, but as far as using isotopes in the field for discovering mineral deposits is concerned, I am not aware of any work along those lines. There has been work on activation analysis of minerals found in the field—of course one can use radioactivity measurements for determining the uranium and thorium content of various minerals—but other than activation analysis I am not aware of the analysis of minerals by alternative methods. Perhaps Mr. Libby, who has a great deal of interest in geochemistry, could comment on this.

The CHAIRMAN: I think perhaps that Mr. Eaton would be able to answer it.

Mr. EATON (USA): I do know of some plans, although the work has not been undertaken yet, for the study of the behaviour of minerals during processing which would involve locating the source of the disintegration of the minerals during the processing of them once they are removed from the mine. This is more in the nature of process research, however, and I am not sure that it is exactly within the scope of the question as it was put. Of course, radioisotopes have also been used a great deal in enriching processes, and I think there have been papers already presented having to do with the study of flotation agents. The rate of absorption of xanthates on sulphide minerals has been studied at MIT, for example.

Mr. ROLLIER (Italy) to Mr. Saeland: Could you tell us if the yield of extraction of iodine-131 by diffusion is higher than the yield of solvent extraction by hexane mentioned two sessions ago?

Mr. SAELAND (Norway): I do not know exactly how great this yield you refer to is—could you tell me that perhaps?

Mr. ROLLIER (Italy): I do not remember the yield actually.

Mr. SAELAND (Norway): I should think the overall yield here for this method described is about 90 per cent.

Mr. A. B. LILLIE (Canada) presented paper P/13.

Mr. R. C. GOERTZ (USA) presented paper P/69.

DISCUSSION OF PAPERS P/13, P/69

Mr. MULLER (Switzerland): I should like to ask Mr. Lillie's opinion about an internationally organized standardization of the size and shape of cobalt-60 pellets, in order to facilitate the replacement of the sources or their second-hand exchange. I think that Mr. Brucer of Oak Ridge has made a suggestion and did some work on that line.

Mr. LILLIE (Canada): I do not think that standardization of the size of the pellets is necessary. However, from the point of view of the containers, the greater standardization there is, the greater flexibility there is in interchanging sources of machines.

Mr. GOERTZ (USA): I should like to ask Mr. Lillie the following question. If he were to construct another isotope-handling facility, what changes would he include? For instance, would he design the facility to handle a wider range of sizes?

Mr. LILLIE (Canada): I believe that a more flexible facility is planned. For the particular purpose at hand, however, I do not believe that there are any major changes in the design. The facility can handle a rather wide range of sizes, provided they are roughly of this particular shape.

Mr. MILES (Australia) to Mr. Goertz: Does the servo-manipulator require substantially more maintenance than the mechanical system, and is this more difficult for contaminated equipment?

Mr. GOERTZ (USA): The servo-manipulator has not been placed into use as yet; it was just completed a few months ago. I think that the answer to the question is: Yes, it will require more maintenance and will be more costly; certainly, it cost more to construct.

The CHAIRMAN: I think that we have seen this morning how true it is that we are already benefiting from radioactive isotopes in a way which makes it clear that this use of the atom is certainly very useful. Even though we may have difficulty in realizing atomic power, this use alone will justify our efforts in the development of the nuclear phenomenon. I wish that the difficulties which we have encountered in developing the uses of isotopes could be more rapidly surmounted. Do delegates realize that nothing that we have heard about this morning was essentially impossible ten years ago? The difficulties have been connected with the slow development of ideas and with financing. The difficulties of irradiation have not been considerable. We can greatly accelerate the use of isotopes. Let us do so. It is a very inexpensive use of the atom, compared to atomic power. We are all for atomic power, but we are also for this most important use.

Session 20C

DOSIMETRY

LIST OF PAPERS

Low-Level Counting and the Future of Isotopic Tracers

By W. H. Johnston,* USA

The ready availability of radioisotopes made possible by the nuclear reactor has recently found many applications in research, development, and control. In broad terms these applications consist of the use of radioisotopes as fixed sources, for activation analyses, and as tracers. The tank-level and thickness gauges which utilize changes in the absorption and scattering of radiations and the pasteurization of foods by the ionization caused by radiation are familiar examples of the use of fixed sources. In activation analysis the specificity of decay-rate and type and spectrum of radiation together with the sensitivity of detection make possible the quantitative measurement of many trace elements by inducing activity with neutron bombardment.

Perhaps the most versatile application, however, is the use of the radioisotope as a tracer. The "bird band" tagging of the interphase in a pipeline, the use of gross specific activity as a measure of the degree of physical mixing, or, conversely, as a volume measurement, and the study of reaction mechanisms by isotopic molecular tagging illustrate the great variety of techniques. In general the use of tracers depends upon the chemical and physical near-identity of the radioisotope to the natural element and upon the great sensitivity by which radioisotopes can be detected. It is in the latter category that recent advances in instrumentation have opened up new vistas for the widespread use of radioisotopic tracers.

Although the variations in half-life, sample mount, type of radiation, and detector make it difficult to select a single example, an ordinary radio-chemical measurement of an activity of 50 counts per minute which is determined with a background of perhaps another 50 counts per minute usually corresponds to an actual disintegration rate from 2000 to 200,000 per minute. Such a measurement is usually made with the relatively small sample of the common commercial Geiger counter. It is clear that at least an order of magnitude increase in primary detection efficiency is possible in principle. In addition another order of magnitude should be gained from the use of larger samples. Furthermore by reducing the background rate by ten-fold another factor of two is gained for equivalent statistical reliability. The present paper is an analysis of how these goals of maxima in detection efficiency and signal to noise ratio can be approached with modern low-level instrumentation, and how these instruments can be profitably used in many tracer applications. Furthermore the potential future for large scale use of tracers is analyzed with attention to the question of safety. The low-level instruments are considered in two classes, the gas-tube counters and the scintillation detectors.

LOW-LEVEL GAS-TUBE COUNTING

The first major application of low-level counting was the work of W. F. Libby and co-workers.[1] Their discovery of carbon-14 in nature and their establishment of the important method of radiocarbon dating required a low-level method for measuring less than 100 disintegrations per minute of this weak beta emitter.

The necessary detection sensitivity was obtained by the use of the screen-wall counter of Libby.[1, 2, 3] The problem of the signal to noise ratio resulting from the high background activity of this relatively large counter was solved by the use of massive shielding for gamma radiation plus electronic shielding for the mesons of the cosmic rays. The mesons were detected in an "umbrella" or perhaps, a "raincoat," of Geiger counters surrounding the sample counter. These counts were electronically subtracted by placing them in anti-coincidence with the counts of the sample counter. In this arrangement the Geiger counters of the envelope can be operated in parallel from a single high voltage supply. A number of satisfactory circuits are available for accomplishing the anti-coincidence operation.[4, 5] Our apparatus is shown in Figs. 1 and 2.

The screen-wall counter with massive shielding and anti-coincidence electronic shielding has proven to be a reliable method. Subsequent developments in gas-tube low-level counters have adopted these techniques of background reduction. The massive shielding usually consists of eight inches of iron or steel. Almost as good results can be obtained with four inches of iron plus four inches of lead. A shield entirely of lead, however, is unsatisfactory because of the contamination by natural radioactivities. Kulp and others have shown that an additional inch of mercury between the iron and counter will further improve the background by removing some gamma radiation probably from contaminants in the shield.[8] This radiation would escape the electronic cancellation. The effects of these massive shields and the anti-coincidence shielding are shown in Table I.

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TABLE I. The Effects of Shielding and Anti-Coincidence in the Reduction of the Background Rate of Gas-Tube Counters*

Shielding	*Background rate in counts per minute*	*Difference*	*Remarks on difference*
None	450		
5 cm lead	142	308	Cosmic radiation and laboratory contamination
20 cm (8 in.) iron	110	32	Contamination of the lead
20 cm (8 in.) iron plus anti-coincidence	5	105	Mesons
20 cm (8 in.) iron plus anti-coincidence plus 1 in. mercury...	2	3	Contamination in iron

* After Anderson,[6,7] and Kulp.[8]

Screen-Wall Counter

The first type of gas-tube counter to be discussed is the screen-wall counter.[1, 3, 7]

The sample in solid form is mounted on the inside surface of a cylinder which surrounds a Geiger tube. The "wall" or cathode of the Geiger tube is an open grid of wires thus eliminating the usual absorption by a "window." The sensitive volume is extended to the surface of the sample by a suitable electric field between the sample and the screen-wall or grid-cathode of the Geiger tube. The effective geometry is essentially 50 per cent and the detection of beta rays leaving the sample is usually 400 cm². With elemental carbon 8 grams sample in which the thickness is equal to the maximum range of the beta radiation is usually used. The self-absorption losses are therefore high. On the other hand the large size of the sample cylinder provides an unusually large sample area. This area is usually 400 cm². With elemental carbon 8 grams are accommodated giving an absolute efficiency of 5.4 per cent for carbon-14.[1]

Internal Gas Counter

The second type of gas-tube counter which is important for low-level counting is the internal gas counter. In its simplest form this counter consists of a Geiger tube with provision for placing the sample in gaseous form inside the counter as part of the counting gas. The discovery and measurement of the distribution of tritium in nature was done with this instrument.[9, 10, 11] The simplicity of this counter and the associated electronics, its ease of operation and high efficiency approaching 100 per cent make this an attractive method provided that the sample can be readily incorporated as part of the counting gas. Anderson and Levi[12] have shown that for carbon-14 the greater sample accommodation of the screen-wall counter approximately compensates for the greater efficiency of the internal gas Geiger counter. For less energetic betas, such as tritium, the gas counter becomes more efficient; for more energetic betas, the screen-wall counter is preferred. This conclusion does not apply to high-pressure proportional gas counting. On the other hand for small samples or in certain routine measurements the internal gas Geiger counter is preferred.

In an effort to increase the sensitivity of measurement of natural radiocarbon, DeVries and Barendsen,[13] Suess,[14] Crathorn,[15] Fergusson[16a] and Williams and co-workers[16b] have developed the low-level internal gas proportional counter using acetylene or carbon dioxide at pressures above one atmosphere. The increased pressure gives increased sensitivity. The proportional region allows further reduction of background rate by pulse height discrimination. The theory, design, and operation of proportional counters have been covered extensively in the literature.[17-20]

Early efforts to use carbon dioxide as a counting gas were unsuccessful because of electron-attachment by electronegative impurities. It is necessary, therefore, to remove these impurities by a procedure such as that of Rafter[21] or DeVries and Barendsen.[13] The carbon dioxide is absorbed on calcium oxide at 700 to

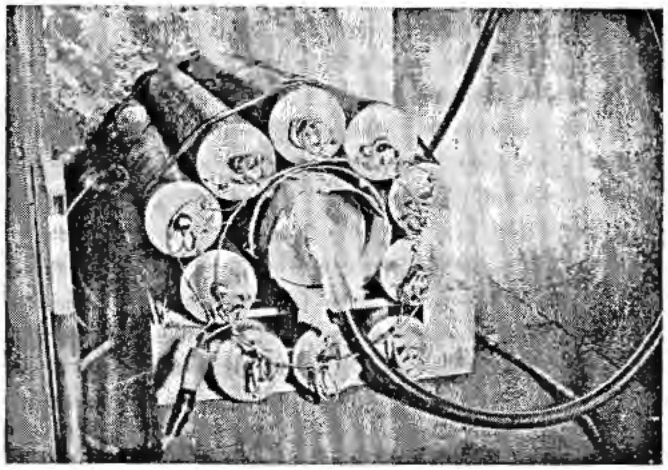

Figure 1

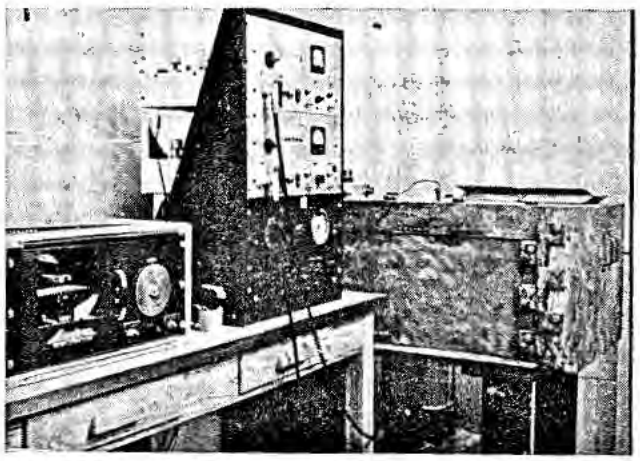

Figure 2

750°C and re-evolved at 800 to 900°C. It has been shown by Fergusson[16a] and others that the electronegative impurities remain on the lime. The purity requirements are very stringent. In order to keep the electron loss by attachment less than 1 per cent, the concentration of oxygen must be less than 1 in 10^6 and the concentration of chlorine less than 1 in 10^7. These purity specifications and the relative complexity of the associated electronics make this beautiful method useful chiefly for such problems as radiocarbon dating of old samples. The absolute efficiency for carbon-14 is about 68 per cent.[16a] Although much higher efficiencies could be obtained, the increase in background rate would increase the statistical error of the net sample count.

Foil Counters

The third type of low-level gas-tube counter is the foil counter of Libby and co-workers.[22, 23, 24] In its simplest form this counter is a cylindrical thin-wall Geiger flow counter. The wall is made of an aluminum-coated plastic film called Mylar[25] which is 0.97 mg/cm² in density thickness. Such a film will pass almost 75 per cent of the betas of carbon-14. Furthermore the cylindrical shape provides a large surface area in contrast to the usual commercial counter. In design cylindrical end pieces of plastic are fixed with respect to each other by thin brass rods. These rods and end pieces support the plastic foil wall which is mounted with the aluminized surface inward as the cathode. The counter gas which is maintained very slightly above atmospheric pressure is usually a mixture of helium and 2 per cent isobutane. This gas is commercially available as "Q" gas.[26] A foil counter and sample holder made by Dr. Schrodt is shown in Fig. 3.

In counting solids with the foil counter the sample is usually mounted on the inside of a split cylinder of plastic. This sample holder is in turn supported concentrically around the foil counter. It is not difficult to obtain about 40 per cent geometry. In mounting the sample it is slurried with a volatile liquid such as methanol or ether plus a small amount of agar in alcohol. Although the operation of spreading this slurry evenly onto the sample cylinder appears to be difficult, it actually can be done easily and routinely. The spreading is done with a glass rod and spatula and the slurry is usually dried briefly with a hair dryer or heat lamp. In the absolute assay or the precision counting of very small samples, it is more convenient to convert them to finite size by homogeneous mixing with an inert material. Schrodt and Libby[23, 24] have shown that talc, acid magnesium metasilicate, is an excellent material for routine use in this manner. Of course care must be taken to avoid radioactive contaminants such as radium. In this regard it is interesting to note that the plastic sample cylinders should be made from ancient carbon compounds, e.g., from petroleum. A cylinder containing only contemporary carbon would add about 6 counts per minute to the background rate of a counter of 1.5 inch diameter by 10 inch length.[23]

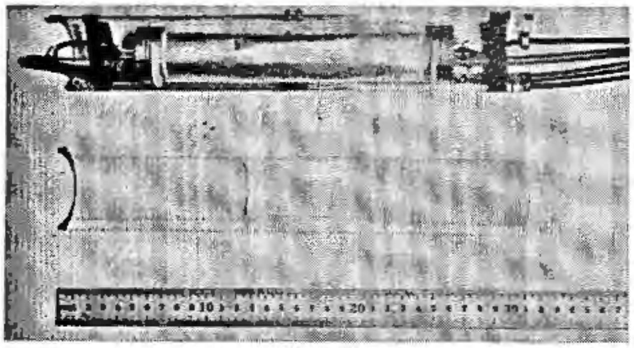

Figure 3

In counting liquids with the foil counter, refrigeration or a cover of rubber-hydrochloride have been used for volatile liquids. If care is taken to check on fractionation effects, a liquid of low volatility can be counted absorbed in blotter paper, as a slurry in talc, or as the liquid in a shallow tray.

In counting gases with the foil counter several modifications may be used. If a small sample is required, a modification of the 4π counter may be used in which the sample is placed in one hemisphere separated by a Mylar foil from the other hemisphere which operates as a Geiger counter with "Q" gas. For larger samples and for greater sensitivity a cylindrical foil counter may be mounted inside a metal or glass jacket as shown in Fig. 4. The sample to be counted is introduced by flushing into the annular space; alternatively the annular space may be evacuated simultaneously with the counter before filling. During these operations a simple mercury U-tube may be used to warn against a dangerous pressure differential. The relative simplicity of instrumentation and the lack of rigid purity specifications on the gas sample make this method desirable for many applications.

An interesting proportional counter using the thin foil between sample and detector is the Sugarman counter.[27] This device is a methane-flow proportional counter with foil window. The precision design and segregation of counter gas give remarkably stable and reproducible operation. Plateaus are long, flat, and reproducible over years of operation; counts of a sample taken on two counters agree within statistics to 0.1 per cent.[28] For moderate-level counting and ordinary low-level counting of strong betas this counter is very useful.

The various gas-tube counters which have been described will be compared and summarized following a discussion of the low-level scintillation detectors.

LOW-LEVEL SCINTILLATION COUNTING

In recent years improvements in photomultiplier tubes have made possible low-level counting with scintillation devices. In general the advantages over gas-tube counters depend upon the greater stopping

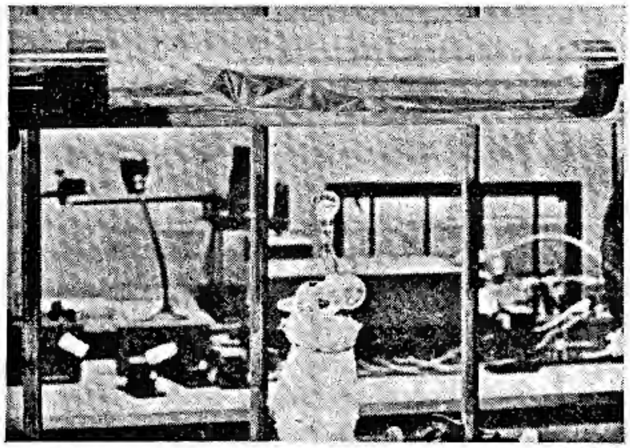

Figure 4

power of a crystal for gamma rays and upon the larger inherent sample handling potential of liquid scintillators for betas. The principles of the scintillation counter have been reviewed extensively.[29–33] The applications to low-level counting will be discussed in terms of the low-level sodium iodide scintillation spectrometer for gammas and the multiple channel liquid scintillation spectrometer for betas. The single channel liquid scintillator will be mentioned briefly.

In low-level gas-tube counting the reduction of the background rate was accomplished by massive shielding and anti-coincidence meson shielding. In low-level scintillation counting some massive shielding is also used. In addition, pulse height analysis by at least two discriminators electronically discards the meson counts and, in many applications, the background events of lower energy than the counted pulses. In view of the energy discrimination less massive shielding is required in many applications. The most satisfactory shielding material is mercury.

Low-Level Scintillation Spectrometer

For low-level gamma counting a sodium iodide scintillation spectrometer is modified in two ways. Firstly the crystal is surrounded with the mercury shield. Secondly the electronics are designed for maximum stability. It should be possible to focus on a photo-peak and remain there during 48 hours of counting. Particular attention must be given to the long-term stability of the high voltage supply which operates the photomultiplier tube. This tube should have a low thermionic noise rate at room temperature.[34]

Several studies have been done using instruments of this type. Arnold's measurements on lutecium-176[35] and a tracer study with chromium-51[36] are examples. In the latter study with only moderate attention to low-level detection, the counting rates were 20 times higher with the scintillation spectrometer as compared with a Geiger counter. Another study was done using iodine-131 for the measurement of a very slow reaction.[37, 38] This apparatus is shown in Figs. 5 and 6. The mercury shield is clearly visible and in Fig. 6 contains a liquid sample. Using a reasonable amount of this tracer the limit of detection with the low-level scintillation spectrometer gave a theoretical limit of greater than 10^6 years for measureable half-reaction. In this study, the counting rate of a liquid sample containing the iodine-131 was 100 times greater in the scintillation spectrometer as compared with a Geiger counter of the same size and geometry as the sodium iodide crystal. The efficiency of detection will vary with the energy of the gammas. Up to 200 kev the efficiency of a one-inch crystal is 100 per cent, at 300 kev it is about 75 per cent, and at 1 Mev approximately 10 per cent.[39] Of course, an efficiency of 100 per cent can be obtained with any given energy by the use of a large enough crystal.

Liquid Scintillators

The development of low-level liquid scintillators was done chiefly by Hayes and his collaborators at Los Alamos[40, 41] and by Arnold at the University of Chicago.[42] In liquid scintillation counting the sample is simply incorporated in solution with a liquid scintillator. This arrangement gives essentially 100 per cent geometry and no self-absorption. Unfortunately the decreased efficiency of liquid scintillators over solid scintillators and the desirability of detecting the lower energy pulses of beta spectra require the measurement of "equivalent electron" pulses or those pulses which result from single electron emission from the photocathode of the photomultiplier tube. These pulses, however, are indistinguishable in size from the thermionic pulses of the tube. This problem is solved by the use of two photomultipliers looking at the solution. By placing the pulses from

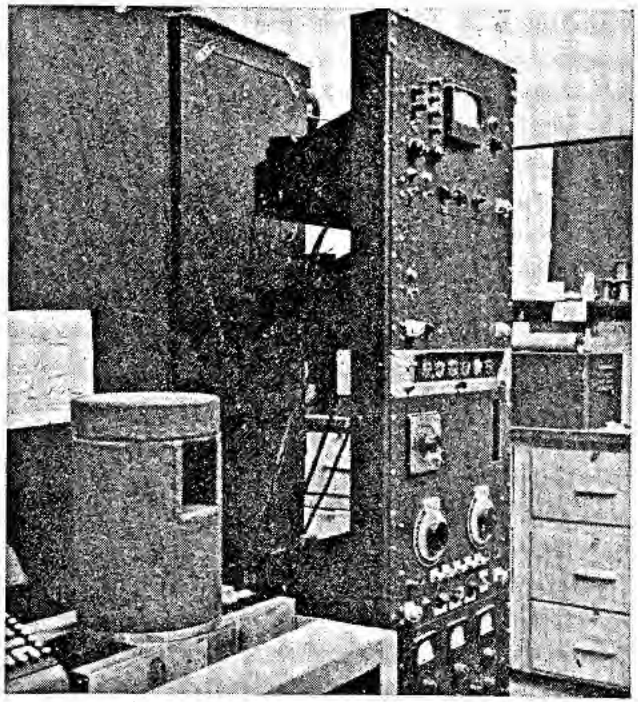

Figure 5

Figure 6

these two channels in coincidence a large discrimination is accomplished against the separate random thermionic pulses of each tube. In addition, some cooling is required, usually to a temperature of −20°C. Such an arrangement essentially eliminates tube noise provided fast electronics are used.[43] Our apparatus is shown in Fig. 7.

Although the choice of the liquid phosphor solution depends somewhat on the choice of photomultiplier, an excellent solution consists of 2,5 diphenyl oxazole as the primary solute and 1,4 di(5-phenyl 2 oxazolyl)benzene as the secondary solute or wavelength shifter in toluene. The sample to be counted is incorporated in this mixture and must not quench the scintillation. Although solutions can be prepared to count almost any beta emitter, the cost of this instrument is not usually justified except for the measurement of weak betas. Thus the counting of tritium and carbon-14 has received much attention.[40–44] Although tritium can be counted in the form of water added with alcohol to the solution above, perhaps the best procedure is that of Nir.[45] Here the THO is mixed with fuming sulfuric acid (or SO_3) and toluene. The tritium enters the toluene by exchange and the latter is separated and used for the solvent of the scintillation solution. With carbon-14 the choice of this method may depend upon the chemical form of the sample. One of the advantages of the multiple channel liquid scintillator is the unlimited sample size which can be accommodated in principle. Thus Reines and his collaborators at Los Alamos have constructed an instrument containing 300 liters of scintillation solution.[46, 47] In this counter, which was used in a search for direct evidence of a neutrino reaction, 90 photomultipliers were used to detect the scintillation pulses. A multiple channel liquid scintillator is shown in Fig. 8.

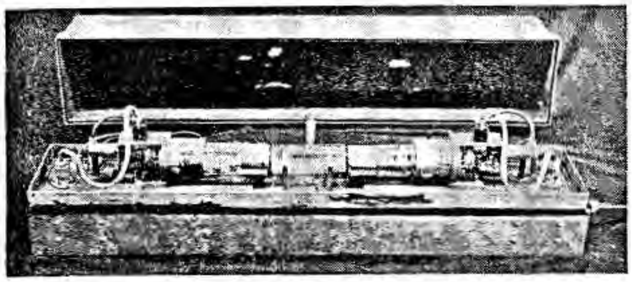

Figure 7

In regard to the sensitivity of the dual channel liquid scintillator Arnold obtains an absolute efficiency for carbon-14 of about 60 per cent for a 30 ml solution containing approximately 50 mol per cent of carbon.[48] The counting rate was 70 counts per minute against a background of 12. In the same instrument the efficiency for tritium was much less, approaching 25 per cent.

The possibility of using a single channel liquid scintillator for low-level counting depends largely on the development of better photomultiplier tubes. By highly selecting his tube, Pringle has obtained an efficiency of 35 per cent for carbon-14 with a background which was approximately 20 per cent tube noise.[49]

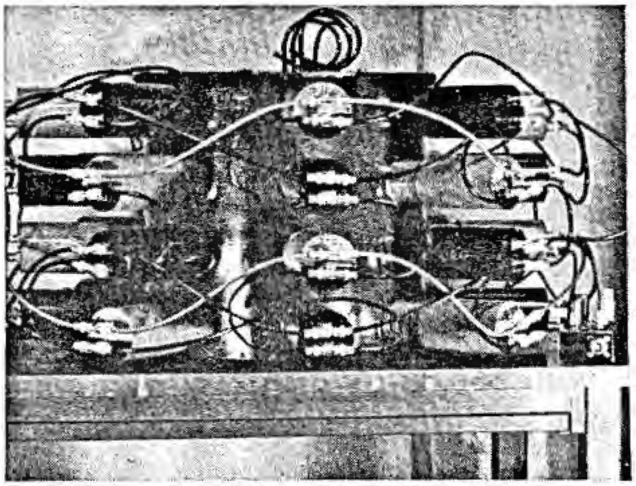

Figure 8

One interesting potential of both liquid and solid scintillation counting is the possibility of multiple-tracers. For example, with a sodium iodide scintillation spectrometer the photo-peaks of such tracers as chromium-51 and iron-59 can be separated.[50] With the liquid scintillator tritium and carbon-14 can be counted simultaneously.[44]

APPLICATIONS AND CONCLUSIONS

In choosing a low-level counting method for a specific job a number of factors must be considered. Some of these are the type and energy of the radiation, the size of the samples which are available, and the degree of sensitivity required. In Table II the low level instruments for beta counting are summarized according to these factors. Weak betas are nu-

The Theory of Cavity Ionization

By L. V. Spencer and F. H. Attix,* USA

Cavity ionization chambers, which are widely used as radiation dosimeters, are often calibrated against primary standard dose-measuring devices such as free-air ionization chambers. Sometimes, however, they themselves serve as primary standards. This use requires an adequate theory of the relationship between the cavity gas ionization and the energy absorbed in the cavity walls.

The physics of the cavity ionization chambers is rather well known. Incident gamma radiation penetrates easily through the chamber walls generating meanwhile, through its interaction with atomic particles, a spectrum of fast electrons. Many electrons cross the cavity and produce ionization within the cavity gas. The amount of cavity ionization is measured and used as an index of the total incident radiation dose. The connection between the cavity gas ionization and the energy absorbed in the cavity walls involves two factors: (a) the ionization produced when a given amount of energy is dissipated in the cavity gas, and (b) the energy dissipated in the cavity gas relative to that dissipated in the cavity walls. The first factor is simply the constant W—the volts per ion pair formed in the gas—which we shall assume to be known. The second factor is what we shall concentrate our attention on in this manuscript.

The cavity ionization depends rather strongly on the material comprising the cavity walls. If the cavity gas and walls are of the same material, e.g., ethylene gas in a cavity with polyethylene walls, the theory is especially simple: the energy dissipated in the cavity gas equals the energy dissipated in an equal mass of wall material.[1] Unfortunately, this situation is seldom realized in practice. More frequently the cavity gas is air and the wall material is not condensed air at all, but is water, polystyrene, aluminum, or sometimes even lead. The classical theory of this more general situation is that of Bragg and Gray, who relate the energy dissipated in the cavity gas to the energy dissipated in the cavity walls through a ratio of the stopping power for electrons in the gas relative to the walls.[2, 3] As generally stated, this relation is thought to hold more exactly the smaller the cavity. Thus, much experimentation in recent years has involved extrapolation to zero cavity size.

The Bragg-Gray theory is based on an exceedingly simple schematization: the photon-ejected electrons are thought of as slowing down continuously, by a sort of "friction" process as they penetrate. Energy is considered as dissipated only along the track and at a rate given by the local stopping power. Accordingly, secondary electrons which may be generated by the fast photon-ejected primary electrons through knock-on collisions are implicitly assumed to travel nowhere, but rather to dissipate their energy at the place where they are generated.

Recent progress in the analysis of secondary "knock-on" electron processes now enables us to determine more accurately the energy dissipated in the cavity gas relative to that dissipated in the wall material. In the following paragraphs we describe a theory which takes into account the existence and travel of fast secondary electrons. The need for such a development was recently indicated in experiments by Attix and DeLaVergne, who found that the cavity ionization measured as a function of wall atomic number differed appreciably from the predictions of Bragg-Gray theory.[4] These measurements are consistent with earlier experiments by Gray, using lead-walled chambers.[3] Gray also measured the cavity ionization as a function of decreasing air pressure.[3] He found a lack of proportionality which has never previously been explained because the Bragg-Gray theory predicts proportionality between the ionization and the gas pressure.

The theory which we describe yields results in good agreement with most of the experiments of Attix and DeLaVergne. It also predicts variations of the cavity ionization with air pressure which agree rather well with Gray's results. Unfortunately, the interpretation of extrapolation to zero cavity size is shown to be more complex than had been believed.

THE SCHEMATIZATION

Consider a small air cavity surrounded by wall material with atomic number Z. Throughout both wall material and air cavity electrons are generated with initial kinetic energy T_0. The electron source strength is everywhere proportional to the local density of material, and the cavity walls are thick compared with all electron ranges.

Through inelastic collisions the source electrons lose energy and generate secondary electrons of various energies. Thereby an electron spectrum $I_Z(T_0,T)$ occurs which traverses all portions of the wall material. We assume that the presence of the small air cavity does not disturb this spectrum, so that the

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* National Bureau of Standards, Washington, D.C.

same spectrum of fast electrons traverses the cavity.[5]

As the fast electrons, mostly from the walls, sweep across the cavity they generate ions and secondary electrons within the cavity. The inelastic collisions which occur within the cavity fall rather naturally into two groups: (a) those which generate very slow electrons with too little kinetic energy to span the cavity, and (b) those which generate fast electrons with plenty of energy to leave the cavity and penetrate the cavity walls. In collisions of type (a) the energy given to the secondary electron remains entirely within the cavity. On the other hand, in collisions of type (b) both primary and secondary electrons contribute little of their energy to the cavity, and this through further collisions of type (a) on the part of both fast electrons. Thus the fast secondary electron resulting from collisions of type (b) might as well be considered part of the incident spectrum, since it is indistinguishable from it.

We therefore schematize the problem as follows: all collisions which result in a net energy transfer less than a threshold value Δ are considered as dissipating their energy on the spot. Conversely, all collisions which result in a net energy transfer greater than Δ are not considered as dissipating any energy at all. The resulting secondary electron is considered as part of the incident spectrum. The energy Δ, which separates dissipative from non-dissipative collisions is taken to be that kinetic energy which an electron requires in order to span the cavity.

It is clear that such a two-group theory is an oversimplification. For example, we still assume that the electron spectrum is characteristic of the wall material even though the cavity must make some modification particularly for energies T not much greater than Δ. Likewise there is the question of the exchange of energy between cavity and walls in the form of very slow electrons. In this latter question our schematization is equivalent to assuming a balance with no net energy transfer, which is fairly reasonable on the basis of semi-quantitative arguments. In general we hope that these types of inaccuracies in the schematization can be allowed for by adjusting the value of Δ rather than changing the two-group schematization. The choice of Δ is not a very critical matter since the calculations are rather insensitive to its value.

CALCULATIONS

We want to determine the energy dissipated per gram of cavity gas relative to the energy dissipated per gram of wall material. In accordance with our schematization this quantity is given by the following ratio:

$$f_z(T_0, \Delta) = T_0^{-1} \int_{\Delta}^{T_0} dT I_Z(T_0, T) S_{air}(T, \Delta) \quad (1)$$

where $I_Z(T_0,T)$ is the electron spectrum, including both primaries and secondaries, and $S_{air}(T, \Delta)$ is the stopping power limited to energy losses smaller than Δ. The integral represents the energy dissipated in the cavity and T_0^{-1} represents the corresponding energy dissipated in the wall material. At energies $T \geqslant T_0/2$, from which region secondaries are excluded by definition, I_Z should be very nearly the reciprocal stopping power of the wall material.

Note how this formula differs from the corresponding Bragg-Gray formula: In Bragg-Gray theory Δ would be set equal to $T/2$ in the quantity $S_{air}(T, \Delta)$, so that *all* energy given to secondary electrons is considered stopping power. Correspondingly I_Z would be taken as the reciprocal stopping power, which is the continuous-slowing-down approximation to the spectrum of primary electrons *only*. Our quantity $I_Z(T_0,T)$ becomes much larger than the reciprocal stopping power at low energies because of the inclusion of "fast" secondary electrons. Correspondingly $S_{air}(T, \Delta)$ is smaller than the stopping power for air because the energy given to "fast" secondaries is not included as stopping power in our schematization. It would be a mistake simply to insert the spectrum $I_Z(T_0,T)$, including secondary electron energy, into the Bragg-Gray integral; for the same energy should simultaneously be subtracted from the stopping power.

The two quantities $I_Z(T_0,T)$ and $S_{air}(T, \Delta)$ are readily calculated. The latter is determined in the same manner as the Bethe stopping power formula.[6] Methods for calculating the former, together with sample results, are in the literature.[7] The product of the two quantities is reasonably well behaved, so that the integral in Equation 1 is easy to carry out numerically. We have therefore performed a number of such integrations. The results are contained in Table I. We have not carried out calculations for extremely small Δ because of our poor knowledge of the cross sections for the production of secondary electrons in this region.

DISCUSSION

There are several comments worth making about Table I. First, since carbon is nearly air-equivalent, having a Z only slightly less than that of air, it is gratifying to note that f_Z for carbon is only slightly less than unity, independent of T_0 and Δ. Variations in f_Z of 1% or less are not significant because the spectra used in the calculations are not that accurate.

Secondly, for fixed Δ and Z, the variations in f_Z with T_0 are slow and very similar to those in the analogous stopping power ratios. This feature is very useful because it allows the correction to Bragg-Gray to take the form of a multiplicative constant which depends on Δ, i.e., the cavity size.

Finally, it can be seen that for constant T_0 and decreasing Δ, f_Z increases more and more rapidly. This is because contributions to the integral in Equation 1 from energies T near Δ are important and involve the stopping power behavior at very low energies. This feature is more prominent for high-Z wall materials, as is expected.

TABLE I. Values for $f_Z(T_0, \Delta)$. All Energies Are in Units of mc^2, and The Stopping Powers Used Were in Units $mc^2/(gm/cm^2)$

Z	T_0	0.005	0.01	0.02	0.04	0.08	0.16
	2.56	0.99	1.00	1.00	1.00	1.00	0.99
C	1.28	0.99	1.00	1.00	1.00	0.99	0.99
	0.64	1.00	1.00	1.00	0.99	0.99	
	2.56	1.15	1.15	1.14	1.13	1.13	1.12
Al	1.28	1.17	1.16	1.15	1.14	1.13	1.12
	0.64	1.18	1.17	1.16	1.15	1.13	
	2.56	1.44	1.40	1.38	1.35	1.34	1.32
Cu	1.28	1.47	1.42	1.39	1.37	1.34	1.32
	0.64	1.50	1.45	1.41	1.38	1.35	
	2.56	1.77	1.69	1.63	1.59	1.56	1.53
Sn	1.28	1.82	1.72	1.66	1.62	1.58	1.54
	0.64	1.86	1.76	1.70	1.64	1.59	
	2.56		2.04	1.93	1.86	1.81	1.76
Pb	1.28		2.11	1.99	1.91	1.85	1.80
	0.64		2.16	2.04	1.95	1.88	

COMPARISON WITH EXPERIMENTS

In the experiments by Attix and DeLaVergne, two plates separated by an air gap were irradiated with gamma rays of different energies. The plates were thick compared with electron ranges but thin compared with photon mean free paths. The ionization per gram of air in the air gap was measured as a function of the plate material and also as a function of the plate separation.

To compare these measurements with our theory, the experimental data was first corrected for X-ray attenuation in the plates, for the contribution to the ionization due to X-rays which are both scattered and absorbed in the plates, and for side losses. The corrected data was then divided by the energy absorption coefficient $(\mu_{en})_Z$ of the gamma rays evaluated at the primary photon energy. The resulting values for f_Z were normalized to agree with theory for a 1-mm gap between graphite plates.

Corresponding theoretical values for f_Z and the analogous Bragg-Gray function were evaluated at the mean energy $\overline{T}_0$ of the photon-ejected electrons. The choice of Δ was made as follows: A theoretical curve of true range vs energy for electrons in air based on the Bethe formula was modified by reducing the ranges 20% to account for foreshortening due to elastic scattering.[6, 8] Using this curve the parameter Δ was identified as the energy corresponding to the "foreshortened" range equal to the plate separation.

Table II contains the comparison for three incident gamma ray energies, namely Co^{60} (1.2 Mev), Cs^{137} (0.67 Mev), and Au^{198} (0.41 Mev). The predictions of the new theory agree rather well with experiments. There is one aspect of this comparison which requires further comment, namely the tendency of the experimental results to fall below theory for high-Z wall materials *and* low incident gamma-ray energies. An explanation for this effect may well be that multiple transits of the cavity by electrons, for high-Z wall materials, enhance the effectiveness of the cavity in distorting the electron spectrum. This distortion would extend to energies T several times the Δ used by us. For low source energies this becomes noticeable and represents a breakdown of the conditions under which all present cavity theory is valid.

Finally, in Fig. 1, variations with air pressure in a lead-walled cavity as measured by Gray (circles) are compared with variations predicted by present theory (solid line) by choosing Δ in the manner indicated earlier in this section to correspond at room pressure to a range in air of 3 mm. The experimental values and theory were normalized to agree at room pressure.

FINAL REMARKS

The strong influence of low-energy stopping powers, which are not well known, as exhibited by the variation of f_Z with Δ, raises the question as to

TABLE II. Comparison of $f_Z(\overline{T}_0, \Delta)$ Values from Present Theory with Experimental Results of Attix and DeLaVergne and with Mean Stopping Power Ratios from Bragg-Gray Theory. The Last Two Columns Relate to a 1-mm Air Gap.

Source	*Z*	*Bragg-Gray*	*Present theory*	*Attix-DeLaVergne*
	6	0.99_2	0.99_5	0.99_5
	13	1.13	1.15	1.14
Co^{60}	29	1.33	1.41	1.40
(1250 kev)	50	1.56	1.69	1.73
	82	1.79	2.03	2.14
	6	0.98_8	0.99_4	0.99_4
	13	1.14	1.17	1.14
Cs^{137}	29	1.36	1.44	1.42
(670 kev)	50	1.60	1.72	1.74
	82	1.85	2.07	2.02
	6	0.98_7	0.99_5	0.99_6
	13	1.15	1.18	1.18
Au^{198}	29	1.39	1.46	1.50
(411 kev)	50	1.64	1.74	1.73
	82	1.90	2.10	1.99

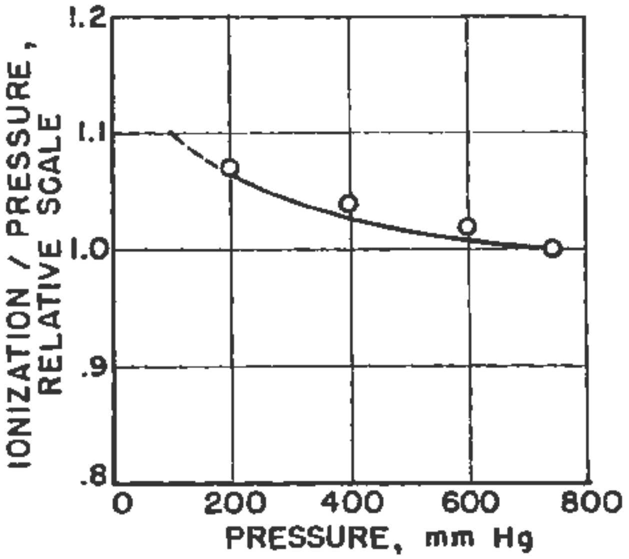

Figure 1. The ionization to pressure ratio as a function of air pressure in a lead-walled chamber. The circles are experimental values obtained by Gray and the solid line represents present theory. The two were normalized to agree at room pressure

the meaning of extrapolation measurements. It is clear that the extrapolated value is not independent of the smallest cavity size for which measurements are made. Thus the concept of "extrapolated value" is not as well defined as is generally assumed. At least for measurements in non-air-equivalent materials, it would seem preferable to relate calculations to definite cavity sizes.

ACKNOWLEDGEMENT

We take pleasure in thanking Dr. Ugo Fano for many discussions of this material and for editorial comments and suggestions.

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Dosimetry of Thermal Neutrons and Gamma Rays in Reactors

By J. Sutton,* I. Draganic,* and H. Hering,† France

Studies on the chemical behavior of certain substances under the action of radiation in reactors require dosimetric follow-up. When irradiation is made —as was our case—in the reflector, the proportion of epithermal neutrons is very small (1%), as was shown by a comparison of the activities of cobalt exposed under graphite and under cadmium respectively. Accordingly, the work limits itself to the determination of gamma radiation on the one hand, and that of the neutrons on the other.

The methods described below, which are used routinely in the laboratories of the Atomic Energy Commission, include some novel principles or details which have already been described in preliminary papers.[1, 2, 3]

For the dosimetry of gamma radiation, an "aerated" or ventilated version of the Fricke ferrous sulfate dosimeter[1] is suitable up to doses of approximately 10^6 rep. The use of aerated solutions of oxalic acid, which has already been described by one of us[2] makes it possible to reach 10^7 rep.

The dosage of the thermal neutrons was carried out with cobalt sulfate solutions, or with filter paper soaked in such salts. The over-all chemical effect of the gamma radiation and of the slow neutrons is achieved by means of ferrous sulfate solutions to which lithium sulfate has been added: the heavy particles released by the Li^6 (n, α) H^3 reaction act, as it were, as "chemical revealers" for the slow neutrons.

We shall limit ourselves, in this paper, to giving the details of the experimental methods.

CHEMICAL PRODUCTS

The chemical products used are of the Prolabo RP brand, except for the lithium sulfate (Analar, BDH). They are used as received, without any additional purification, since preliminary tests have shown that additional recrystallizations bring about no perceptible change in their behavior under irradiation.

The Mohr salt used contained less than 10 ppm of Cu^{++} (polarographically determined) which is well under the minimum amount which could influence the results. The water was distilled, under nitrogen flow, with $K_2Cr_2O_7 + H_2SO_4$, then with $KMnO_4 + NaOH$ and finally, without any dissolved reagent, in special equipment entirely made of silica. The cobalt sulfate was free from Ni.

METHODS

In order to chart the intensity at various points around a 7.5 curie source of Co^{60}, we used $10^{-3}M$ Mohr salt solution and a 0.8 N sulfuric acid concentration. An amount of 10 ml of the aerated solution was inserted into Pyrex test tubes of constant thickness and diameter, closed with polychlorovinyl stoppers. The solutions were thoroughly mixed prior to determination.‡ This routine precaution is particularly necessary. The finite dimensions of the source (made up of a cylinder 12 mm both in diameter and height) and test tubes cause the flux density to vary a little from one point of the samples to the next. Assuming that we have $G_{Fe^{+++}} = 15.6$ we find flux densities which vary, according to the points taken, between 10^{13} and 10^{14} ev/sec/ml.

In the reflectors of nuclear reactors, where the total doses received are much greater, the ferrous sulfate dosimeter can be used according to Rolin's technic,[3] which we employed by substituting silica ampules for the glass ones, the use of which was being ruled out by the fluxes currently found in the French reactors. The ampules were cleaned with a hot sulfo-nitric mixture, washed in distilled water then, again, in tridistilled water, and dried at 120°C. They were then half filled with the solution and cooled in liquid nitrogen, until some oxygen condensed, after which they were quickly sealed with a small gas-oxygen flame. Several ampules prepared in this fashion were opened without having been subjected to any irradiation. We noted that absolutely no oxidizing had taken place in the course of the sealing process. By this device, we measured doses reaching 10^6 rep in an aerated solution, with intensities varying between 4 to 11×10^{14} ev/sec/ml in the Châtillon reactor, and between 14 to 70×10^{14} ev/sec/ml in the Saclay reactor, according to the distance between the ampule and the heavy water container.

Rolin[3] also described thermal neutron dosimetry with cobalt salt impregnated paper discs. We have

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‡ By the absorption of 304 mμ light waves.

improved on the precision of this method by using an Agla micropipette for soaking the discs, and by a careful electrolytic analysis of the cobaltic solution. These monitors, which can be reproduced to ± 0.5%, contained 1 mg of Co in the case of one reactor, and 10γ in another. The paper, which was thoroughly wetted by the solution used, was dried under an infra-red ray lamp. Each irradiated ampule carried a circular paper disc, protected by two filter paper squares, and fixed with adhesive tape. Counting took place a fortnight after irradiation was finished, in order to allow the short-lived activities to decay. These monitors kept well in the Châtillon reactor, but after about one week, in the Saclay reactor (doses above 8×10^7 r), they became fragile, and mounting them for counting purposes turned difficult. Counting was by means of a bell-type counter and a very readily reproducible set-up. The whole instrumental set-up was calibrated by means of discs impregnated in an active cobalt sulfate solution, itself standardized by an absolute coincidence method due to M. Grinberg. The neutron fluxes so determined in our experiments varied from 0.8×10^{11} to 1.5×10^{11} neutrons/cm^2/sec for the reflector of the Châtillon reactor, and from 0.1×10^{12} to 1.0×10^{12} neutrons/cm^2/sec for that of the Saclay reactor.

RESULTS

I. Here is a summary of the results obtained with solutions of ferrous salts and lithium.[4] Some 1-ml samples of 0.02 *M* Mohr salt in 0.8 *N* sulfuric acid, and 0.09 *N*, 0.45 *N* or 0.90 *N* in lithium sulfate, are sealed under oxygen pressure in common glass ampules. These are irradiated in the reflector of reactor "Zoé," along with similar ampules filled with a lithium-free Mohr salt solution, and others which contain 1 ml of a cobalt sulfate solution in 0.8 *N* sulfuric acid, and some cobalt impregnated paper monitors. For a given irradiation, the amount of oxidized ferrous sulfate increases as a linear function of the lithium solution. For a given lithium concentration, it increases as a linear function of the absorbed neutron dose (determined, despite the none too convenient geometry, by the activity induced in the cobalt sulfate solutions). By using $G_{\mathrm{Fe}} = 15.6$ for the oxidizing of ferrous iron by gamma radiation, the efficiency of oxidation by the fission fragments of the Li^6 nucleus gives a value of 5.0 ± 0.6, which is in fair agreement with the findings of other authors.[5] Some irradiations carried out inside a bismuth brick show that the difference in the quantities of Fe^{2+} iron oxidized in the presence and in the absence of lithium respectively, is independent of the γ flux. On the other hand, when irradiation was carried out under cadmium, the quantity of oxidized Fe^{2+} was constant, whether or not lithium was present in the solutions. This further demonstrates the fact that the excess oxidation found under bismuth or graphite is truly due to the neutrons. Thus, the solutions of Mohr salt and lithium sulfate in 0.8 *N* sulfuric acid are excellent dosimeters for the gamma rays associated with thermal neutrons.

II. The irradiation of aerated solutions of oxalic acid by gamma rays was carried out, not only with sources of Co but also in the reflectors of both reactors.

For Co^{60} irradiation, the solutions were placed in Pyrex tubes. In the reflectors, use was made of solutions sealed in glass or silica ampules, with an excess of oxygen. Oxalic acid titer was determined hot, in the presence of sulfuric acid, with a $\mathrm{KMnO_4}$ solution. Determination was made at the same time on a sample of the original solution. The difference between the values found gave the result sought.

Under low concentrations and for small doses, the method becomes inaccurate. Thus, for the titration of solutions of 2 to 10 mM/l having been subjected to doses of about 3×10^{18} ev/ml, more $\mathrm{KMnO_4}$ is needed as a control, due to the formation of $\mathrm{H_2O_2}$ which takes place faster than oxalic acid is decomposed. This effect becomes negligible when the oxalic acid concentration exceeds 50 mM/l.

We have endeavoured, without success, however, to reveal intermediate products of oxalic acid decomposition, such as formaldehyde and the formic, glyoxalic and glycocollic acids;[6] the reaction products appear to be only $\mathrm{CO_2}$ and $\mathrm{H_2}$.[7] Other compounds may appear following the irradiation of concentrated solutions under very high intensities. We shall, at a later date, publish the results given by such experiments, in aerated solutions or otherwise, with tagged oxalic acid.

In view of the inaccuracy of the volumetric determination of small variations in concentration, the lower level of practical utilization is about 10^{19} ev/ml. Prolonged irradiation by cobalt sources has shown that the ratio of the amount of decomposed acid to the dose received remains linear up to 10^{21} ev/ml at least. The yield, *G*, indicated above remains the same as the initial concentration varies from 50 to 700 mM. On the other hand, we have measured, in P2, doses which reached up to 4×10^{21} ev/ml approximately, assuming that the oxidation yield remain constant up to such doses. This assumption is justified by the fact that, for each of the positions chosen in the reflector and calibrated by cobalt activation and ferrous sulfate oxidation, the ratio of the consumed oxalic acid to cobalt activation remains constant, at least up to the maximum dose shown above. The results of the experiments carried out in P2 show greater dispersion than those in Zoé or with cobalt sources, but agreement between the three series remains acceptable. If $G_{\mathrm{Fe}} = 15.6$ for γ radiation, one will find, for the decomposition oxalic acid, 4.9 ± 0.4 molecule/100 ev with cobalt radiation and 5.2 ± 0.5 in the reflectors of Zoé and P2. Irradiations under bismuth and cadmium have shown that the thermal neutrons have no influence. The oxalic acid solutions so studied between concentrations of 50 and 700 mM/l, in radiation fields of 0.3 to 120 rep/sec constitute an excellent dosimeter for γ-rays for prolonged irradiation in the reactors. They offer the additional advantage of being practically devoid of activation under such irradiations.

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Absolute Dosimetry of Cobalt-60 Gamma Rays

By J. S. Laughlin, S. Genna, M. Danzker, and S. J. Vacirca,* USA

The output of any radiation source can be precisely specified by its intensity, or quantity of radiation. As defined by the International Commission on Radiological Units,[1] quantity of radiation is the time integral of intensity and can be expressed in ergs per square centimeter. Intensity can be determined directly and precisely with the calorimetric method described here which measures temperature changes of the order of 10^{-5} degrees Centigrade.[2]

Knowledge of the absorbed dose is even more essential in many radiation experiments. Absorbed dose[1] refers to the amount of energy imparted per unit mass at a particular point in an absorbing material, and is expressed in rads. (One rad equals 100 ergs/gram.) The calorimetrically determined quantity of radiation can be converted into absorbed dose at given depths in an absorbing medium whose physical properties are known. This has been done for the cobalt-60 gamma rays in various materials and the absorbed dose so determined has been compared with that obtained from cavity ionization measurements in the same absorbing medium.

COBALT SOURCE

Experiments were performed with the Memorial Center telecobalt unit. The unit is commercially known as the "Theratron" and is manufactured by the Atomic Energy Commission of Canada. The cobalt-60 source is in the form of a number of small pellets contained in a cylindrical steel shell. The shell has an inside diameter of 2.5 cm and a length of 1.2 cm. A 2.5 cm diameter lead collimator defines the beam. The collimator is 18 cm long and its exit diaphragm is 47 cm from the 1000 curie cobalt-60 source.

CALORIMETER APPARATUS

A single lead-tungsten absorbing cylinder, illustrated in Fig. 1, is suspended in a vacuum chamber (Fig. 2) for use as the energy absorber for the telecobalt gamma ray beam. The absorber is suspended with nylon threads through eyelets attached to the roof of the vacuum chamber. Worm screw adjustments permit accurate alignment of the absorbing cylinder.

The absorber is constructed in three sections. The center section, or socket, consists of a silver plated cylindrical brass shell filled with lead. The two end sections, or plugs, are machined from "Hevimet" tungsten alloy. After mounting the thermistor in one plug and the heater element in the other plug, the separate elements are screwed together and soldered at the joints to insure good thermal conductivity. The diameter of the absorber is 8.9 cm and its thickness is 7.8 cm.

A Western Electric No. 12A rod-type thermistor is used as the temperature-sensitive element. The No. 12A thermistors are about 3 cm long and 3 mm in diameter. They have a high negative temperature coefficient of about 3.8%/°C and a total resistance of about 10,000 ohms at 26°C.

A rectangular groove 4 cm long, 4 mm wide, and 4 mm deep, is cut in the rear tungsten plug for the thermistor rod. The heavy electrodes of the thermistor were removed and replaced with insulated 36 B&S gauge copper wire. In order to obtain good thermal contact between the thermistor and the tungsten plug, one end of the thermistor is securely soldered to the tungsten. The remainder is insulated with a thin sheet of teflon. The entire thermistor is then imbedded in the rectangular groove with Wood's metal.

An insulated nichrome wire heating element, imbedded in Wood's metal within the front plug, serves to introduce a known amount of energy for calibration. Enameled insulated nichrome wire is soldered at one end to the tungsten plug and at the other end to 36 B&S gauge enameled copper wire. A cylindrical well machined in the "Hevimet" receives the nichrome heating element as well as a few turns of the copper wire. Wood's metal serves to secure the wire to the tungsten plug with good thermal contact.

The surface of the cylinders and the inside wall of the vacuum chamber are silver plated and finished to a high polish in order to minimize radiation heat transfer. A thin aluminum baffle surrounds the sides and ends of the cylinder in order to dampen radiation heat transfer effects of temperature fluctuations in the chamber walls.

The calorimeter vacuum chamber is immersed in a thermostatically controlled bath of rapidly circulating water. A centrifugal pump and heat exchanger unit continuously circulates and cools the water bath. Cooling is effected by means of a water cooled copper coil. A mercury thermoregulator controls two knife heaters immersed in the water bath.

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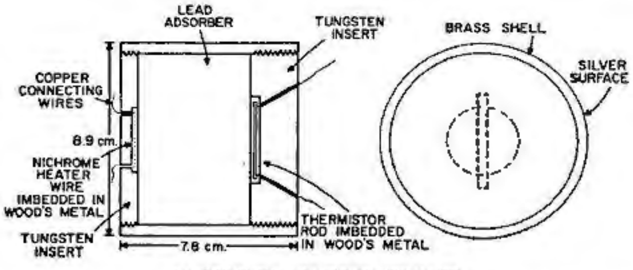

Figure 1. Construction of the calorimeter cylinder which is used to absorb the cobalt-60 gamma rays

Electrical connections to the thermistor and heater coil are made through Kovar seals on the roof of the chamber. The thermistor is connected as one arm of a Wheatstone bridge. All the elements except the control resistance box are immersed in an oil bath on the lid of the chamber in order to eliminate variable thermal emf's. The junctions of the control resistance box are insulated to eliminate rapid temperature fluctuations due to room drafts.

The bridge unbalance emf is amplified by a dc chopper amplifier and recorded continuously with an electronic potentiometer. Since the voltage output of the bridge is directly proportional to the resistance change in the thermistor arm, the bridge can be operated off balance to yield a continuous record of the resistance change of the thermistor.

CALIBRATION OF CALORIMETER APPARATUS

The heater resistance imbedded in the absorbers, as mentioned earlier, serves to introduce the known energy standard for calibration. Copper lead connections made inside the cylinder to the heating coil are brought outside the vacuum chamber through Kovar seals. A constant current through the heater is used for calibrating the instrument. The energy input is determined from the constant potential drop across the pre-determined heater coil resistance. The potential drop is measured with a Leeds & Northrup type K2 potentiometer during current input.

A series of 21 calibrations were performed yielding a sensitivity value of 0.351 ohms/joule with a standard deviation of ± 0.005 ohms/joule. The results were found to be independent of the energy input rate used for the calibrations.

IRRADIATION WITH COBALT-60 GAMMA RAY BEAM

A one-inch diameter lead collimator with its exit diaphragm at 47 cm from the source was used for all energy measurements. The beam was admitted into the calorimeter through two thin aluminum foil windows (0.007 gm/cm^2). The calorimeter was positioned so as to locate the absorber along the beam axis. The geometrical beam penumbra diameter was about 4.5 cm at the face of the absorber. This arrangement results in negligible side scatter loss through the walls of the absorber and insures the total absorption of the beam.

In order to remove any electrons that might be projected in the collimator a magnet with a field strength of approximately 10,000 gauss was positioned with its field perpendicular to the beam during irradiation of the calorimeter absorber. Ten-minute irradiations were employed. The thermistor resistance change for 19 absorber irradiations was computed as described earlier. The resultant value of resistance change per minute is

$$dR/dt = 0.0470 \text{ ohms/minute} \qquad (1)$$

with a standard deviation of ± 0.0006 ohms/minute.

It is pertinent to convert the resulting resistance change into the temperature change of the absorber. Since the sensitivity of the thermistor is approximately 380 ohms/°C, the rate of temperature change is about

$$dT/dt = 1.24 \times 10^{-4} \pm 1.6 \times 10^{-6} \,^{\circ}\text{C/min} \qquad (2)$$

A few minor corrections for backscattered and transmitted energy must be made before the final value of the energy output of the collimated beam is determined. The absorber cylinder dimensions were sufficiently large to reduce the side scatter and transmitted energy corrections to a negligible magnitude. Estimates of the loss of energy due to side scatter and transmission were made from measurements with a cylindrical ionization chamber. Integrated ionization measurements laterally around the absorber and in the rear of the absorber yielded an energy loss from the absorber of 0.25 per cent due to side scatter, and 0.45 per cent energy transmitted. In the corrections, it was assumed on theoretical grounds that the ergs/cm^2-roentgen for the lower energy scatter quanta is of the same order of magnitude as that of the higher energy quanta. The backscattered energy albedo was taken as 0.1 per cent according to the theoretical computations of Hayward and Hubbel.[3]

Combining the above results with the above corrections, the total energy output rate becomes

$$dE/dt = 0.135 \pm 0.002 \text{ joules/min} \qquad (3)$$

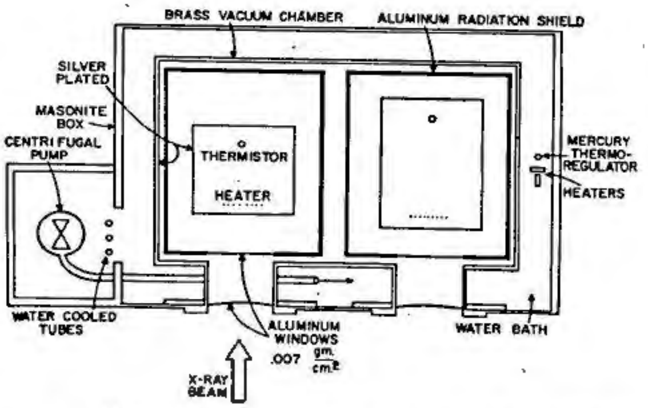

Figure 2. Horizontal cross section of the calorimeter construction

SECONDARY STANDARD COMPARISON—VICTOREEN THIMBLE CHAMBER

A suitable secondary standard for the total energy output of the beam is the integrated roentgen-cm²/min along a plane perpendicular to the axis of the gamma ray beam. Neglecting the energy absorbed in air, any plane intercepting the beam will have the same rate of energy flow through it. Similarly the integrated roentgen-cm²/min is a constant of the beam. The plane need not be perpendicular for the above to hold but is chosen as such for convenience.

The integration was performed in two steps. The roentgen/min was first measured at 120 cm from the source along the central axis. The measurement was performed with a 25-roentgen Victoreen thimble chamber previously calibrated at the National Bureau of Standards. A 3 mm wall phenol-formaldehyde (Catalin) cap was fitted over the thimble during calibration in order to measure the cavity ionization under equilibrium conditions. A 3 mm cube anthracene crystal detector was used for the relative intensity survey. The determination yields an integrated value of 401 ± 13 roentgen-cm²/min assuming an N.B.S. Victoreen calibration accuracy of ± 3 per cent, and experimental accuracy with the Victoreen chamber of ± 1 per cent, and an anthracene crystal intensity distribution accuracy of ± 1 per cent.

Combining the above results, the resultant *quantity of radiation per roentgen* in the cobalt-60 gamma ray beam is:

$$Q/r = 3370 \pm 130 \text{ ergs/cm}^2\text{-roentgen} \quad (4)$$

THEORETICAL QUANTITY OF RADIATION

The computed quantity of radiation per roentgen has been calculated from values of the true portion of the mass absorption coefficients[4] for X-rays in air. It is assumed in the computation that the average energy dissipated per ion pair formed in air is independent of the energy of the ionizing particle and is equal to 34.1 ev. If the average energy of cobalt-60 gamma rays is 1.25 Mev the true portion of the mass absorption coefficient for air is equal to 0.0268 cm²/gm. The corresponding theoretical quantity of radiation per roentgen is equal to 3280 ergs/cm²-roentgen.

Absorption and scattering of the gamma radiation within the cobalt source itself results in a degradation of the emitted photon energies. An estimation of the degradation of the emergent gamma ray beam yields an absorption coefficient of 0.0271 cm²/gm. The corresponding theoretical quantity of radiation is equal to 3250 ergs/cm²-roentgen for the degraded cobalt gamma rays.

EXTRAPOLATION CHAMBER APPARATUS

In order to correlate the calorimetric results with cavity ionization chamber measurements, an extrapolation chamber was constructed. The chamber is illustrated in Fig. 3. Four precision machined screws, having a pitch of 2 revolutions per mm, support the top movable frame of the chamber. An interlocking gear system, driven by a low speed motor, actuates the four screws simultaneously. A selsyn system geared to the screws drives a revolutions counter in a control box permitting accurate adjustment of the plate separation outside of the irradiation room. The revolutions counter is geared to indicate 10 counts per revolution of the machine screws.

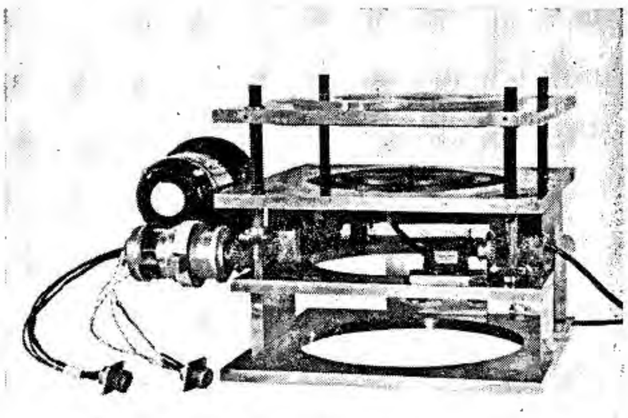

Figure 3. Photograph of the extrapolation chamber. Radiation is incident upon the upper wall of the chamber. The polystyrene chamber walls are supported by lucite rings which are mounted in 23 cm diameter holes cut into the aluminum support plates. The upper aluminum plate moves on four precision machine screws having a screw pitch of 0.05 cm/revolution. Additional thicknesses of wall material can be added to the upper or lower chamber walls as desired. The plate separation is controlled in the control room during radiation

Two parallel chamber walls of any material are easily mounted on the aluminum frames. In this experiment the chamber walls are machined of polystyrene. The upper wall of the chamber consists of a polystyrene disc 14 cm in diameter and of variable thickness. The thickness used for the absolute measurements is 0.726 gm/cm². The inner surface of the upper wall, coated with a uniformly thin layer of colloidal graphite (Aquadag) serves as a movable high voltage electrode. The lower wall of the chamber, machined from polystyrene, is 2.54 cm thick. The chamber frame is so designed that any additional desired thickness of absorber may be added below this lower plate to insure maximum scatter contribution. Aquadag coated on the inner surface of the lower wall forms the collecting electrode and the guard ring. The collecting electrode diameter is 3.011 cm as measured by a traveling microscope to the center of a circular groove (0.006 cm wide) insulating the electrode from the guard ring.

EXTRAPOLATION CHAMBER MEASUREMENTS

Ionization currents were measured with the lower electrode of the chamber positioned at 75 cm from the source with its center on the central axis of the beam. The magnet, mentioned earlier, was positioned with its field perpendicular to the gamma ray beam. The collimated beam has a uniform intensity distribution over an area of about 7 cm diameter. The intensity falls off to about 1 per cent of the maximum beam intensity at about 20 cm diameter. A broad beam was used in order to compensate for the loss of ionization due to electrons escaping through the sides of the parallel plate chamber. Under broad beam conditions the loss of electrons is compensated for by electrons returning into the collecting volume from the irradiated guard ring region.

A null type electrometer circuit is used to measure the chamber current as a function of the revolutions counter reading. The electrometer circuit is calibrated against a standard current source that was previously calibrated at the National Bureau of Standards. A constant saturation field intensity of 10 volts/mm is maintained across the electrodes for all plate separations. For each plate separation two readings were taken; one with positive polarity on the high voltage electrode, the other with negative polarity. The average of the two readings is taken as the measure of the true ionization current.

The ionization currents are correlated with the roentgen as measured with the 25 roentgen Victoreen thimble mentioned earlier. A plot of the esu/roentgen as a function of plate separation as indicated by the revolutions counter reading is given in Fig. 4.

The ionization charge collected per roentgen per register reading is determined from a least-squares analysis of the experimental data. The variation of plate spacing based on measurements of the capacity of the parallel plates agreed with the register reading calibration. Combining the above ionization determination with the area of the collecting electrode, the ionization charge density with an upper plate thickness of 0.726 gm/cm^2 of polystyrene becomes 0 96 esu/cm^3 per roentgen with a standard deviation of ± 0.01 esu/cm^3-roentgen.

In another polystyrene extrapolation chamber exposed to the same gamma ray beam, the relative amount of ionization produced in acetylene and air was determined by Dr. Failla to be 1.31. This factor together with the absolute ionization in air value given above yields 1.26 ± .01 esu/cm^3 in acetylene per roentgen. The Bragg-Gray calculation of the energy absorbed in the surrounding polystyrene is greatly simplified since, aside from the polarization density effect, the wall and the gas have the same electronic stopping power. The value of the average energy dissipated per ion pair formed in acetylene relative to air is independently known (Failla[5]) to be 0.78.

ABSORBED DOSE ON THE BASIS OF CAVITY IONIZATION

The dependence of cavity ionization on the local absorbed radiation dose is related to the manner in which the radiation gives up its energy to electrons. Since all local absorbed dose rates are produced directly or indirectly by energetic electrons, it is important to concentrate on the problem of the actual electron spectrum. The method of calculating the total degraded electron spectrum and its application to absorbed dose and cavity ionization theory has been developed by Fano[6] and Spencer.[7, 8] An expression that uses the complete flux spectrum $n_{tot}(E)$ of the electrons is

$$D_W = \frac{1}{\rho_W}\int_{\Delta}^{E_{max}} n^W{}_{tot}(E)\cdot\frac{dE}{dx}\bigg|_{<\Delta}^{W}\cdot dE \qquad (5)$$

for the absorbed dose in Mev/gm. The letter w refers to the wall material. Then ρ_W is the wall density in gm/cm^3, $n_{W tot}(E)$ is the total electron flux in electrons/cm^2-sec-Mev and $dE/dx|^W{}_\Delta$ is the stopping power in Mev/cm for energy limit Δ. Δ is arbitrary and may have any value less than or equal to $E_{max}/2$.

When the total electron spectrum is used to get the ionization in a small cavity in the wall material, the energy limit acquires a natural significance. Then the ionization in esu/cm^3 is given by:

$$J_v{}^a = \frac{1}{W_a}\int_{\Delta}^{E_{max}} n^W{}_{tot}(E)\cdot\frac{dE}{dx}\bigg|_{<\Delta}^{a}\cdot dE \qquad (6)$$

where the letter a refers to the air in the cavity. Then the $\overline{W}$ is the average energy in Mev dissipated per creation of one esu of charge. The energy limit must be taken as the energy of those electrons whose probable range in the cavity is equal to the average dimension of the cavity. The case in which one or two of the dimensions of the cavity are excessively elongated, as in a parallel plate ionization chamber, may also be handled by calculating an average electron path length for an isotropic electron flux penetrating the cavity. For wall materials with an average

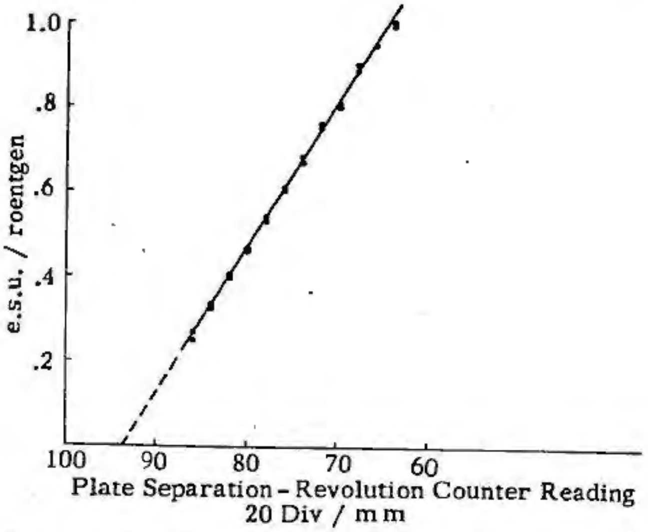

Figure 4. Plot of the extrapolation chamber ionization charge collected per roentgen as a function of the revolutions counter reading. The plate separation is obtained from the calibration of 20.0 divisions/mm. The roentgen is measured with a Victoreen thimble chamber calibrated at the National Bureau of Standards

atomic number close to the average atomic number of the cavity material, the dependence of D/J_v (ratio of absorbed dose to cavity gas ionization density) on cavity dimension will be small.

A calculation for 1.25 Mev gamma rays using Equations (5) and (6) gives $D/J_v = 98.8$ erg/gm per esu/cm^3 for a polystyrene wall and air cavity when the parallel plate spacing is about 0.2 mm. For a spacing of 0.6 mm the value of D/J_v is 98.6 erg/gm per esu/cm^3, neglecting leakage or finite beam effects.

For certain special wall materials and a limited range of gamma energies some of the difficulties associated with the interpretation of measurements to give D/J_v can be avoided. This occurs when a gas is available which has the same atomic composition as the wall material. Then, for photon energies which produce electrons for which the density (or polarization effect) in the wall material is negligible, the electron flux spectra in the wall and the gas of the same atomic composition are the same.[9]

An analysis of this special case shows that the ratio D/J_v is proportional to the ratio of the ionization currents for the gas to air, other things remaining constant, and the ionization chamber spacing does not appear explicitly in the formula. Using $\rho_g = 1.173 \times 10^{-3}$ gm/cm^3 at STP and $\overline{W}g = 26.6$ ev per ion pair for acetylene we get $D/J_v = 99.1$ for a polystyrene to air chamber. This agrees with the calculated value 98.8 above within experimental error.

ABSORBED DOSE ON THE BASIS OF INCIDENT QUANTITY OF RADIATION

With the gamma ray beam incident on the surface of an absorber, the absorbed dose D at any depth x is given by the relation:

$$D\ (x) = Q \cdot \overline{\mu}_a \cdot f\ (x)\ /\rho \qquad (7)$$

where Q is the quantity of radiation in ergs/cm^2 at $x = 0$, and ρ is the absorber density. The average value of that portion of the total absorption coefficient which corresponds to kinetic electron energy is $\overline{\mu}_a$ in cm^{-1} and is obtained by averaging over the gamma ray spectrum:

$$\overline{\mu}_a = \int \mu_a\ (E) \cdot Q\ (E)\ dE \ \Big/ \int Q\ (E)\ dE \qquad (8)$$

The factor $f(x)$ combines the effect of attenuation of the incident gamma rays to depth x and the effect of build-up due to scatter. $f(x)$ is evaluated by extrapolating the asymptotic portion of the central axis depth dose curve to the $x = 0$ point. The asymptotic curve is, in addition, displaced by an amount necessary to adjust for the forward scattering and range of the electrons set in motion by the gamma rays. This new curve represents the energy absorbed from the gamma ray beam at the point at which the absorption occurs, or at which the electrons are set in motion. An approximate method to obtain the displacement of the asymptotic depth dose curve when the beam is not too narrow is to integrate the central axis depth dose curve from 0 to an extrapolated infinity, and then to equate this to the area under the displaced curve which has been extrapolated to $x = 0$ as well.[10] The displacement is kept as an unknown parameter to be solved for and gives 0.97 mm in the present case. The factor $f(x)$ is then the ratio of the value of the displaced curve at $x = 0$ to the value at the depth in question. At the depth of the cavity (0.726 gm/cm^2), $f(x) = 0.986$.

The value of the average true absorption coefficient for polystyrene for cobalt-60 gamma rays is 0.0300 cm^{-1}. An estimation of the effect of self-absorption in the cobalt source using a calculated spectrum results in an increase of 1% in the coefficient to 0.303 cm^{-1}. However, this spectrum effect may be found experimentally to be greater, which would require a larger average coefficient. Using the calorimetrically determined quantity of radiation of 3370 erg/cm^2-r for the cobalt-60 gamma rays we get $D/r = 96.1$ ergs/gram at a depth of 0.726 gm/cm^2 in polystyrene per roentgen which corresponds to $D/J_v = 100$ erg/gm per esu/cm^3, which is again in reasonable agreement with the previous results.

CONCLUSION

It is demonstrated that it is technically feasible to determine precisely the quantity of radiation in the gamma ray beam from a cobalt-60 source. It is further demonstrated that the quantity of radiation combined with a depth dose curve yields the absorbed dose at any desired depth in an absorber. The absorbed dose so determined in a polystyrene absorber on the basis of a calorimetric measurement agrees closely with the absorbed dose calculated on the basis of cavity ionization. The use of the total degraded electron spectrum was illustrated in this latter calculation. The validity of the spectrum and stopping power components of this calculation was independently checked by measuring the ionization in acetylene gas in a cavity in polystyrene.

The utility of this calorimetric method as a primary standard suitable for the calibration of secondary standards is established. Its utility in the determination of absorbed dose is also exhibited. The validity of the Bragg-Gray cavity ionization relation employed with the total degraded electron spectrum is justified by comparison with the calorimetrically determined absorbed dose.

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Measurement of Tissue Dose as a Function of Specific Ionization

By H. H. Rossi and W. Rosenzweig,* USA

Results of radiobiological experimentations have made it appear that the biological effect of exposure to ionizing radiations is affected by two physical parameters: (*a*) the total energy delivered at the point of interest, and (*b*) the spatial distribution of this energy along the tracks of the ionizing particles. The first of these quantities is now measured in rads (multiples of 100 ergs/gm) and can be measured by a variety of methods, most of which are based on the well-known Bragg-Gray theorem. In particular, tissue equivalent ionization chambers of the type developed in this laboratory[1] make it possible to determine this quantity within a few per cent over a wide range of intensities.

The second quantity involved may be expressed in terms of linear energy transfer (LET, Zirkle[2]) for which kev/μ is a convenient unit. Another parameter which is more or less equivalent is specific ionization which can be expressed in terms of numbers of ion pairs per micron. In the present state of the art it is not clear whether energy loss and specific ionization are proportional to each other in a solid such as tissue, as the nature and energy of the ionizing particle is varied. However, it appears likely that any deviations from proportionality are not great.

Any differences in radiobiological effectiveness (RBE) of ionizing radiations are at present attributed to variations in LET or specific ionization. The connection between RBE and LET is, however, not simple and is rarely, if at all, considered one of strict proportionality. Consequently, the objective of physical dosimetry must be a detailed measurement of dose as a function of LET or specific ionization, rather than a mere determination of the average value of this parameter. In a first attempt at measurement of the specific ionization spectrum, we have developed an instrument which has been described elsewhere.[3] It is the purpose of this presentation to analyze certain aspects of the measurement which apply not only to the instrument itself, but also to some fundamental aspects of radiobiological experimentation.

Basically, the instrument is a proportional counter of spherical shape which is filled with a suitable counting gas (methane) at low pressure and has a wall made of conducting tissue equivalent plastic (material having the same atomic composition as tissue). This choice of the wall material results in a distribution of ionizing secondaries that is the same as that occurring in tissue. The ionization produced by these secondaries when traversing the cavity depends on both the specific ionization and the length of their trajectories. It is possible to allow for the latter effect on a statistical basis and consequently a proper analysis of the pulse-height spectrum produced by the counter yields a detailed spectrum giving dose as a function of specific ionization in the counting gas. This approach has a number of limitations. One of these derives from the fact that being a gas operated device, the response is comparatively slow and presently employed models must be operated in the range of the permissible limit of exposure to ionizing radiation. Another limitation arises from the assumption that particles traverse the cavity in rectilinear motion. Despite the low pressures employed, this requirement is strictly met only for positively charged nuclear fragments, since curvature of electron tracks remains a rather probable process. However, since most variations in RBE seem to occur after the LET has reached levels in excess of about 10 kev/μ (i.e., values which are mostly contributed by heavier particles) this restriction is not too serious.

Boag[4] has furnished theoretical LET distributions for protons ejected by fast neutrons traversing water. Since our device seems particularly suited for analysis of neutron exposure we have attempted to reproduce his data experimentally. The results are shown in Figs. 1, 2, and 3 for the Pu-Be spectrum, the Po-B spectrum and monoenergetic neutrons arising from the D-D reaction and having an energy of about 3 Mev. The solid curves represent experimental data and the dotted lines are based on Boag's theory which, in the case of the radioactive sources, has been suitably integrated. It will be seen that the general agreement is good, but it appears that the curves registered with our device are flatter than the theoretical curves. This is particularly evident in Fig. 3 where substantially monoenergetic neutrons are involved. We do not believe that this disagreement is entirely due to experi-

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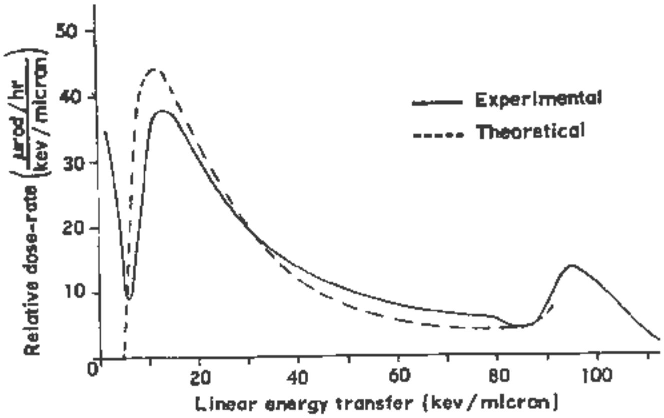

Figure 1. Dose distribution spectrum: Po-Be source

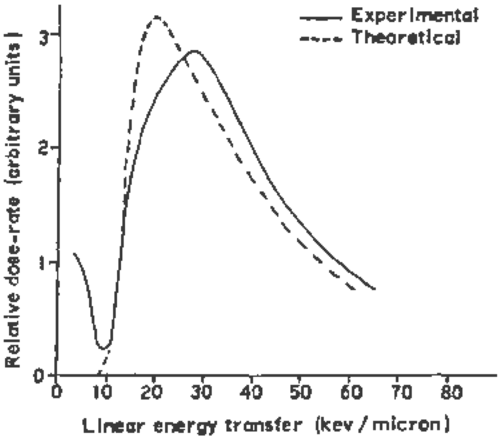

Figure 2. Dose distribution spectrum: Po-B source

mental shortcomings. In fact, checks on the linearity of the counter and of the electronic system employed have been satisfactory and, in addition, the total dose registered by the device is in agreement with an integral dose measurement performed with a tissue equivalent chamber after small corrections for contaminating electromagnetic radiation were applied.

The reason for the discrepancy is probably due to differences in the theoretical and practical aspects of the concept of LET. It must be realized that LET as normally considered is a statistical average over a number of discreet processes. Since the energy loss of ionizing particles is not continuous, its value when taken over a small interval will fluctuate about the assumed average value both because the number of interactions along a given length of track may vary and because the energy expended in successive interactions is also variable (Landau[5]). Consequently, identical particles of the same energy yield a distribution of LET. The width of this distribution becomes larger as the test interval chosen becomes smaller, since the degree of divergence will be larger for a smaller statistical sample. On the other hand, if the sample chosen becomes too large, the energy of the particle will vary within the interval resulting again in an increase of LET values. There must, thus, exist an optimal sample size for which the spread is least. The numerical evaluation of the optimal interval requires a detailed statistical analysis, but it is evident that its size will vary, depending on the nature and the energy of the particles involved.

It is to be noted that these considerations apply equally to the process of physical measurement as well as to radiobiological theory since the same effects will be operative in both the counter gas and biological structure traversed.

When pulse-height measurements are extended into the region of large values, it is evident that the main portion of the spectrum which is caused by protons is followed by a long band of pulses representing a very high degree of specific ionization. This is obviously due to carbon nuclei recoiling from the walls of the chamber. Since the yield of such particles is comparatively low and the pulse spectrum produced extends over a large range of pulse-heights, the counting rate per channel is quite low and with equipment and sources used thus far, the precision is not too great. In addition, the relation between energy loss and specific ionization for such particles is not too well known, hence the inclusion in an actual LET spectrum must be regarded as an approximation until further date can be obtained. Nevertheless, these particles which are of interest in radiobiology are clearly registered and an approximate value of the dose delivered can be obtained. However, this determination must be made in the lower range of pressures of counter operation since the range of these particles becomes comparable to counter dimensions at pressures of the order of 50 mm Hg. Thus it may be concluded from Fig. 4, which was obtained with a Po-Be source, that a dose contributed by these recoils is approximately 6% of the proton dose for LET up to 425 kev/μ. Since the counting rate beyond the corresponding pulse height is still finite an additional contribution must be delivered beyond this point. It is believed to be small.

The immediate practical use to which this instru-

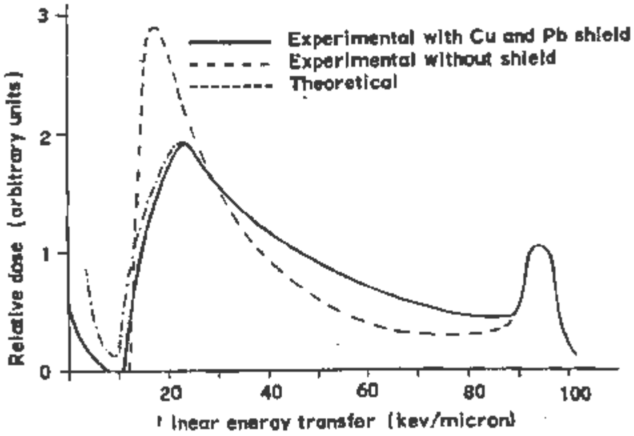

Figure 3. Dose distribution spectrum: monoenergetic neutrons from D-D reaction

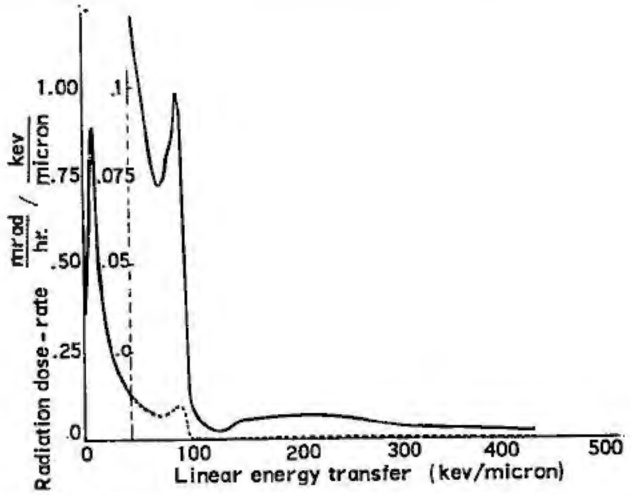

Figure 4. Dose distribution spectrum for Po-Be source showing contribution due to heavy recoils

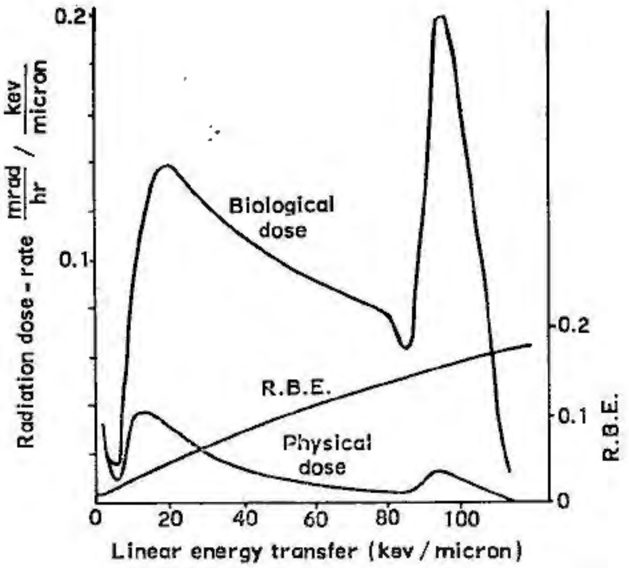

Figure 5. Radiation hazard due to Pu-Be source

ment has been put is one of health physics surveys around neutron sources where the energy distribution of the radiation received is very poorly known and an evaluation of the biological hazard consequently quite uncertain unless the LET distribution of secondaries produced in tissue is evaluated explicitly. The device is used to determine this quantity directly for particles heavier than electrons. Any dose that may be delivered by electrons arising from gamma or X-radiation is measured by subtraction of the total dose as registered by a tissue equivalent chamber from the heavy particle dose registered by the counter. The latter is simply equal to the area under the curves shown. Any evaluation of the actual health hazard may then be performed by multiplying doses delivered at various levels of LET by whatever RBE is assumed to be applicable. As an example, Fig. 5 shows an evaluation of the hazard from a Pu-Be source applying the recommendations of the National Committee on Radiation Protection.[6] Any other RBE values may be applied equally simply.

Efforts are under way to extend the operation of the device to higher intensities so that biologically effective doses or lower doses from pulsed generators may also be measured.

ACKNOWLEDGEMENT

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Portable Instruments for Beta-Ray Dosimetry

By W. C. Roesch and E. E. Donaldson,* USA

In circumstances where personnel are exposed to radioactive isotopes with little or no shielding, the main contribution to the dose rate is frequently from beta rays. At one cm from a one millicurie source emitting 1.0 Mev gamma rays the dose rate is 5.8 rad/hr;[1] for beta rays, it is 425 rad/hr. The majority of radioactive isotopes emit both beta and gamma rays in comparable numbers; near thick sources or shielded sources the gamma rays will be responsible for most of the dose rate but near point sources or thin sources, the beta-ray dose rate will predominate.

The dose from beta rays is received in the surface layers of the body and hence is subject to different maximum permissible limits than that for more penetrating radiation.[1] If the half-value layer in tissue (that thickness necessary to reduce the dosage rate to one-half its initial value) is one mm or less, the maximum permissible weekly dose in the skin is 1500 mrad. However, if it is greater than one mm, the permissible weekly dose is 600 mrad. The permissible dose decreases to 300 mrad for radiation penetrating to a depth of 5 cm. The 300 mrad limit will usually apply to gamma rays because of their greater penetration. Because of these different permissible limits, it is an advantage to be able to measure the surface dose rate separately from the dose rate to the deeper tissues.

Conventional survey meters designed for beta-ray measurements were found to be inadequate for most problems. Their response at moderate distances from point beta sources is the same as that of skin but they do not correctly measure the skin dose rate for extended sources. In practice the readings of the instruments must be multiplied by large correction factors and for routine operation the largest correction factor is used in all situations. In these situations the calculated dose rates may be several times as large as the true values. Another limitation arises because the large size of the instruments does not permit accurate measurement of the dose rate near sources of radiation, e.g., the dose rate to a hand in contact with or near a contaminated surface.

Experience in atomic energy plants has indicated the desirability of being able to measure beta-ray dose rates accurately in order to assure greater safety to the workers, and to permit them to work up to permissible doses when required. The following work was undertaken to provide simple but sufficiently accurate instruments for direct field measurement of beta radiation.

THE EXTRAPOLATION CHAMBER

An extrapolation chamber similar to that introduced by Failla[2] was used as a standard in all measurements of the beta-ray dose to tissue. The use of the extrapolation chamber is based upon the fact that the dose absorbed from beta radiation at any given point in a tissue-equivalent plastic is the same as the dose absorbed at a similar point in the original tissue. For purposes of beta-ray dosimetry a flat slab of lucite is taken to be representative of the tissue near and at the surface of the human body.

The problem is to measure the dose absorbed in the lucite exposed to beta radiation by determining the ionization per unit mass of the gas in a vanishingly small cavity in the plastic. It is necessary that the dimension of the cavity in the direction of the highest flux of beta particles be much less than the range of the beta particles in the gas so that no appreciable fraction of the electrons terminate their flight in the cavity. The extrapolation chamber provides the means of measuring the ionization under these conditions as well as assuring that the cavity has reached the size referred to as "vanishingly small" for the beta radiation under consideration. The most important features of the chamber are: that it be backed with a plastic slab of thickness greater than the range of the most energetic electrons present; that there be some provision for varying the spacing of the electrodes; and that there be some means for varying its depth in plastic from a few milligrams per square centimeter up to the range of the radiation considered.

The construction of the extrapolation chamber is shown in Fig. 1 in which the method of adjusting the electrode separation is demonstrated. The guard ring places a limit on the diameter of the chamber while the thickness of the front window or screen determines the effective depth at which the ionization is measured. In use, the chamber volume is varied by adjusting the electrode separation and the ionization current is plotted as a function of chamber volume. It is always found that as the separation is reduced, this curve approaches a straight line. The slope of this line which yields the ionization per unit volume for an infinitely thin chamber is employed in the calculation of the dose absorbed by the plastic.

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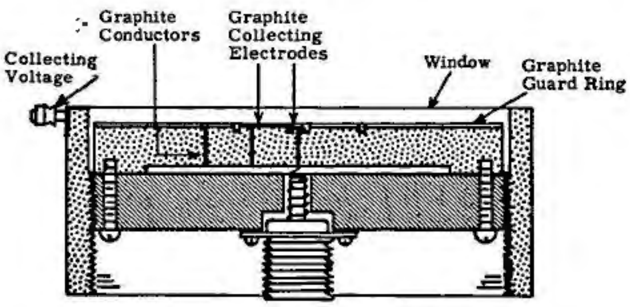

Figure 1. Section of the extrapolation chamber

Because the extrapolation chamber is used as a standard for beta dose measurement and because it is known that conventional survey instruments do not measure the dose from extensive sources correctly, it was necessary to investigate its directional response. In fact, this information is needed for judging the adequacy of any proposed survey instrument. The normalized angular response of the extrapolation chamber to a movable Sr^{90}-Y^{90} beta source is shown in Fig. 2. The ionization measured is uniform over a large solid angle about the normal to the chamber window.

The criteria to be used in judging any proposed survey instrument can be summarized as follows: (1) it shall have a small predictable dependence upon the energy of beta radiation and shall read beta dose, (2) it shall have no greater dependence on the energy of gamma radiation than other instruments and shall read gamma dose, and (3) it shall have nearly the same angular dependence as the extrapolation chamber.

USE OF MONOENERGETIC ELECTRON BEAMS

For the determination of the dependence of instrument response on beta-ray energy, monoenergetic electrons from a Van de Graaff accelerator were used as well as nuclear beta rays. Because uniform radiation fields of large extent are needed for comparison of the instruments, it was necessary to operate at distances of several meters from the exit port of the accelerator where the electrons were somewhat degraded from their initial energy. The effective energy in each case was determined from the range measured in lucite.

In order to obtain low dose rates from the accelerator, it was necessary to maintain the total beam current of the accelerator below 0.5 microamperes. Also, the background intensity of X-rays had to be reduced by directing the electron beam through a graphite tube past adjacent shielding material.

There is a difference in the depth-dose distribution for accelerator electrons and nuclear beta rays which results from the differences in their energy spectra.[8] Due to the presence of very low-energy electrons in a beta-ray spectrum, the depth dose from a nuclear beta source decreases sharply with increasing depth. For high-energy electrons from the Van de Graaff accelerator, however, the dose rate initially increases with increasing depth because of multiple scattering. After a depth of 500 mg/cm² is reached, the depth dose resembles that from a beta spectrum. Such effects must be taken into account when chamber response is being compared for the two sources of electrons.

THE MODIFIED CP SURVEY CHAMBER

The standard CP survey instrument has a deep cylindrical chamber with walls made of 3 mm thick phenolic plastic and a thin front window which is left uncovered for beta radiation and covered for gamma rays. It is known that the standard CP chamber has a strong directional effect for beta rays. The angular dependence of the standard CP was measured in the same way as it was for the extrapolation chamber and the result of the measurement is shown in Fig. 2. In practice the narrow angular response of the CP leads to large errors in the measurement of tissue dose resulting from exposure to extended sources.

It was believed that a shallow cylindrical chamber approximately 20 cm in diameter and 2.5 cm deep would satisfy the criteria stated earlier and at the same time would have approximately the volume of the standard CP. A test chamber was fabricated using 3 mm thick phenolic tubing 20 cm in diameter with a lucite base 1.3 cm thick and a 7 mg/cm² window to simulate the inert layer of skin. The depth of the chamber was made variable from 1 to 8 cm by a thread adjustment similar to that used in the extrapolation chamber. It was found that this chamber, when set at a depth of 2.2 cm, was satisfactory in all respects and possessed the same angular dependence as the extrapolation chamber.

The sensitivity of the modified CP chamber with a 7mg/cm² window was compared with the sensitivity of the extrapolation chamber with a 1 mg/cm² window by exposing the two instruments to electrons from the accelerator. As shown in Fig. 3, the relative sensitivity of the modified CP chamber falls off with decreasing electron energy. This fall-off can be explained by considering the absorption in the

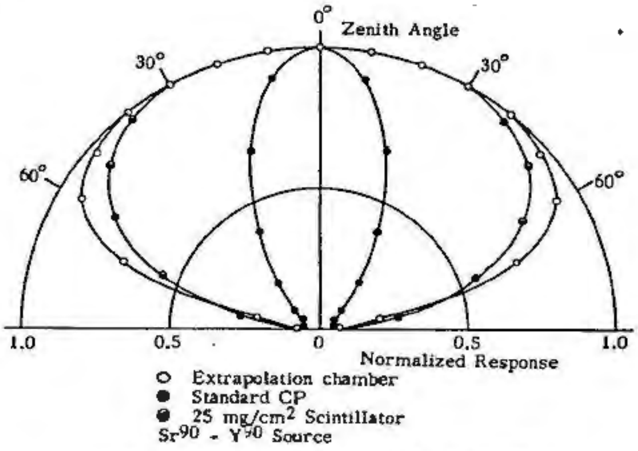

Figure 2. Angular dependence of the instruments

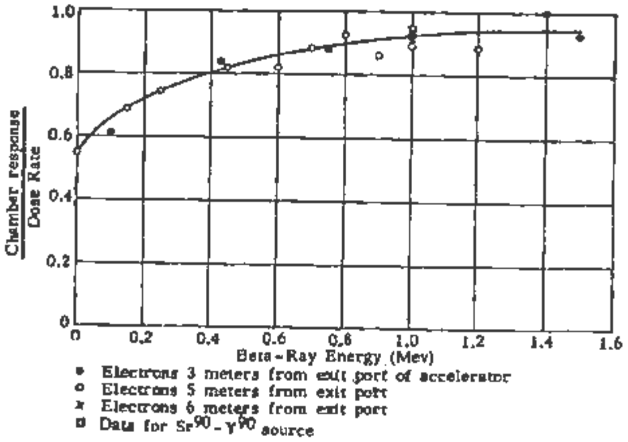

Figure 3. Chamber response vs beta-ray energy

7 mg/cm² window and is desirable from the standpoint of measuring the beta dose to skin.

The sensitivity of the modified chamber to photons with energy 0.01 to 1.25 Mev was measured and the results of the measurement are shown in Fig. 4. The sensitivity was normalized to a free-air ionization chamber for energies from 0.01 to 0.10 Mev and to thimble chambers for energies 0.146 to 0.197 Mev; the points at 0.80 and 1.25 Mev were obtained from standardized radium and cobalt sources. In Fig. 4 two sets of results are represented; one for a 7 mg/cm² window and the other for a 6 mm tissue-equivalent Bragg-Gray screen. The screen was of sufficient thickness to give equilibrium for electron production in the chamber walls and to remove those electrons produced in the source and in air by the primary radiation. The shape of the curve at low energies while undesirable is characteristic of most ionization instruments and seems unavoidable, but the response at high energies is quite satisfactory.

The instrument can be simply calibrated for field use by exposing it to a high-energy gamma source with the Bragg-Gray screen in place. Skin dose rate measurements in the field are taken with the screen removed. When used in this way, the instrument measures the total dose rate to surface tissues due to the non-penetrating beta radiation and it also measures the contribution due to penetrating gamma radiation which it reads about ten per cent too high. For measurement of the dose rate due to penetrating radiation, the chamber is used with the screen over the front window in the conventional manner.

THE THIN SCINTILLATOR SURVEY INSTRUMENT

The difficulties in making a large flat ion chamber with a thin but strong front window could be avoided if the detecting medium were a solid which could support the window. This suggested the use of a scintillation detector. Studies have been made of the use of anthracene crystals for gamma-ray dosimetry but little is known about their use for beta-ray measurements.[4, 5, 6] The present work was undertaken to determine the properties of scintillation detectors for the dosimetry of both beta and gamma radiation.

The scintillation material chosen for study was anthracene because it is approximately tissue-equivalent in composition and because it is the most efficient light emitter of the known organic scintillators. Because the beta particles are rapidly absorbed in matter, the scintillator must be thin, e.g., for 1.0 Mev beta rays it should be much less than 60 mg/cm² thick. Thin crystals could have been cut from large ones but they would have been quite fragile. It was found that machinable and reasonably strong scintillators could be made by grinding anthracene flakes to a fine powder, slurrying them with three times their volume of benzene, and molding the slurry at 12,000 lb/in² at 135°C for thirty minutes. The resulting tablets were faced on a lathe, mounted on glass disks with silicone stopcock grease, and machined to the desired thickness.

Scintillators as thin as 0.08 mm were made and this method was found to produce detectors with reproducible characteristics. Any method of mounting sufficiently thin layers of anthracene powder or flakes should produce adequate dosimeters. The considerable variation among the individual detectors, however, would make necessary a separate calibration procedure for each one.

A DuMont 6292 photomultiplier tube was used with a negative high voltage supply for these measurements. A high voltage gradient across the tube envelope and scintillation crystal will raise the background current and will eventually destroy the photocathode. To avoid these difficulties an aluminum light shield over the scintillator was kept at the negative potential of the cathode. To protect personnel from the high voltage a grounded shield covered the inner shield and was spaced about 3 mm from it by means of a thin lucite ring. The total thickness of aluminum amounted to 7 mg/cm². The anode currents were measured with a vibrating reed electrometer.

The response of scintillators of different thicknesses to a Sr^{90}-Y^{90} source was measured. The angular dependence of the thinnest scintillator (25 mg/cm²) is shown in Fig. 2 to approach that of the extrapolation chamber rather closely, while thicker scintillators have response curves approaching that of the CP. The area under these curves (multiplied by the sine of the angle) will determine the response to a large

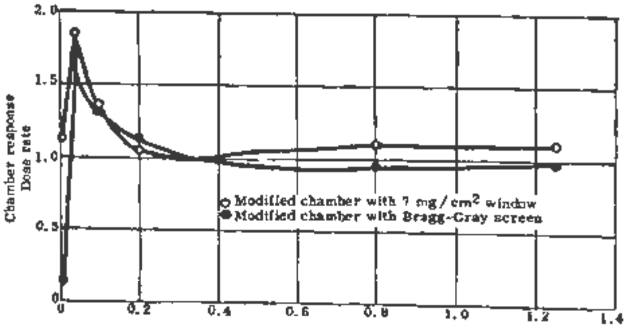

Figure 4. Chamber response vs photon energy

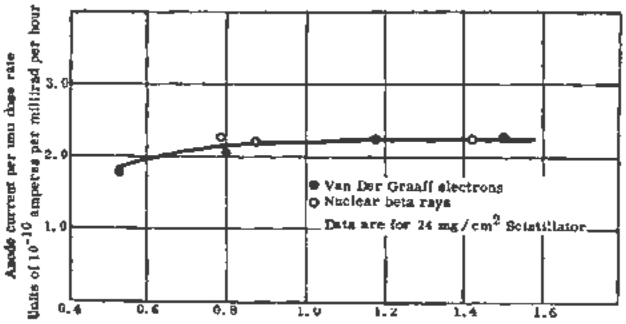

Figure 5. Scintillator response vs beta-ray energy

wall covered with a uniform thin layer of beta emitting material. In this case, the 25 mg/cm^2 scintillator would give a result differing by only 11 per cent from that of the extrapolation chamber.

A 24 mg/cm^2 scintillator was chosen for further study and its response to a Sr^{90}-Y^{90} source, to a P^{32} source, to a Ru^{103}-Ru^{106} source, and to electrons from the Van de Graaff accelerator was compared to the response of the extrapolation chamber. The results are shown in Fig. 5. The current per unit dose rate is independent of electron energy except at low energies where it decreases with decreasing electron energy.

The response of the same scintillator to gamma rays with energies between 0.0089 and 1.25 Mev is shown in Fig. 6. The gamma-ray dose rates were determined with a free-air ion chamber below 146 kev and with thimble chambers above 146 kev. The current per unit dose rate is independent of photon energy except that below 200 kev it undergoes a sharp decrease. This decrease is due to a saturation effect in the emission of light from anthracene by electrons of high specific ionization.[7] Two points are given for both radium and Co^{60} sources. The lower value in each case was obtained with a 6 mm lucite sheet in front of the thin window of the detector; this sheet was used at all energies down to 25 kev. With the lucite in front of the window, the gamma radiation and its secondary electrons are in equilibrium at the position of the scintillator, consequently, it functions as a Bragg-Gray detector. Without the lucite, secondary electrons formed in the source, in air, in surrounding objects, and in the detector can penetrate the thin window and produce light in the scintillator.

At a dose rate of 5 mrad/hr the 24 mg/cm^2 scintillation detector produces a current of 10^{-9} amperes which equals the photomultiplier dark current at the voltage (1175 volts) used. At higher tube voltage the signal to noise ratio increases so that detection levels of about one mrad/hr are easily obtainable.

SURVEY PRACTICE

Survey instruments of the type described here can be calibrated with radium or other gamma-ray sources of known emission in the same way as the usual gamma-ray instruments, provided that the thin front window is covered with a thick piece of the wall material so that the instrument responds as a Bragg-Gray detector.

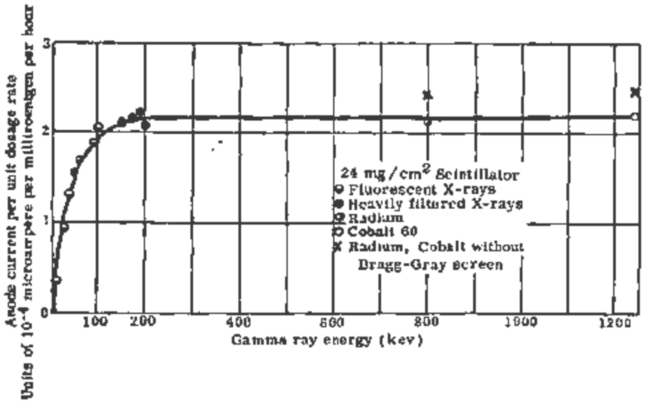

Figure 6. Scintillator response to photons

These instruments record the combined beta- and gamma-ray dose rates at the surface of the body. When different exposure limits are to be applied to the non-penetrating and penetrating components of the radiation these can be differentiated by means of absorbing screens. A 6 mm screen, for instance, will ordinarily eliminate all radiation of half-value layer about 1 mm or less; at the same time it will turn the chamber into a Bragg-Gray chamber for measurement of the more penetrating radiation. Radiation measured without the screen will be subject to the 1500 mrad/week limit; that measured with it, to 300 mrad/week.

CONCLUSIONS

Two instruments, a modified CP survey chamber and a scintillation survey detector, have been developed for use in the routine measurement of skin dose resulting from any combination of beta or gamma radiation. The results of the improvement in accuracy of dosimetry are most noticeable in cases of fields of radiation varying rapidly with distance and in cases involving extended sources. Work with radioactive materials may be done more efficiently because the accurate measurement of dose rates eliminates the need for large safety factors in exposure calculations.

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A High Level Dosimeter for the Detection of Beta and Gamma Radiation and Thermal Neutrons

By S. Dondes,* USA

In the study of the radiation chemistry of the nitrogen-oxygen system (to be published at a later date), a simple high level dosimeter for the detection of beta and gamma radiation and thermal neutrons was found in the use of nitrous oxide. The equipment for sample filling and sample analysis is not expensive and includes a small vacuum system and possibly a colorimeter. The physical properties of nitrous oxide as: ease of purification, small cross-section to thermal neutrons; indefinite shelf life; and usefulness in the temperature range of —80°C to 200°C add to its attractiveness. In addition, the products of decomposition, by beta and gamma irradiation, namely: nitrogen, oxygen and nitrogen dioxide are readily measured by vacuum techniques, and if desired, the nitrogen dioxide (in sufficient quantity), may be measured colorimetrically without opening the irradiated vessel. These products of decomposition do not react among themselves and may be measured at any time after irradiation.

To date, calibration of the nitrous oxide dosimeter has been made, using the Brookhaven National Laboratory Reactor, between 10^6 and 10^{10} roentgens, with the range 10^4 to 10^6 in process.

EXPERIMENTAL

Tank nitrous oxide (Matheson Co.) was purified, inserted and sealed in quartz vessels, 20 cm^3 (see Fig. 1), to a pressure of 650 mm. Where high doses were desired, 5 mg of uranium-235 oxide, was inserted as a powder prior to the addition of the gas. After sealing, the samples were irradiated at predetermined time intervals in the Brookhaven National Laboratory Reactor, operating at 24 megawatts and in a region where the neutron flux was 2.7×10^{12} neutrons/cm^2/second and the temperature about 70°C.

After irradiation, the seal was broken in vacuum and the nitrogen and oxygen measured after condensing the nitrous oxide remaining and the nitrogen dioxide formed in a liquid nitrogen bath. The ratio of the nitrogen and oxygen formed, was measured by burning the oxygen with hydrogen. Nitrous oxide remaining was measured at dry ice temperature, and the nitrogen dioxide at room temperature. A complete analysis takes about thirty minutes. In actual practice, the nitrogen and oxygen only need be measured at the low dosage and the nitrogen dioxide at the high. Visual inspection will determine which to use.

RESULTS

The production of nitrogen, oxygen and nitrogen dioxide with varying doses, is shown in Fig. 2. At low dose rates, the nitrogen dioxide is negligible and difficult to measure. The sum of nitrogen and oxygen formed is shown in the curve, as would be measured in normal practice. At high doses, between 3×10^7 and 3×10^9 roentgens, nitrogen dioxide is readily perceptible, and can be measured colorimetrically (using a suitable filter for the colored quartz), with the vessel unopened. A double check may be made by measuring the nitrogen and oxygen. Doses from

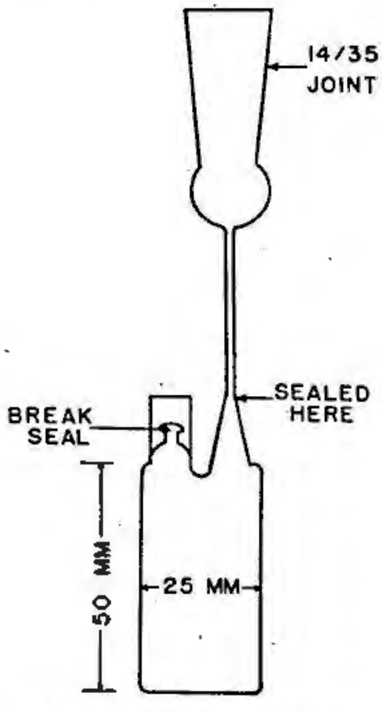

Figure 1. Quartz reaction vessel

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* Rensselaer Polytechnic Institute.

10^6 to 3×10^7 roentgens may be measured colorimetrically by increasing the length of the vessel from two to fifty inches.

At doses above 10^{10} roentgens, the decomposition of nitrous oxide approaches an equilibrium, as shown in Fig. 3, thus the dosimeter no longer is accurate, however, under controlled conditions, may be useful in this region.

DISCUSSION

The kinetics of the nitrous oxide decomposition by ionizing radiation is fairly simple in the initial stages, but becomes more complex, the latter not fully detailed here. However, the complete study of the oxides of nitrogen has been accomplished and will be published at a later date. From this study, we learned that nitrous oxide does not become attacked by nitrogen or oxygen atoms or any ion reactions and thus seems to be an ideal dosimeter. In the initial stages, the dosimeter is independent of intensity and temperature to a region of 3×10^7 roentgens.

The major reactions in the decomposition of nitrous oxide by ionizing radiation are as follows:

$$N_2O \longrightarrow \left| \quad N_2O^* \left\langle \begin{array}{l} \nearrow \; N_2 + O \\ \searrow \; N + NO \end{array} \right. \right. \qquad \text{I a}$$

$$N_2O \longrightarrow \left| \quad N_2O^+ + e^- \left\langle \begin{array}{l} \nearrow \; N_2 + O \\ \searrow \; N + NO \end{array} \right. \right. \qquad \text{I b}$$

$$2\,NO + O_2 = 2\,NO_2 \qquad \text{II}$$

$$NO_2 + O = NO + O_2 \qquad \text{III}$$

$$NO_2 + N \left| \begin{array}{ll} \nearrow \; 2\,NO & \text{IV a} \\ \longrightarrow \; N_2O \quad O & \text{IV b} \\ \searrow \; N_2 \quad O_2 & \text{IV c} \end{array} \right.$$

$$NO^+ + e^- = N + O \qquad \text{V}$$

In the region of 3×10^7 to 3×10^9, the dosimeter is slightly temperature and intensity dependent, where the curve has a slight bend (see Fig. 2). At intensities greater than 3×10^3 roentgens per second, and a total dose of over 10^8, the curve will be straightened since the nitric oxide becomes more decomposed by ion reactions, at higher intensities, and thus less nitrous oxide can be reformed. (Details will be presented in a later paper.) However, the primary decomposition of nitrous oxide will be exactly proportional to the dose.

With the decomposition of nitrous oxide by ionizing radiation being almost temperature independent, the lower temperature of usefulness is limited by the vapor pressure of the nitrous oxide, which at —81°C is one atmosphere and the higher temperature limited by the thermal decomposition of nitrous oxide. Thus, at 200°C, nitrous oxide is stable to thermal decomposition. For short time irradiations, temperatures above 200°C could be used. The thermal decomposition of nitrous oxide is a first order reaction and the constant of decay is given by the formula:

$$K = 4.2 \times 10^9 \, e^{-53{,}000/RT} \text{ sec} \qquad \text{VI}$$

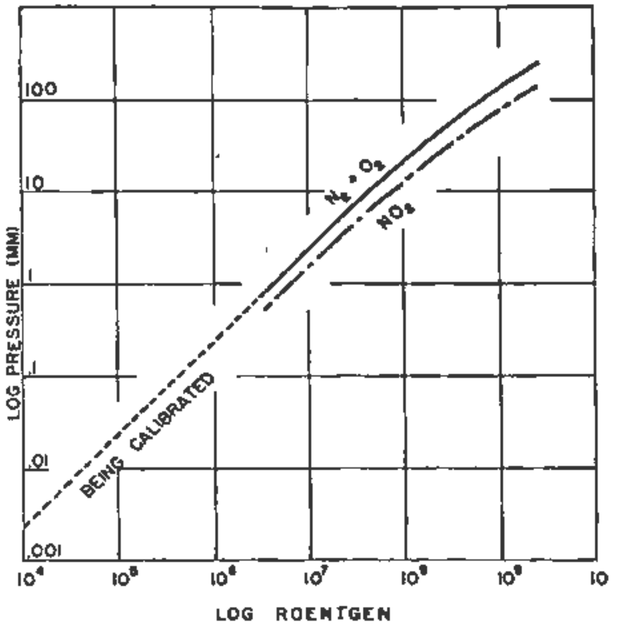

Figure 2. Calibration of N_2O dosimeter

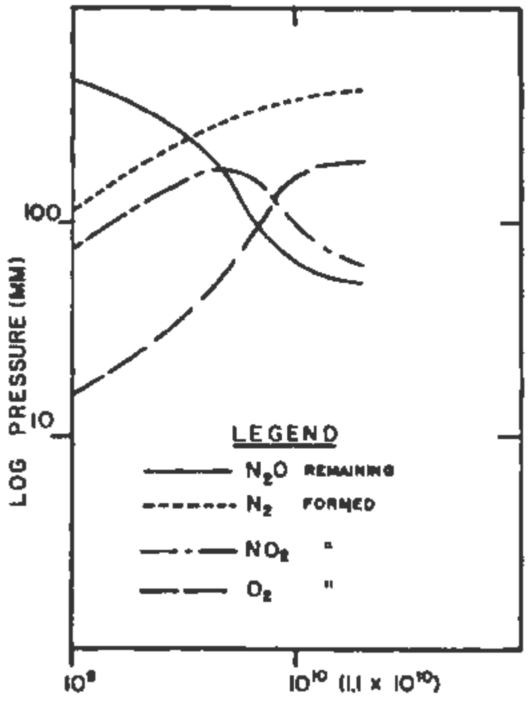

Figure 3. N_2O at equilibrium

At very low temperatures, from —40°C to —80°C, the dosimeter may read somewhat higher since the nitrogen dioxide may freeze out and the back reaction (IV b) will decrease.

The measurement of thermal neutrons was accomplished by adding 5 mg of uranium-235 oxide to the reaction vessel. Under these conditions, the decomposition due to the neutron induced fission was about one hundred times greater than that due to the beta and gamma radiation in the pile. The uranium-235 can be readily substituted with other materials such as boron, lithium or natural uranium.

Using pure uranium-238 and a cadmium shielding, fast neutrons, above 1 Mev, could also be measured. Experiments of this type are presently being studied. Since the beta particles do not penetrate the walls of the quartz vessels, the major measurement is gamma radiation. Minor corrections can be made of the beta radiation of the radioactive quartz and of the knock-on of fast neutrons on the nitrous oxide.

CONCLUSIONS

The dosimeter is designed for high intensity irradiations, such as: (*a*) determination of the dosage for enzyme deactivation in the region of 10^6 to 10^7 roentgens; (*b*) determination of the dosage for food sterilization in the region of about 3×10^5 to 1.6×10^6 roentgens; (*c*) on the lower limit, the determination of the dosage for pasteurization in the region of 10^4 to 10^5 roentgens; (*d*) determination of thermal and fast neutron flux within a pile; (*e*) dosage contours within a pile; and others requiring high dosage measurements.

It should be emphasized that this is an absolute dosimeter requiring no further calibration and only a simple vacuum system or a cheap colorimeter. The entire apparatus is inexpensive, rugged and simple to use.

ACKNOWLEDGEMENTS

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Use of the Fricke Ferrous Sulfate Dosimeter for Gamma-Ray Doses in the Range 4 to 40 kr

By J. Weiss, A. O. Allen and H. A. Schwarz,* USA

In the past few years, the increased use of high-intensity gamma-ray sources in various industrial and other research laboratories has emphasized the need for a good standard dosimeter. Although there are many methods available for determining these high dose rates, the Fricke ferrous sulfate dosimeter appears to be the easiest to use and reproduce.

A good dosimeter should be independent of intensity over a wide range and also independent of wave length, slight temperature changes and pressure variations. A brief investigation of ionization chambers and other non-chemical dosimeters will indicate their limitations in fulfilling these requirements.

A chemical dosimeter should have, in addition to the above mentioned requirements, the following characteristics:

1. It should be simple and convenient to use.
2. The extent of reaction due to irradiation should be easily and accurately measurable.
3. It should be reproducible and easily made from shelf reagents (insensitive to impurities).
4. The amount of reaction should not be strongly dependent on the concentration of the reagents.
5. The amount of reaction should be proportional to the dose.

Of all the chemical dosimeters in use at present, ferrous sulfate solutions appear to fulfill best the above requirements.

The history of the ferrous sulfate dosimeter begins in 1928, when Fricke and Morse[1] first proposed the use of $FeSO_4$ in $0.8N$ H_2SO_4 solution as a dosimeter solution for measuring X-ray doses from an X-ray machine. Since that time there have been several modifications and improvements in the basic method of using and analyzing these solutions. One important change was suggested in 1951 by Dewhurst,[2] who demonstrated that chloride ions would inhibit the oxidation of ferrous ion by certain organic impurities in the system. This modification eliminated the necessity of using triply distilled water and recrystallized ferrous sulfate in making up the dosimetric solutions.

Another contribution was made in 1952, when Hardwick[3] suggested that the irradiated solution of ferrous sulfate be analyzed for ferric ion concentration by direct reading of the absorbency at a wave length of 305 mμ.

In the present paper, we wish to indicate the ease with which this dosimeter may be used, even by untrained personnel. Information is presented on the reproducibility found by several individuals with varying degrees of experience, and on comparisons of dose rates measured on the same sources in different laboratories.

We recommend the following procedure for performing $FeSO_4$ dosimetry.

Dissolve:

2 gm $FeSO_4 \cdot 7H_2O$ or $Fe(NH_4)_2(SO_4)_2 \cdot 6H_2O$
0.3 gm NaCl
110 cm^3 concentrated (95–98%) H_2SO_4 (analytical reagent grade) in sufficient distilled water to make 5 liters of solution.

The sample containers are filled with this solution and placed in position where the radiation intensity is to be measured. The containers should be at least 8 mm ID, made of glass (or polystyrene), well cleaned and rinsed thoroughly with distilled water. The sample should be large enough to fill the spectrophotometer cell that is to be used. The duration of the radiation period should be accurately noted. The absorbency (optical density D) of the sample is compared with that of the unirradiated solution at 305 mμ in a spectrophotometer. For best results, the spectrophotometer should be thermostated, since the extinction coefficient has a rather large temperature coefficient of +0.7% per degree Centigrade.[4,5] At least, the temperature of the sample in the spectrophotometer should be noted.

The dose rate R is given by the formula

$$R\ (r/hr) = \frac{10^9}{\epsilon \cdot Y} \times \frac{D_{sample} - D_{blank}}{time}$$

where ϵ is the molar extinction coefficient and Y is the ferrous sulfate yield in units of micromoles per liter per 1000 r. In this laboratory, ϵ has been determined to be 2174 at 23.7°C. For best accuracy, ϵ should be determined on the spectrophotometer to be used. However, the extinction coefficients published by the various laboratories are within 3% of this value.

The size and shape of the dosimeter container is

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* Brookhaven National Laboratory.

not important as long as the internal diameter is greater than 8 mm. Weiss[6] originally reported that the ferrous oxidation rate increased markedly in cylindrical cells as the internal diameter fell below 8 mm. Ghormley[5] has confirmed this diameter effect, but found that the oxidation rate in 4 mm ID cells was only 3% greater than in the larger ones, a considerably smaller effect than was reported by Weiss. The solutions used in Weiss's work did not contain Cl^- and might have been affected by impurities. Repeating this work we have found a 6% greater yield in 4 mm ID cells. This effect is not noticed in polystyrene cells. Presumably it arises from the fact that more secondary electrons per unit volume are generated in glass than in water or in polystyrene.

The dosimeter is quite insensitive to changes in solute composition. Dainton and Sutton[7] have noted a decrease in the yield of the reaction below 10^{-4} M $FeSO_4$, and Ghormley and Hochanadel[8] have noted a decrease in the yield around 0.1 M $FeSO_4$. Ferrous sulfate and ferrous ammonium sulfate have been used interchangeably in this laboratory with no noticeable difference.

V. Hogan, in this laboratory, has determined the dependence of the yield on H_2SO_4 concentration. In $0.1N$ H_2SO_4 the oxidation rate was 3% lower than at the standard concentration ($0.8N$) and in $4N$ H_2SO_4 the yield was 7% greater. This approximately parallels the increase in electron density of the solution.

The NaCl is used to suppress the effect of organic impurities in the solution. In general, organic impurities tend to increase the Fe^{++} oxidation rate. The effect of varying the NaCl concentration is difficult to determine, since the effect is different with different impurities. However, chloride ion tends to catalyze the thermal air oxidation of $FeSO_4$ and thus becomes troublesome if present in concentrations greater than 10^{-2} M. Donaldson and Miller[9] have found that Cl^- does not inhibit the effect of unsaturated organic material, but fortunately most contaminants found in nature are saturated.

TABLE I. Effect of Different Water Sources on the Yield of the $FeSO_4$ Dosimeter

Source of water	*Oxidation rate (μM/min)* *With chloride*	*Without chloride*
Chlorinated well water	11.41	11.45
Distilled water*	11.47	11.74
Distilled water†	11.51	11.94
Distilled water‡	11.50	11.64

* Ordinary distilled water passed through aluminum pipes.
† Laboratory distilled water prepared in all glass system.
‡ Water distilled from basic permanganate and acid dichromate.

In Table I, the oxidation rates are given for solutions prepared with and without NaCl using various sources of water. It is seen that without added Cl^- most waters give a high oxidation rate. The exception is chlorinated well water. While this undoubtedly has the greatest impurity concentration, it also contains sufficient Cl^- to suppress the impurity effects. When 10^{-3} M Cl^- is added to these solutions, the yield drops to its normal value within experimental error. In practice, organic impurity effects can be found by following the oxidation rate as a function of time. In the presence of impurities, a plot of Fe^{++} oxidized vs dose will not be linear through the origin but will exhibit a positive intercept.

TABLE II. Reproducibility of Ferrous Sulfate Dosimeter Measurements by Individuals

Dosimeter measurements	*Average number of determinations per measurement*	*Average deviation from mean*	*Operator*
8	5	0.6%	H. Schwarz
2	12	0.6%	Jerome Weiss
14	4	0.7%	V. Allen
12	3	0.5%	J. Steimers

The ferrous sulfate dosimeter has proven itself to be highly reproducible when used by either experienced or inexperienced personnel. Table II lists the results of determinations made by four different individuals using various irradiation conditions. Neither of the two latter operators had ever used a spectrophotometer or the dosimeter before these series of determinations were made. Each was given approximately four hours of instruction and was closely supervised during the first day's operations. After that time, they were allowed to work by themselves except for occasional conferences to find out if they were having any difficulty.

Column 1 of Table II gives the number of different conditions used (dose rates, geometry, etc.). Column 2 lists the average number of determinations made for any given set of conditions, and column 3 lists the average deviation for all the determinations by each experimenter.

At Brookhaven, Co^{60} sources are prepared and shipped to various other laboratories in the country. Comparisons are therefore available between dose rates measured by $FeSO_4$ for the same source, first at Brookhaven by Brookhaven personnel and then at the purchaser's laboratory by their personnel. Some comparisons are shown in Table III. The results are normalized to the same date using 5.2 years for the half life of Co^{60}. Most of these experimenters had never used a ferrous sulfate dosimeter before these sources were obtained by their laboratories.

We have stated that the Fricke dosimeter is useful in the range of 4000 to 40,000 r. The lower limit is set by the analytical method. In a 1 cm absorption cell, the absorbency will increase by 0.137 which can be read to 1%. If a longer cell is used, the method can be extended to correspondingly lower doses. The upper limit is set by oxygen consumption in the sample. In Fig. 1, the Fe^{++} oxidation in 10^{-2} M $FeSO_4$ is shown. Above 40,000 r, the yield gradually decreases to the value observed in deaerated solutions. Presumably the upper limit can be extended to doses of about 200,000 r by saturating the solution with pure oxygen before irradiation (and increasing the $FeSO_4$ concentration fivefold).

The absolute value of the yield of the Fricke dosimeter has been measured by several methods independent of ionization chambers. One of the more recent values and probably the most precise was determined by Schuler and Allen[10] by measuring the energy input from a Van de Graaff generator. They determined G (ions of Fe^{++} oxidized per 100 ev energy absorbed) to be 15.45 ± 0.11. Calorimetric values obtained by Hochanadel and Ghormley[11] and by Burton, Lazo and Dewhurst,[12] and an internal beta radiation value determined by Donaldson and Miller[9] agree with this number within their respective stated accuracies. It can be calculated from the ion chamber work of Weiss and Bernstein[13] that the yield in terms of roentgens is 16.0 ± 0.3 μM/1000 r. Comparison of these two numbers involves the value of W (energy per ion pair) in air. Discussion of W values lies beyond the scope of the present paper.

The yield values referred to here were determined with 1 and 2 Mev electrons and with X-rays from a 2 Mev electrostatic generator. Nothing is known about the wave length dependence of this dosimeter to the precision which we have been discussing. However, according to theory, the yield should remain constant as the wave length is decreased from those employed here and should tend to decrease with increasing wave length at lower energies. Farmer, Rigg and Weiss,[12] using 250 kvp X-rays, compared the dosimeter to an ion chamber and found G for ferrous sulfate to be 16.4 ± 0.8. It would appear that the yield is constant up to this wave length. Presumably the yield may be taken as wave length independent for γ-ray applications.

TABLE III. Interlaboratory Comparison of $FeSO_4$ Dosimetry

Source location and experimenters	*Their dose rate (r/hr)*	*Our dose rate (r/hr)*	*%deviation*
Dow Chemical Co., Midland, Mich. W. K. Kramer	(*a*) 404,000	407,000	0.8
T. C. Engelder	(*b*) 123,500	124,000	0.4
Stanford Research Inst., Stanford, California H. J. Eding S. I. Taimuty	797,000	797,000	0.0
University of Chicago, Chicago, Illinois A. Zimmer	446,000	438,000	1.8
Massachusetts Institute of Technology, Cambridge, Massachusetts S. Davison	126,000	125,000	0.8
Columbia University New York, New York N. Barr	198,000	199,000	0.5

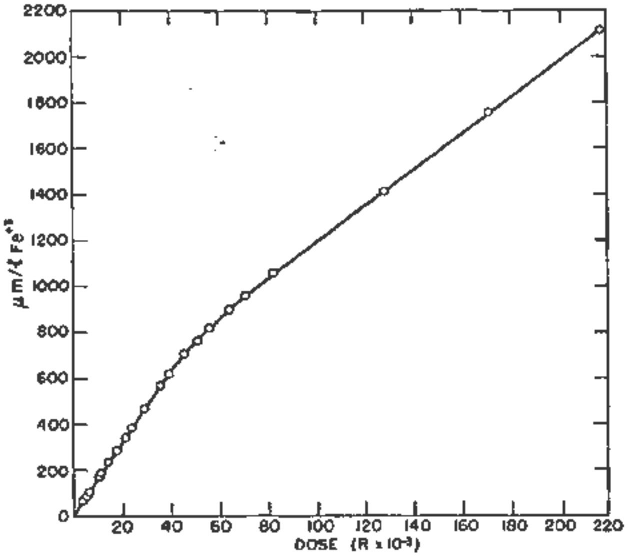

Figure 1. The oxidation of 0.01 *M* $FeSO_4$ in 0.8 *N* H_2SO_4

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The Role of Liquid Scintillators in Nuclear Medicine

By F. N. Hayes, Ernest C. Anderson, and Wright H. Langham,* USA

Liquid scintillation solutions[3, 12] share with other scintillating materials the property of converting energy which is deposited in them by ionizing radiations into visible light.[16] The unique advantages of the liquid scintillators are derived directly from their liquid state; all the other scintillating materials (some organic and inorganic crystals and certain special plastics) are solids. These advantages are two in number: first, the solvent power of the liquid scintillators for a great variety of materials, and second, the comparatively low cost of liquid scintillators of very large volumes and arbitrary shapes. This paper will discuss a few of the applications of these properties which have been made to biochemical problems. Since the field is a wide and very active one, attention will be limited to selected examples from the work of the Biomedical Research Group at Los Alamos and no attempt will be made to review the current literature. Many of the techniques discussed here are as yet unpublished, but will appear in due time in the appropriate journals.

PROPERTIES OF LIQUID SCINTILLATION SOLUTIONS

No pure liquid is known or expected to have the characteristics of a good scintillating material.[12] However, when certain organic compounds which are themselves scintillators in the pure crystalline state are dissolved in appropriate solvents, the resulting solution is a scintillator. These solutes, which are responsible for the actual light emission process, are linear chains of aromatic rings: terphenyl is the simplest example. The rings may be benzenoid or heterocyclic, (e.g., the oxazoles[3, 4, 7, 8]) and conjugated alkene chains sometimes occur (e.g., diphenyl hexatriene). The solute need only be present in low concentration; 5 grams per liter is the optimum for diphenyloxazole (PPO) in toluene. Therefore, most of the ionization energy is absorbed by the solvent, and very efficient energy transfer mechanisms must exist in order that the energy reach the solute for efficient light emission. This fact practically limits the acceptable solvents to alkyl benzenes.[3, 9, 13] However, it is possible to modify the solvent properties of the solvent by the addition of certain other liquids which decrease the light emission only moderately.[9] These liquids, known as "diluents," include aliphatic hydrocarbons, ethers, and alcohols. Finally, a secondary solute, itself a scintillator, may be added (in about 1% of the concentration of the primary) in order to shift the wavelength of the light emitted by the primary solute to longer wavelengths. This may be desirable for a number of reasons: better match to the response of the photocathode of the photomultiplier tube, or greater optical transparency of the solution at longer wavelengths.

The scintillation solution which we have found to be the best for most applications is 4 grams per liter diphenyloxazole (PPO) and 0.05 grams per liter 1,4-di[2-(5-phenyloxazolyl)]-benzene (POPOP) in toluene. This solution gives the largest pulse height and the longest wavelength of emission (4300 Å). The use of terphenyl in place of the PPO gives nearly the same pulse height but the solubility of the terphenyl is considerably less and it is more difficult to keep in solution.

The general composition of a liquid scintillation counter may, therefore, be: (1) the main solvent, whose primary function is the transfer of energy to the solute; (2) the primary solute, which absorbs energy from the solvent and emits a fraction of it as visible light; (3) the secondary solute which shifts the emission spectrum to longer wavelengths; (4) the diluent which modifies the solvent properties of the system without too great interference with the light emission process; and (5) photomultiplier tube(s) and associated electronic equipment to convert the light pulses into electrical pulses, to analyze their energy, and to record the results.

In only one respect are the liquid scintillators seriously inferior to the solid, inorganic crystals; namely, in their energy resolution. The light emission of a liquid scintillator is, within limits, proportional to the energy of the primary ionizing event, but the low atomic number of the organic compounds compared with NaI, for example, renders their response to gamma rays considerably inferior. The NaI crystal has a very high stopping power for gamma rays, and a large part of the cross section is due to the photoelectric effect in which the full energy of the gamma ray is deposited in the crystal. This gives rise to a sharp line, the "photopeak" in the energy spectrum. In the organic materials, on the other hand, the interaction with gamma rays is almost entirely Compton scattering, which results in a broad con-

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tinuous spectrum. For gamma ray spectrometry, the NaI crystal is always greatly superior, and only in special cases such as those discussed below is the liquid scintillator a serious competitor for gamma measurement. In the case of beta emitters, however, the initial spectrum is a broad continuum and good resolution is generally of little importance.

TRACER MEASUREMENTS WITH LIQUID SCINTILLATORS

Some of the nuclides which are most useful for tracer studies in biological systems are notoriously difficult to measure because they emit only soft beta rays and no gammas. Carbon-14 and sulfur-35 (average energy about 50 kev) calcium-45 (85 kev) and especially tritium (average energy 5 kev) are outstanding examples. Because these beta rays have very little penetrating power, efficient counting is possible only if they are part of the detecting system itself, so that self-absorption is eliminated. This has been possible for C^{14} and tritium with gas sample counters (both Geiger-Muller and proportional) but only at the expense of considerable time spent in chemical processing of the sample. Such processing necessarily destroys the chemical identity of the sample. One of the important advantages of the techniques to be discussed is that the sample can often be recovered unchanged for further study. A program has been underway for some time at the Los Alamos Laboratory to investigate the possibilities of taking advantage of the solvent powers of liquid scintillators to count tracer samples with a minimum of chemical preparation and to apply these techniques to biochemical studies. Some specific applications of the techniques developed are discussed in paper P/842.[14] The present discussion will be devoted principally to the instrumentation aspects of the problem.

In a good liquid scintillator, the low energy betas of the carbon-14 and tritium spectra produce on the average 350 and 40 photons per disintegration, respectively. With an efficient optical system for collecting these photons by a photomultiplier tube having a good Cs-Sb cathode, the average number of photoelectrons per disintegration will be 35 for carbon-14 and 4 for tritium. A representative photomultiplier (RCA 5819 or DuMont 6292) will have a thermionic emission background of the order of 100 per second at 4 equivalent electrons. It is clear, therefore, that the first step in counter design is the elimination of this large background.

The thermionic emission background can be reduced by selection of photomultipliers and refrigeration and can be eliminated by coincidence techniques. The most stable and sensitive instruments use all three, especially for tritium counting; for carbon-14 measurements, refrigeration is usually unnecessary. The coincidence technique as used at Los Alamos is briefly as follows:[11] two photomultiplier tubes receive the light from the scintillation solution. The pulses from each are independently amplified and those corresponding to a selected energy range are fed into a coincidence circuit. A count is recorded only if a pulse of an appropriate energy occurs simultaneously in both channels. The thermal noise pulses, being random and uncorrelated in the two tubes are, therefore, eliminated. This system necessitates a considerable number of electronic circuits: two fast amplifiers (rise time 0.1 μsec) with gains of from 1000 to 40,000 and very good overload characteristics, a pulse analyzer consisting of four Harwell discriminators using delay line clipping and providing independent upper and lower bounds in both channels, and a fast coincidence circuit with a resolving time of about 0.1 μsec. In addition a scaler, timer and high voltage supply are needed. While somewhat formidable in appearance, such an apparatus is practical; three units are in routine use in this laboratory and similar systems are commercially available.†

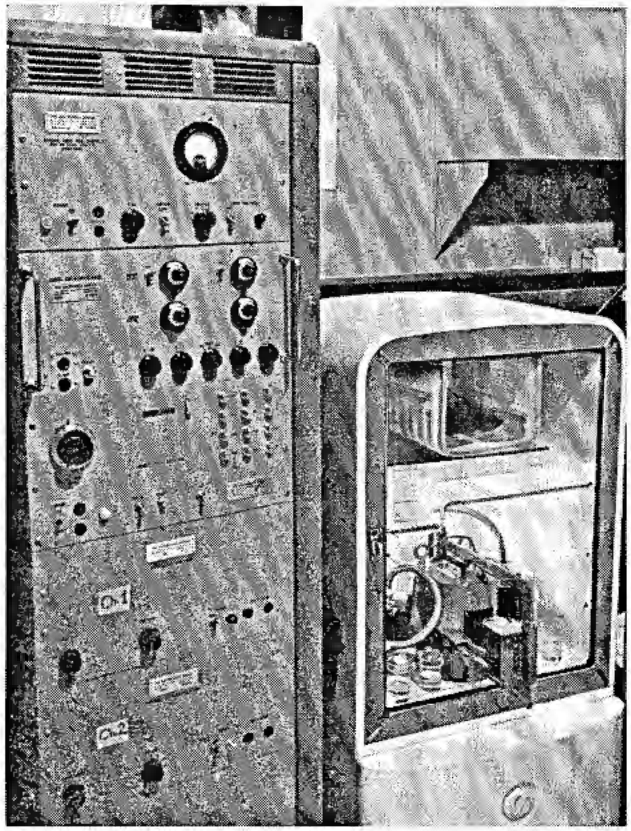
Figure 1. Liquid scintillation counter for carbon-14 and tritium

Figure 1 is a photograph of one of the Los Alamos units. The electronic circuits are contained in the relay rack at the left. At the right is the refrigerator containing the lead shield. The door of the shield is open, which shows the manner of introducing the sample bottle. When the shield door is closed, the sample is thereby carried into the shield to the photomultiplier tubes, which are at right angles to one another and which enter the shield from the rear.

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† From Packard Instrument Co., LaGrange, Illinois; and Technical Measurements Corp., New Haven, Connecticut.

Other sample bottles can be seen beside the shield.

A number of different techniques have evolved for measuring samples of different solubility characteristics. The simplest samples to handle are those which are soluble in a good scintillation solvent, usually toluene in our work. An extreme and unusual case occurs when the sample itself is actually such a solvent. An example is the study of the contemporary assay of natural radiocarbon being carried out by Hayes and described elsewhere.[5, 10] *p*-Cymene (*p*-isopropyl benzene) is easily prepared (by catalytic dehydrogenation) from a number of natural terpenes which are found in the wood, leaves, and fruits of many trees, shrubs and grasses. Therefore, it is possible to study the distribution of natural carbon-14 in such materials using only the simplest chemical processing. It is highly significant of the sensitivity of the liquid scintillator technique, that *p*-cymene, commonly prepared from "live" carbon sources, is practically useless as a primary solvent because its natural radiocarbon content raises its background by a factor of 8 above that of "dead" solvents in the ordinary counting apparatus.

The natural radiocarbon content of fermentation ethanol also has been measured by liquid scintillation counting.[1] Ethanol is not in itself a good scintillation solvent, but is a "diluent" which can be tolerated at concentrations up to 30% in toluene without too serious a loss of pulse height.

Substances of more conventional biochemical interest which are directly soluble in the scintillator include the sterols and the fatty acids. The sterols, particularly cholesterol and its relatives, have been studied by R. G. Gould of this laboratory using both single and double tracer techniques.[14] Because of the energy resolution possible, it is feasible to determine both tritium and carbon-14 in the same sample by proper adjustment of the energy gates. The two counts can be made simultaneously in some of the commercial models.

An additional advantage of the scintillation counting of steroids which should be pointed out is the tremendous range of sample sizes which can be accommodated. Gould has found that cholesterol samples from 0.01 mg to 1000 mg can be used without affecting the counter efficiency.

Sometimes samples which are themselves not soluble in toluene can be dissolved by the addition of an appropriate diluent. An example which has been extensively studied in the counting of tritium-labeled water by the addition of ethanol to the scintillator.[6] The technique has been applied to the determination of tritium water in blood, urine, perspiration and other body fluids. In some cases, a preliminary separation of water from the fluid is desirable; in most cases, the sample can be used directly. The sample is added to ten to twenty times its volume of absolute ethanol and the resulting alcohol-water mixture added to the scintillator solution in amounts up to 50% (the optimum for greatest sensitivity). This technique has been used in the study of body water volume, water turnover time, and the effect of irradiation upon the rate of uptake of water and upon the general water balance of the animal.

It has been observed that some inorganic salts dissolved in body fluids will remain in solution in the water-ethanol-toluene mixtures. It is, therefore, possible to count inorganic tracer ions by this technique. A simple preliminary treatment may be desirable, for example, if the sample is a concentrated, highly colored urine. By passing the sample through an anion-exchange resin the color is removed and anions, which are easily precipitated, such as phosphate, are replaced with chloride. In a representative example, 25-cm^3 samples of urine were poured into the resin columns (Dowex 1-X4, a quaternary ammonium styrene type, chloride form). Each sample was eluted with water; 40 cm^3 were collected beginning when chloride ion appeared in the eluate. In this case, the nuclide being studied was Cs-137 and 90% of the activity in the urine was found in this 40-ml fraction. This fraction was then added to 300 ml absolute ethanol and mixed with 600 ml of PPO-POPOP-toluene scintillator. Counting efficiency for the Cs-137 was observed to be over 100%. (Some of the counts came from the 2.6 minute Ba^{137m} daughter.)

Because of its wide applicability, the most promising counting method is one which is being currently studied by Hayes and Rogers. They have found that some materials which are insoluble in the scintillators can be suspended by grinding with absolute ethanol. When the resultant paste is added to a toluene scintillator, the dispersion is sufficiently fine that no sedimentation occurs during the counting period. Furthermore, the diameters of the particles are so small that no self-absorption of carbon-14 beta rays occurs. The suspension has surprisingly little effect upon the pulse height.

In a typical series of experiments, eight $BaC^{14}O_3$ suspensions were prepared using from 14 to 235 mg ground in 2.5-ml aliquots of absolute ethanol. The $BaC^{14}O_3$ was prepared from a quantitative Van Slyke oxidation of the Los Alamos standard benzoic acid.[5] Suspensions of cold $BaCO_3$ were made for background determinations. The scintillator used was 4 grams per liter of PPO and 0.05 grams per liter of POPOP in toluene. The counting efficiency (about 60%) for the $BaC^{14}O_3$ suspensions were determined, after which the standard C-14 benzoic acid was added to the suspensions and the efficiency for dissolved C^{14} determined. The ratio between the counting efficiency of suspended versus dissolved C^{14} was found to be 0.93 ± 0.3 over the entire concentration range. Absolute counting efficiency ranged from 63% for the dissolved benzoic acid with no suspension, to 58% for the dissolved benzoic acid in the presence of the most concentrated suspension. The

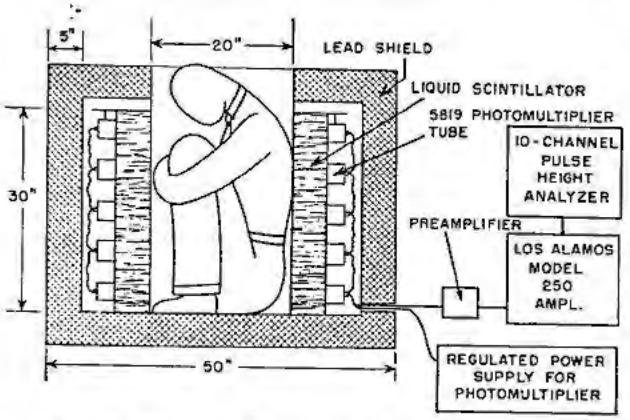

Figure 2. Schematic diagram of original human counter

small value of this decrease indicates that surprisingly little light was lost in spite of the completely milky appearance of the suspension.

The technique has also been tested using labeled phenylalanine, since amino acids are a very important class of compound for which no solvent has been found. Preliminary results indicate that the ratio of suspension to solution efficiency is close to 0.9 in this case also.

Finally, the possibility of counting tissue by this method is being studied. Various mouse tissues containing carbon-14 have been lyophilized, suspended and counted. The results so far are very promising and the efficiency is comparable to that obtained with $BaCO_3$ and phenylalanine.

LIQUID SCINTILLATORS AS LARGE 4 π DETECTORS

The second use of liquid scintillators which has been studied intensively at Los Alamos is their application to the determination of gamma emitting nuclides *in vivo*. While it was noted above that the poor energy resolution of liquid scintillator prevents it from competing with NaI crystals in gamma ray spectrometry, there is a type of gamma counting problem in which the choice between them is not obvious. This is the case in which it is desired to determine the concentration in an animal of a gamma emitter which is present at a very low concentration; for example, the determination of Ra in the human when its concentration is considerably less than the maximum permissible amount. While the NaI crystal counter can be set with great precision on the narrow photopeak of one of the Ra daughter gamma rays with a very low background, the limited size of the crystals available limits the detector to a very small solid angle. Long counting times are necessary and great stability is required of the apparatus. In the case of the liquid scintillator, however, it is quite feasible to construct a large "well-type" counter capable of containing a man and giving essentially 4 π geometry. While the background of such a detector is extremely large (of the order of 1000 per second), the counting efficiency is sufficiently high (about 15%) that one can measure as little as 10^{-9} curies of Ra equivalent in 15 minutes of counting time (the maximum permissible body burden of Ra is 10^{-7} curies). Because of the short counting period, no exceptional demands are made upon the stability of the electronics.

Several such detectors have been constructed at Los Alamos in the shape of hollow cylinders of various sizes. The liquid scintillator surrounding the hole is about 12 cm thick. While the two ends are left open, they constitute a fairly small fraction of the total solid angle, so that the geometry counting is essentially 4 π. A brief physical description of the detectors follows.

The original human counter is shown schematically in Fig. 2. Its dimensions were approximately 80 × 80 cm, and the central hole was 50 cm in diameter. It used 45 photomultiplier tubes (2-inch diameter cathodes) and 150 liters of solution. It was barely large enough to accommodate an adult human, and was, in fact, a temporary modification of one of the neutrino counters.[2] Results obtained with this instrument have already been reported.[15] A smaller version of very similar design has been used as a monkey counter. Its dimensions are 40 × 40 cm with a 20 cm diameter hole, and 32 photomultiplier tubes are used.

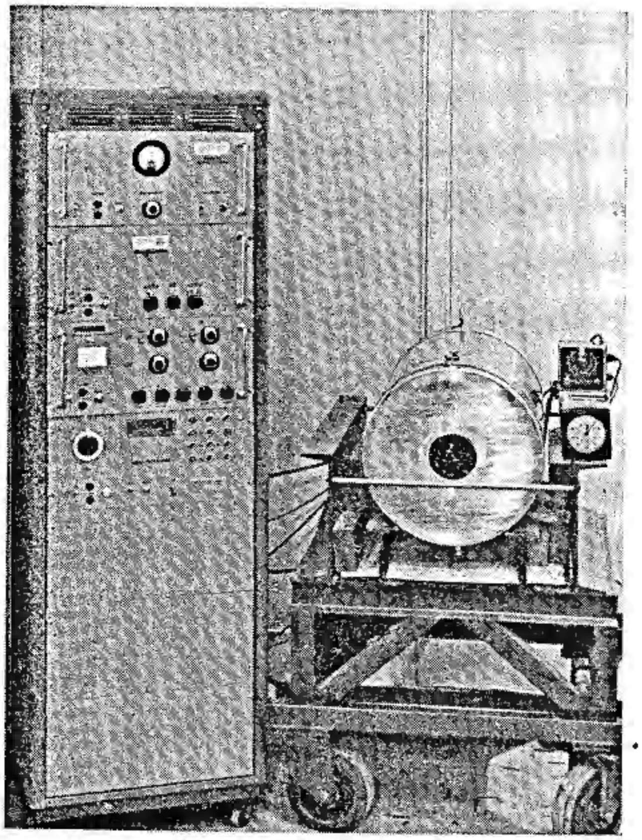

Figure 3. Small animal counter

A slightly different geometry is used in the dog counter (45 × 70 cm, 8 photomultipliers),[17] and the small animal counter (30 × 40 cm, 6 tubes) which is shown in Fig. 3. In these two detectors, the number

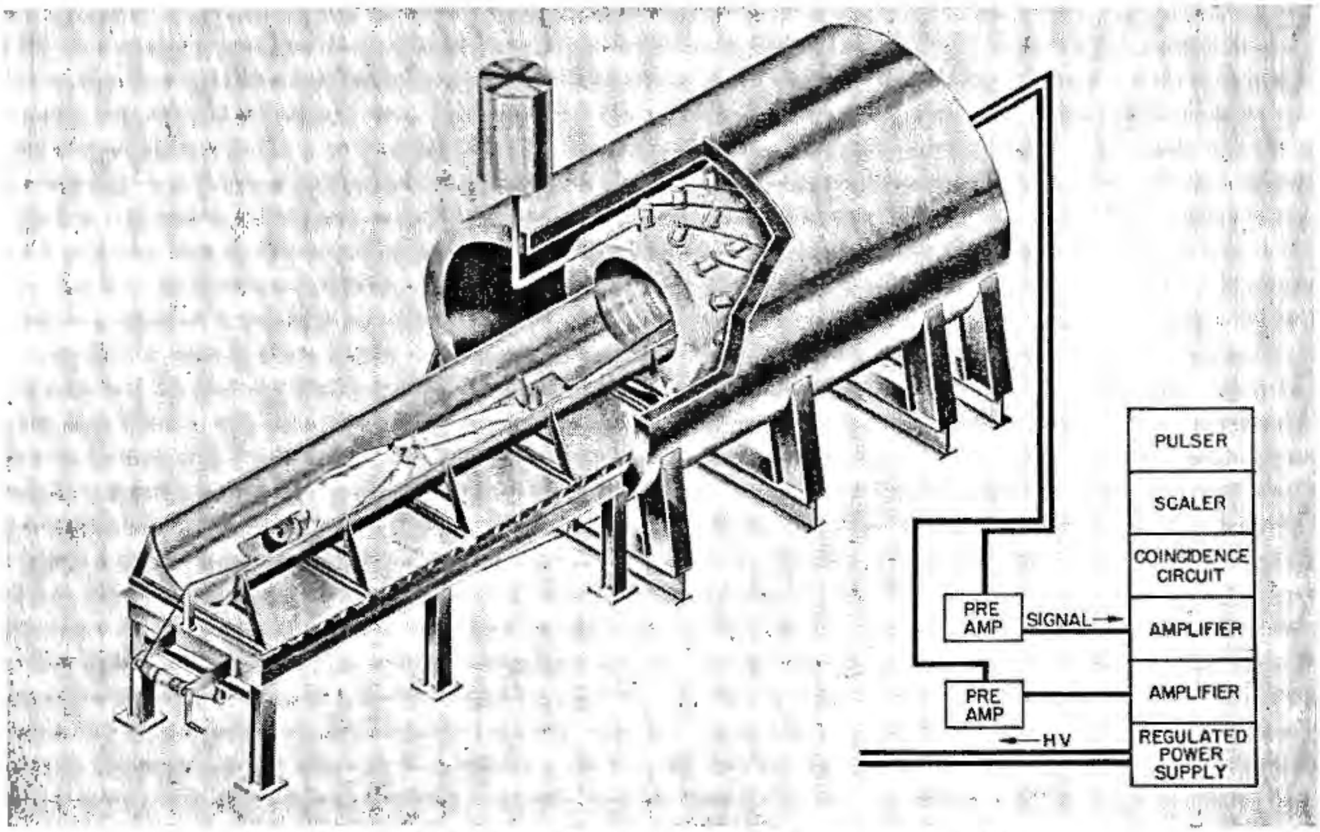

Figure 4. Scintillation detector for human subjects; new human counter

of photomultiplier tubes is greatly reduced and they are placed at the ends instead of the sides of the cylinder. The dog counter has four tubes at each end, while the small animal counter has all six tubes at the rear. In this arrangement, the energy resolution is considerably impaired, but in practice the counting efficiency and the sample to background ratio for gamma rays with energies above 1 Mev is not affected. The shielding around the small animal counter is a stainless steel annular tank filled with mercury, 3.5 cm thick. While the discriminator circuit shown in the relay rack is a double channel coincidence unit, this detector is usually operated with a single channel only.

An improved model of the human counter, twice the length of the original and using 108 photomultipliers, is now nearing completion. An artist's drawing is shown in Fig. 4. The tubes are on the sides and a large number are used since it is hoped to use this instrument for measuring very soft gamma rays (Bremsstrahlung) for which high optical efficiency is of the greatest importance. (It may be noted that in the drawing the human figure is rather too thin and gives an erroneous impression of the size of the detector. The central hole is only 46 cm in diameter, or barely large enough to accommodate the subject.)

Using these detectors, measurements have been made of the natural K^{40} level in human subjects, and tracer studies of Na^{24}, I^{131}, and Fe^{59} have been made on dogs, rats and monkeys. Van Dilla has used the dog counter in a study of the retention of Ra and Th isotopes and of Sr^{85} in the beagle. He has recently shown that Sr^{90} can also be detected by Bremsstrahlung, but the efficiency is low because of the continuous spectrum and low energy of this radiation. It is hoped to improve the Bremsstrahlung counting efficiency in a new model of the dog counter now under construction.

The basic problem in the detection of soft gamma rays is that of light collection; in order to obtain reasonable pulse heights for gamma rays down to energies of less than 100 kev, it is necessary that special precautions be taken to maximize the amount of light reaching the photocathodes. Since it is economically impossible to cover more than a few per cent of the total wall area with photomultiplier tubes, one must rely on repeated reflections of the light before it eventually reaches a phototube. This implies a highly reflective wall and an extremely transparent solution, since the photons may travel many meters before absorption. It has been shown[17] that for a somewhat simplified model of the geometry, the fraction of the total light reaching the photocathodes is given by $F/1-rt$ where F is the fraction of the wall area occupied by photocathodes, r is the reflectivity of the wall, and t is the transmission of the solution for the average light path between reflections. Clearly the difference between $rt = 0.90$ and $rt = 0.95$ is a factor of two in pulse height.

Special grades of anatase TiO_2 have been found the best material for the reflecting walls which are immersed in the scintillator. (MgO while intrinsically whiter is useless for internal coatings because of its low refractive index.) This pigment is used to prepare a special paint with an epoxy resin binder. The transparency of the solvent toluene is increased by treatment with activated alumina and by using a secondary solute to shift the emission spectrum as far as possible to longer wavelengths. The most satisfactory solution so far developed uses *p*-terphenyl as the primary solute and 1,4 di-[2-(5-phenyloxazolyl)]-benzene (POPOP) as the "wave-length shifter." The wave length of maximum emission of this solution is 4300 Å, compared with 3550 Å for terphenyl alone.

Triethylbenzene has been used as the solvent in some of these detectors. While the pulse height is less than that from toluene, it has the advantage of being less of a health and fire hazard.

The electronic circuits used in conjunction with such a detector have few special requirements. A coincidence unit is not essential at high energies, since connecting all the photomultiplier anodes in parallel in effect performs this task. However, some of the pulses obtained from these tanks are extremely large, up to 150 Mev. Therefore, the overload requirements of the amplifier are rather extreme.

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Liquid Scintillation Counting of Natural Radiocarbon

By F. N. Hayes,* Ernest C. Anderson,* and James R. Arnold,† USA

Professor W. F. Libby's interest in low-level counting techniques and their development led to the discovery of natural radiocarbon[2] and its application to dating.[11, 12] The outstanding success of radiocarbon dating in illuminating many problems in archaeology and geology is now well known, and the techniques promise to have many other applications in the uses of C^{14} and H^3 (both natural and synthetic) in tracer studies. For these reasons, there is considerable interest in the techniques themselves and in their improvement. While the original discovery of natural C^{14} was made with gas counters, Libby's dating method is based on the counting of solid samples of elemental carbon inside a special Geiger-Muller counter called the "screen-wall" counter. The counter is surrounded by a ring of conventional Geiger-Muller counters used in anti-coincidence with it for the reduction of background. The majority of the dates so far determined have been measured with this procedure, but it does have several shortcomings which offer grounds for improvement. The two principal weaknesses are the low counting efficiency (about 5%) and the susceptibility to contamination by airborne radioactivities, which result from the large surface area of the solid carbon. The low counting efficiency is partly compensated for by the large sample used, but this in turn constitutes an additional difficulty in those cases in which only small amounts of sample are available.

COMPARISON OF GAS SAMPLE WITH SCINTILLATION COUNTING

Two improved counting methods have been proposed and worked out in some detail: (1) gas sample counting, using a proportional counter filled with acetylene,[8, 23] carbon dioxide,[5, 9, 25] or methane[6] prepared from the sample; and (2) liquid scintillation counting[3, 4, 17, 21] in which the sample is converted into a suitable solvent or diluent for a scintillation solution.

Approximately it may be said that the gas counter uses samples of smaller size than the conventional screen-wall method (8 grams) and obtains its improvement in sensitivity by the greatly increased counting efficiency. The liquid scintillation technique, on the other hand, shows up to best advantage when sample size is greatly increased; its use of liquid samples with high densities makes practical the use of very large samples. Not only is the counter more compact, subtending a smaller solid angle to extraneous radiations, but the mass of shielding is greatly reduced and pulse height discrimination can be used to eliminate mesons and much of the gamma ray background. Both gas sample and scintillation counting effectively avoid the contamination problem by the nature of the samples and ease of avoiding their exposure to the atmosphere. The gas sample method has been reduced to practice by several laboratories and a large number of dates obtained by acetylene counting have been published by Suess. The liquid scintillation method, largely because of difficulties in the synthesis of the sample material, has so far been used chiefly for measurements of the contemporary assay and for studies of synthetic "aged" samples.

Table I presents a comparison of some of the counting methods which are either in use or in a fairly advanced stage of development. In addition to the basic characteristics of the method (amount of sample required, contemporary rate and background), some calculated capabilities are given. These are first, in column 5, the statistical precision (one standard deviation) theoretically obtainable in 48 hours of counting time, assuming that the background must be determined along with each sample. [This assumption is not necessarily true for all the methods; e.g., the double screen-wall[1] permits simultaneous measurement of background.] It is further assumed that counting time is divided between sample and background in the most efficient manner (that is, in the ratio of the square roots of the counting rates).[13] The statistical precision has not been given in terms of the corresponding error in the age because in such terms it rapidly becomes unrealistic (8 years for example for method 11) because of other factors influencing the precision. For example, it is not *a priori* obvious that sufficient stability can be obtained in the electronic circuitry to maintain a precision of a few tenths per cent. That such stability exists must be specifically proved. The best reproducibility so far obtained by Arnold is 0.3%, analysis of the data indicating the error at this level to be indeed purely statistical, if appropriate precautions are taken. Isotopic fractionation, in nature and in sample preparation, mixing of the biosphere with the rest of the natural radiocarbon "reservoir," contamination and similar effects will

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also become very prominent in any attempts to extend the precision. In brief, adequate counting statistics are a necessary but not a sufficient condition to guarantee a corresponding precision in dating.

The counting time for the measurement of a contemporary sample to an error in age of 100 years (1.25% error in activity at 1σ) is given in the sixth column and is another possible basis of comparison. Whether this counting time can be approached as a limiting time for a determination depends on the complexity of sample preparation. If it is possible to duplicate the facilities for sample preparation, which are usually much less expensive than the counting apparatus, some advantage can be taken of the potential reduction in counting time.

The last column gives the theoretical maximum age limit of the method. The same limitations which were mentioned in connection with column 5 apply to these numbers.

One might predict that the simpler chemistry and electronics and the smaller sample requirement of the gas counting method will recommend its use for the average dating application, and that the scintillation method may find principal application in the precision study of the contemporary assay and in the dating of extremely old samples. The problems to be investigated by radiocarbon dating are so diverse that several methods will undoubtedly be required for their solution.

INSTRUMENTATION FOR SCINTILLATION COUNTING

The liquid scintillation method is based on the ability of certain solutions to convert a part of the energy deposited in them by ionizing radiations into visible light.[24] The conversion is relatively inefficient so that only a small number of photons are produced (less than 10 per kev). The photon production is, however, proportional to the energy deposited so that the magnitude of the light pulse is (within limits) proportional to the energy of the initial event.

For highly efficient counting of soft beta rays, the sample is made an integral part of the scintillating solution,[14, 15] so that the entire energy of every beta ray is available for light excitation. The main solvent is usually an alkyl benzene and the solute (which is responsible for the light emission) is a compound consisting of aromatic rings linked in a linear array (the simplest example is *p*-terphenyl). A photomultiplier tube is used to detect the photons and to amplify the pulse by a large factor (of the order of 10^6). The pulses are further amplified by a conventional linear amplifier (gain of 10^3 to 10^4) and those of the desired energy range are selected by a pulse analyzer. Since the average energy of the C^{14} beta is about 50 kev, the energy range must extend down to 5–10 kev to obtain good efficiency. These pulses may correspond to the emission of only a single electron from the cathode of the photomultiplier tube. Under these conditions, one encounters an undesirable characteristic of the photomultipliers, their "dark current." Even in the absence of any true signal, thermionic emission from the photosensitive cathode produces a large number of "noise" pulses (of the order of 10 per second). These are eliminated by the following method:

Two photomultiplier tubes (e.g., Dumont 6292) with their associated circuits receive the light from the scintillation cell, and after energy selection only those pulses which occur simultaneously (within 1 μsec or less) are passed by a fast coincidence circuit. Since the thermal noise pulses are random and uncorrelated in the two tubes, they are rejected in this process. Refrigeration is sometimes used to reduce thermionic emission somewhat and to make the task of the coincidence circuit easier. (An alternative approach to the problem has been adopted by Pringle *et al.*[21] Through careful selection of the photomultiplier tube and close attention to the optics of the system, they are able to reduce tube noise so that single channel operation is possible.)

The resulting electronic apparatus is somewhat more complex than the rather similar circuits used in gas proportional counting. The principal difference is the need for two amplifiers and analyzers instead

TABLE I. Comparison of Counting Methods

Method	*Size of sample (grams)*	*Contemporary rate (cpm)*	*Back-ground (cpm)*	*Contemp-orary precision**	*Count time 100-yr. error†*	*Maximum age, (yrs.)‡*
Libby screen-wall[12]	8	6.7	4.0	1.28%	50 hr	25,000
Suess acetylene 1 liter, 1 atm[22]	1	10.7	2.3	0.78	19 hr	31,000
Crathorn, acetylene 3 liters, 1 atm[8]	3	39	16	0.47	7 hr	34,000
de Vries, CO_2 0.3 liter, 3 atm[25]	0.5	5.6	2.6	1.29	51 hr	25,000
Brannon, CO_2 0.7 liter, 10 atm[5]	6	45	13	0.40	5 hr	36,000
Fergusson, CO_2 8 liters, 3 atm[9]	13	103	14	0.23	1.6 hr	42,000
Pringle, $\cdot C^*H_3$ 20 ml[21]	2.3	15.5	3.3	0.67	13 hr	33,000
Pringle, methanol in toluene, 20 ml[21]	1.9	14.2	6.2	0.80	20 hr	29,000
Audric, acetylene in toluene, 80 ml[4]	7	45	40	0.57	10 hr	31,000
Arnold, hexane, octane in toluene, 100 ml[3]	47	182	26	0.18	1 hr	44,000
Hayes, cymene from terpenes, 90 ml[17]	67	470	60	0.11	22 min	48,000

* Theoretical minimum possible error (1σ), statistics only, for contemporary sample.

† 1σ for contemporary sample = 100 years, 48 hours counting time; sample and background both unknown.

‡ Assuming 4σ is limit; 24 hours each on sample and on background.

of one and the addition of the coincidence unit. If the ultimate precision indicated in Table I for the scintillation methods is to be attained, great stability in counter characteristics is essential. This is best achieved by making the counting rate as nearly independent as possible of all variations in the electronic and optical parameters of the system. This is accomplished to a great degree by employing "balance point" operation.[3]

CHEMISTRY OF SAMPLE PREPARATION

The main difficulty retarding the application of the scintillation methods to actual dating is the complexity of the chemical syntheses required to convert the sample to a suitable form for counting. The carbon of the sample must be converted in good yield to an organic liquid which is either itself an efficient solvent for the scintillation system, almost necessarily an alkyl benzene,[16] or compatible with an efficient solvent with a minimum of interference with light emission, such as an aliphatic hydrocarbon, an alcohol, or an ether. This second class of liquids can be referred to as "diluents." A third class of organic liquids comprises those compounds which interfere by "quenching" the light emission process (e.g., CS_2, amines, ketones, etc.) or by absorbing the light (colored substances). Members of this third class are clearly unsuitable.

Methods of synthesis which have been proposed and studied experimentally are shown in Table II. (The equations are not balanced, since in many cases the stoichiometry is unknown.) These include examples of syntheses of both solvents and diluents.

TABLE II. Synthetic Methods for Scintillation Dating

1. Partial synthesis of toluene[21]
    - $C^*O_2 + \phi MgBr = \phi C^*O_2MgBr$
    - $\phi C^*O_2MgBr + HCl = \phi C^*O_2H$
    - $\phi C^*O_2H + LiAlH_4 = \phi C^*H_2OH$
    - $\phi C^*H_2OH + Br_2 = \phi C^*H_2Br$
    - $\phi C^*H_2Br + LiAlH_4 = \phi C^*H_3$
        - Yield: 50%
        - Sample carbon in scintillator: 13%
        - Counting efficiency: 50%
2. Synthesis of methanol[7, 20, 21]
    - $C^*O_2 + LiAlH_4 = LiAl(OC^*H_3)_4$
    - $LiAl(OC^*H_3)_4 + ROH = C^*H_3OH$
        - Yield: 50–90%
        - Sample carbon in scintillator: 11%
        - Counting efficiency: 30%
3. Synthesis of acetylene[4, 8]
    - $C^*O_2 + Li = Li_2C^*_2$
    - $Li_2C^*_2 + H_2O = C^*_2H_2$
        - Yield: 65%
        - Sample carbon in scintillator: 10%
        - Counting efficiency: 45%
4. Synthesis of hexane and octane[3, 19, 23]
    - $C^*O_2 + SrCl_2 = SrC^*O_3$
    - $SrC^*O_3 + Mg = SrC^*_2$
    - $SrC^*_2 + H_2O = C^*_2H_2$
    - $C^*_2H_2 + Cu_2Cl_2 = Cu_2Cl_2 \cdot x(C^*_2H_2)$
    - $Cu_2Cl_2 \cdot x(C^*_2H_2) = Cu_2Cl_2 \cdot (C^*_2H_2)_x$
    - $Cu_2Cl_2 \cdot (C^*_2H_2)_x = Cu_2Cl_2 + C^*_6H_6 + C^*_8H_8$
    - $C^*_6H_6 + C^*_8H_8 + H_2 + Pt = C^*_6H_{14} + C^*_8H_{18}$
        - Yield: 30%
        - Sample carbon in scintillator: 50%
        - Counting efficiency: 25%

Note: In this table, C* designates carbon from the sample.

Obviously, the total synthesis of a good solvent is the more desirable in that the major portion of the solution is then made up of the sample and no losses in pulse height need be incurred. However, the difficulty of the total synthesis of benzenoid hydrocarbons from CO_2 is such that one is led to consider alternatives in the hope of simplifying the chemistry. One of these is the partial synthesis of a suitable solvent, e.g., preparation of toluene from sample CO_2 and inactive phenylmagnesium bromide.

In this case, an excellent solvent is obtained but only the methyl group is derived from the carbon of the sample and hence the counting solution is 13% by weight sample. This technique has been used by Pringle and his colleagues.[21]‡ A somewhat shorter synthesis, proceeding through ethyl benzoate directly to toluene was used by Blau (unpublished). The difficulty with this type of method is that the overall counting efficiency is not much better than that obtainable with methods involving much simpler syntheses.

Several examples of diluent synthesis have been worked out. In addition to the toluene synthesis, Pringle *et al.*[21] have also used W. G. Brown's simple and elegant reduction of CO_2 to methanol with $LiAlH_4$.[7, 20] This synthesis gives a diluent containing 40% by weight sample carbon. Unfortunately, only about 25% methanol can be incorporated into the system without undue loss of pulse height, so that only 10% of the system is sample carbon.

Audric and Long[4] have used a synthesis of acetylene already developed for gas counting.[8] The acetylene can be dissolved at low temperature in a solvent system (toluene-ethanol) to an extent of about 12% by weight sample carbon. This is comparable to the amount in the preceding two examples.

Arnold has combined a slightly different acetylene synthesis as used by Suess[23] with a synthesis discovered by Nieuwland[19] leading to a mixture of hexane and octane via copper acetylide. The mixture of aliphatic hydrocarbons obtained can be added to the scintillation system in concentrations as high as 60%, which is equivalent to about 50% by weight sample carbon. The synthesis is complex and the yields are about 30%, but it has been proved by careful C^{13} measurements that isotopic fractionation in the synthesis is very slight. This synthesis is being used on a routine basis and preliminary results are given below.

Hayes has investigated the possibility of by-passing the difficulty of synthesis by suitable selection of the sample. *p*-Cymene (*p*-isopropyltoluene), an excellent scintillation solvent, can be easily prepared from a large number of natural terpenes. "Dead" cymene for background measurement is prepared from petro-

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‡ We are grateful to Professor Pringle for allowing us to quote his results in advance of publication.

chemical propylene and toluene. The terpenes are widely distributed in essential oils found in the wood, leaves, and fruits of trees, shrubs and even grasses. Some of these oils are commercially available under circumstances which permit the determination of the locale and time of harvest. A study of the contemporary assay based on this principle has been begun and some results are given in the next section.

RESULTS§

The early measurements of Anderson[12] on the C^{14} assay of contemporary wood showed an essential constancy regardless of geographic origin within the precision of the screen-wall counter method. Small deviations can arise, however, from a number of effects, such as isotopic fractionation, incomplete mixing of the "exchange reservoir", and the introduction of "fossil" carbon into the atmosphere by the combustion of coal, oil and gas (the Suess effect).

Suess has reported[22, 23] a decrease of a few per cent in the C^{14} assay during the twentieth century, presumably due to the last named effect. There is some evidence that this effect is local, being much more marked in industrial regions than elsewhere. Arnold has, therefore, measured the C^{14} content of annular sections of a tree from Lake Geneva, Wisconsin (about 50 miles northwest of Chicago), and of one from the Peruvian Amazon by the scintillation method. The results are shown in Table III. As expected, the Peruvian assay is higher and the Wisconsin specimen shows a steady rise with increasing age of sample. The depression of the activity of the contemporary Wisconsin sample is about 4%, and even the 50-year old Wisconsin sample is slightly below the Peruvian level.

TABLE III.* Measurements of the Suess Effect

Sample no.	*Source of sample*	*Rings deposited*	*C^{14} assay, dpm/gm*
CS-1	Lake Geneva, Wisconsin	1933–1953	13.00 ± 0.04
CS-2	Lake Geneva, Wisconsin	1913–1933	13.15 ± 0.06
CS-3	Lake Geneva, Wisconsin	1890–1913	13.32 ± 0.04
CS-4	Peruvian Amazon	1934–1954	13.62 ± 0.05

* Results of J. R. Arnold.

An interesting example of the application of natural radiocarbon measurements to a rather important and different type of problem is the study of the origin of "smog" and haze over cities. By measurement of the C^{14} assay of carbon obtained by filtration of large quantities of air, it can be learned what fraction originated from biological sources (such as pollen or burning of garbage and waste paper) and what fraction came from fossil fuels (such as gasoline and coal). Measurements by Arnold of samples collected by F. A. Patty and G. Clayton showed that 25% of the carbon in Los Angeles smog is of living origin while in Detroit smog, the figure is about 12%. Large-scale tagging of individual sources of solid particles is practical and would make possible a more detailed analysis.

TABLE IV.* Specific Activity of p-Cymene

Source of the cymene	*C^{14} Assay,† dpm/gm*
Eastman Kodak, "White Label"	13.5 ± 0.1
	14.0 ± 0.1
Newport Industries (55-gal. drum)	13.5 ± 0.1
	13.4 ± 0.2
Synthetic camphor	13.8 ± 0.1
Average‡	13.6 ± 0.2

* Results of F. N. Hayes.
† Error is standard deviation of counting statistics.
‡ Error is estimated from scatter of individual measurements.

Results obtained by Hayes for the absolute specific activity of *p*-cymene are given in Table IV. Three samples of *p*-cymene were measured: Eastman Kodak "White Label", Newport Industries commercial (55-gallon drum sample), and material prepared by D. G. Ott of Los Alamos from synthetic camphor by the P_2O_5 distillation method.[10] The initial source of all three materials was ascertained to be Florida gum turpentine. Gum turpentine is derived from the exudate of living trees and it is, therefore, contemporary, although the exact year of collection was not established. ("Wood" turpentine, on the other hand, is derived from the distillation of old stumps and is of uncertain age.) The commercial cymene is prepared from pinene from turpentine by isomerization and dehydrogenation. Synthetic camphor is also synthesized with pinene as the starting material by a rather complicated series of reactions. The yield in the laboratory conversion of camphor to cymene is only 50%, so that isotopic fractionation is a danger. However, the agreement of the activity of this product with that of the other cymenes indicates there is little, if any, such effect. Natural camphor samples from harvest of various years are, therefore, being collected for comparison, as well as sample of terpenes from other sources.

Considerable care was taken in the determination of counter efficiency. A sample of $BaCO_3$ containing about 16% C^{14} was converted to CO_2 and divided into four well-mixed aliquots. Three of these aliquots were submitted for independent mass-spectrometric analysis of their C^{14} content. Excellent agreement was obtained among the three determinations. The fourth aliquot was converted into benzoic acid (yield 95%). A portion of the benzoic acid was converted to CO_2 and again analyzed for C^{14} mass-spectrometrically. The result was in agreement with the original CO_2 assay.¶ One mg of the benzoic acid was dissolved in one liter of toluene along with 100 mg of inactive benzoic acid. From this stock solution, 1 ml aliquots (determined gravimetrically

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§ Since the results of Arnold and Hayes were obtained by different methods in different laboratories, they are reported separately.

¶ We are indebted to D. L. Williams of Los Alamos for this C^{14} standard. A detailed discussion of the standard will be published soon by him.

not volumetrically) were added to the counting solutions to determine the counter efficiency. The disintegration rate was calculated using 5570 years as the C^{14} half-life. It is believed that the overall accuracy of the calibration is good to 1% or better. An aliquot of the benzoic acid standard in toluene solution was used by Arnold to determine the efficiency of his counter, so that the results in Tables III and IV do not constitute completely independent determinations of the absolute assay. However, the samples were quite different and the chemical processing was very dissimilar so that the observed agreement of the Peruvian and Florida samples is very gratifying. It indicates that there are no large chemical errors (fractionation, contamination, etc.) in either procedure. We, therefore, believe the error in these numbers to be less than 3% on an absolute scale. The small Suess effect observed in the Florida cymenes is evidence for the local nature of large depressions of the activity such as that found near Chicago.

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Medical Scintillation Spectrometer

By J. E. Francis and P. R. Bell,* USA

Scintillation counting of radio iodine and other gamma emitting substances in biological work has always been extremely difficult with equipment commercially available due to the inclusion of scattered radiation in the measurements. The inclusion of scattered radiation can easily lead to an error of a factor of two or greater making precision dosimetry very difficult.

This new instrument is a scintillation spectrometer which measures the spectrum of gamma radiation falling on it. By selecting the full energy peak, scattered radiation will not be detected. The unit permits accurate thyroid uptake measurements, assay of material to be used, detection of radioactive impurities, the measurement of samples for tracer chemistry, and high contrast scanning of distributions of activity in tissue.

The instrument contains a complete linear amplifier, a single-channel analyzer, a count integrator together with a preset timer, and the high voltage supply for the photomultiplier. It is relatively inexpensive and compact being contained in one thirteen by seventeen inch chassis.

Two special collimators have been designed for use with the spectrometer. The first accepts radiation only from an arc of 30 degrees in front of the scintillation crystal and very little in the back direction. The second is designed for measuring the distribution of activity in various body organs.

CIRCUIT

Figure 1 is a schematic diagram of the photomultiplier, preamplifier and linear amplifier. Radiation falling on the detector crystal causes the emission of light photons which pass through the shield crystal and are collected on the photocathode of the phototube. These photons cause the emission of electrons which are multiplied in the tube. By taking the signal from the last dynode a positive step pulse is obtained. This positive pulse is placed on the grid of a cathode follower (*V*-1) which drives a cable carrying the signal to the input of the amplifier. The step pulse from the cathode of *V*-1 is fed into a 1500 ohm delay line through its characteristic impedance to obtain a pulse one microsecond in duration. The differentiated signal is placed on the input grid of the first feedback loop (*V*-2, *V*-3) of the linear amplifier. A portion of the signal is fed through a network to the cathode of *V*-2 to compensate for the dc resistance and complex frequency response of the delay line. The first feedback loop has a gain of approximately seventy. A step potentiometer provides gain control over a range of sixteen to one.

V-4 and *V*-5 comprise the output group which has a gain of one hundred and twenty. The output pulses vary from zero to one hundred volts.

The pulses from the amplifier are fed into the single-channel analyzer which is shown in Fig. 2. The analyzer contains two discriminators (*V*-6, *V*-7) whose discrimination level can be varied from a few volts to ninety volts by means of a multiturn potentiometer labeled *E*. The two discriminators are biased a fixed distance apart determined by the setting of a second potentiometer labeled Δ*E*.

The discriminators work in the following way. Normally when there is no signal into the lower discriminator (*V*-7), the cathodes of *V*-7 are held at approximately ninety volts by the bias on the grid of side *B*. No current is flowing in side *A*, and the grid of *V*-7*A* is negative with respect to the cathode by a voltage determined by the setting of the potentiometer *E*. In order to get an output signal from the plate of *V*-7*A* a signal from the amplifier must be large enough to raise the grid of *V*-7*A* high enough to transfer the current from *V*-7*B* to *V*-7*A*. In order to sharpen this transfer point a crystal diode is connected from the plate of *V*-7*A* to the junction of *R*-39 and *R*-40. This effectively lowers the plate resistance of *V*-7*A* to the forward resistance of the diode until *V*-7*A* is drawing approximately 1.3 milliamperes and is in a region of higher gain. When the tube draws more than 1.3 milliamperes, a back voltage is developed on the crystal diode and its resistance goes from a few hundred ohms to several thousand ohms. The increase of the impedance at this point causes a rapid transfer of current from *V*-7*B* to *V*-7A. The upper discriminator *V*-6 operates in the same manner.

For a small signal lying below the value determined by the setting *E* neither discriminator is triggered, and there is no output pulse. When an input signal is large enough to trigger only the lower discriminator, the plate of *V*-7*A* goes negative and charges the ten μμF coupling condenser (*C*-20) which goes to the second grid of the anti-coincidence tube *V*-8. When the input signal goes down below the trigger level, the plate of *V*-7*A* returns to its original voltage, and a positive signal is produced at the second grid of the anti-coincidence tube *V*-8

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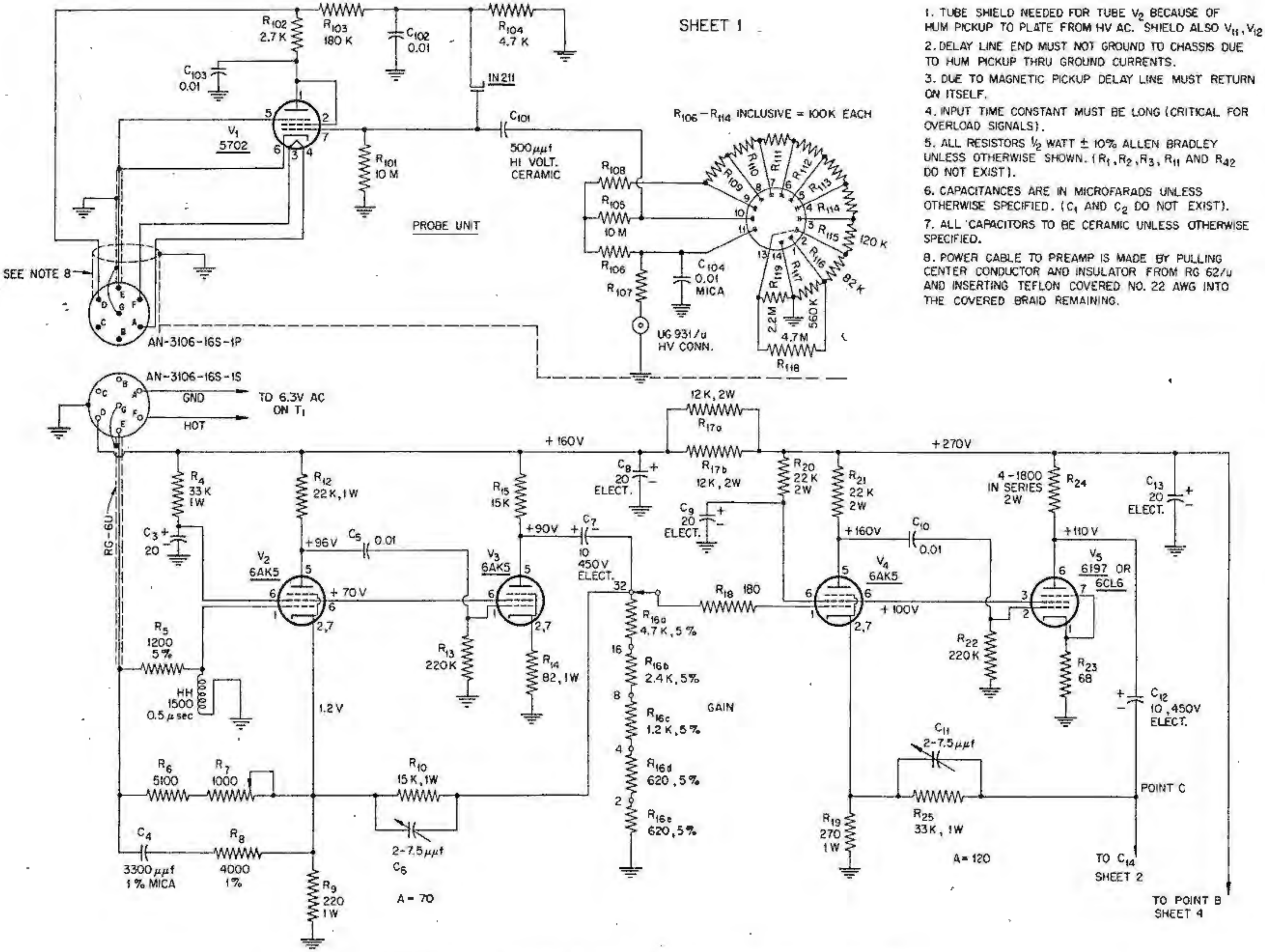

1. TUBE SHIELD NEEDED FOR TUBE V_2 BECAUSE OF HUM PICKUP TO PLATE FROM HV AC. SHIELD ALSO V_{11}, V_{12}.
2. DELAY LINE END MUST NOT GROUND TO CHASSIS DUE TO HUM PICKUP THRU GROUND CURRENTS.
3. DUE TO MAGNETIC PICKUP DELAY LINE MUST RETURN ON ITSELF.
4. INPUT TIME CONSTANT MUST BE LONG (CRITICAL FOR OVERLOAD SIGNALS).
5. ALL RESISTORS ½ WATT ± 10% ALLEN BRADLEY UNLESS OTHERWISE SHOWN. (R_1, R_2, R_3, R_{11} AND R_{42} DO NOT EXIST).
6. CAPACITANCES ARE IN MICROFARADS UNLESS OTHERWISE SPECIFIED. (C_1 AND C_2 DO NOT EXIST).
7. ALL CAPACITORS TO BE CERAMIC UNLESS OTHERWISE SPECIFIED.
8. POWER CABLE TO PREAMP IS MADE BY PULLING CENTER CONDUCTOR AND INSULATOR FROM RG 62/u AND INSERTING TEFLON COVERED NO. 22 AWG INTO THE COVERED BRAID REMAINING.

Figure 1. Schematic diagram of photomultiplier, preamplifier, and linear amplifier

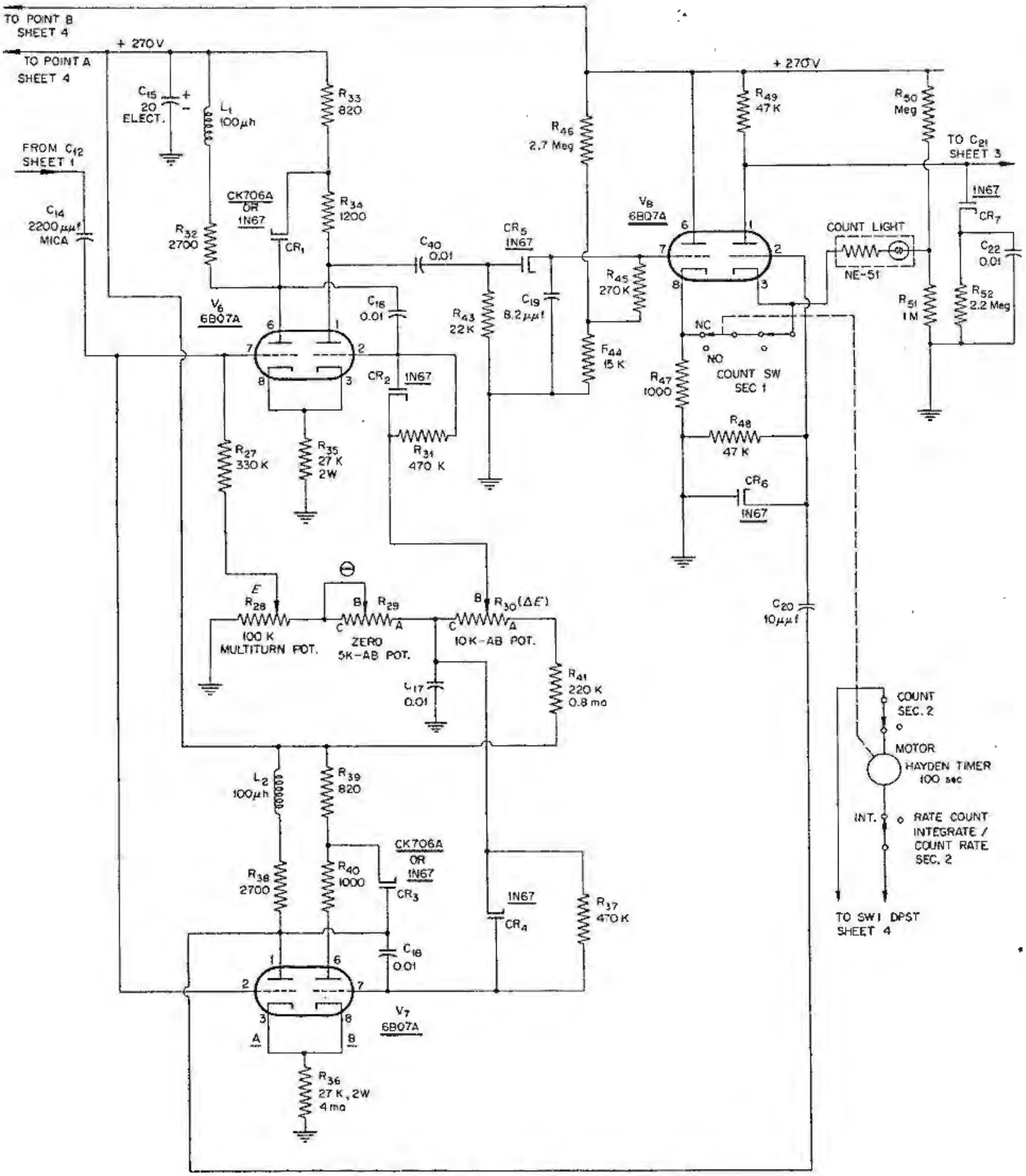

Figure 2. Schematic diagram of single-channel analyzer

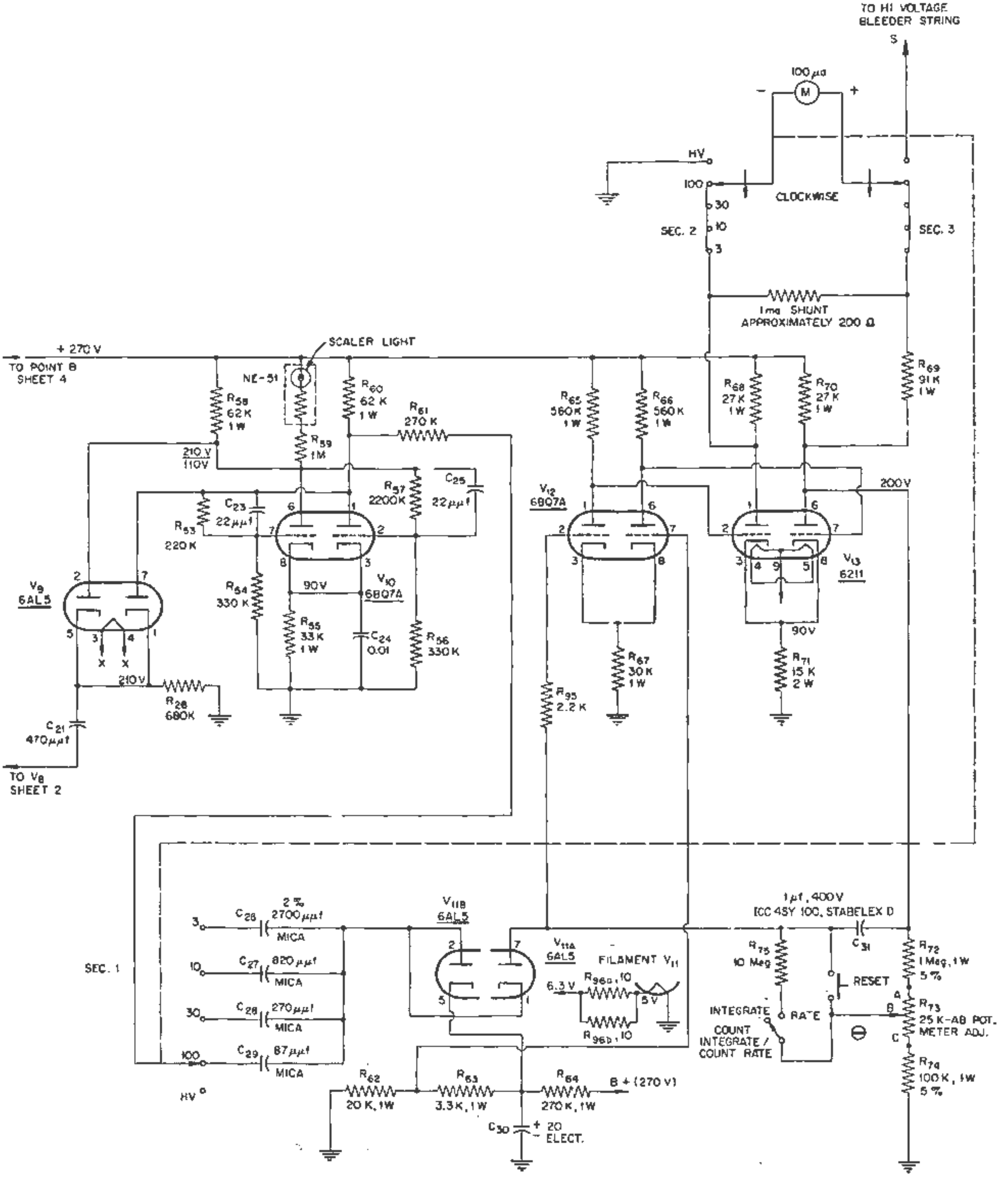

Figure 3. Schematic diagram of data storage

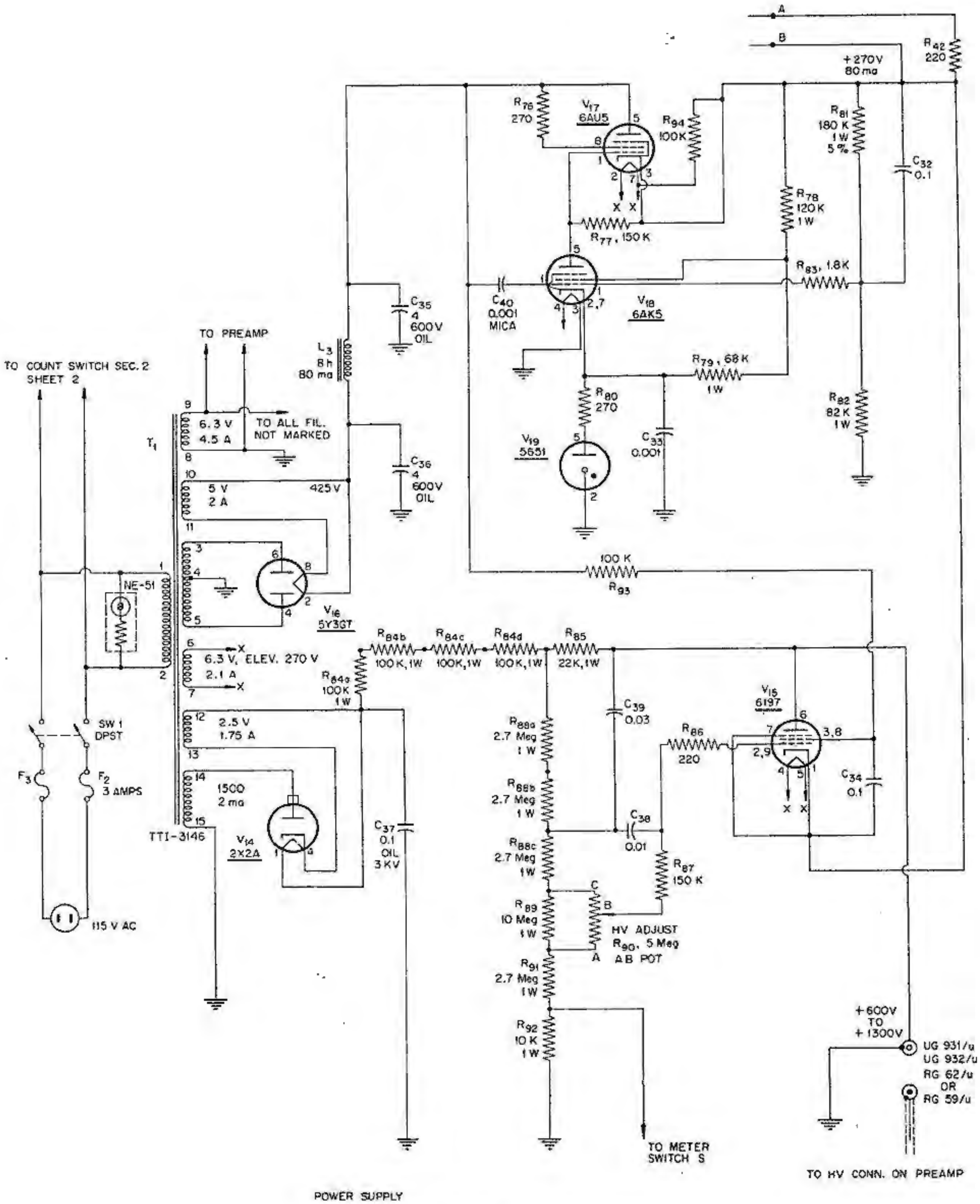

Figure 4. Schematic diagram of power supplies

which decays away with the time constant determined by the total shunt capacity and the 47k (R-48) grid resistor. This transfers the current from the first half of V-8 which is normally "on" to the second half giving a negative output from the second plate of V-8. If, however, the input signal from the amplifier is large enough to trigger both discriminators, a positive signal is obtained from the second plate of the upper discriminator (V-6) in addition to the signal from the lower discriminator. The signal from the upper discriminator is lengthened by a 1N67 (CR-5) diode and placed on the first grid of V-8. This keeps the first half of V-8 "on" even when the signal from the lower discriminator appears at the second grid. By this means no output signal is obtained.

It should be noted that the signal from the lower discriminator does not appear on the grid of V-8 until after the input pulse is no longer large enough to trigger the lower discriminator. This is necessary because of the finite rise and fall time of the pulses. On-off operation for counting is obtained by means of two switches in series connecting the cathodes of V-8 together. The switch operated by the timer is normally closed. Counting is started by closing the count switch. At the end of one hundred seconds the switch operated by the timer opens causing the counting operation to cease because the cathode of V-$8B$ is now disconnected and no signal can be obtained from the plate of V-$8B$.

Figure 3 shows the schematic diagram of the data storage section. The output pulse from V-8 is fed into a scale of two scaler. Each plate of V-10 has two stable states and is triggered from one state to the other by successive, identical pulses which are fed in through the diode V-9. When the scaler (V-10) plate changes from one stable state voltage to the other, the coupling condenser (C-26 to C-29) in use is charged or discharged through V-11. So for each two pulses into the scaling circuit a charge $q = V \cdot C$ is placed on the grid of V-$12A$ where V is the difference in voltage of the two stable states of V-$10B$ and C is the coupling capacity.

V-12 and V-13 is a two-stage dc amplifier with a feedback condenser of one μF from the plate of V-$13B$ to the grid of V-$12A$. When a charge is placed on the grid of V-$12A$ by the coupling condenser and the diode V-11, the plate of V-$13B$ moves up which pulls the grid of V-$12A$ up until equilibrium is reached again. The plate of V-$13B$ has to go up 45 volts and the plate of V-$13A$ down 45 volts to obtain one milliampere current in the cross resistor (R-69) and meter between the two plates. By using a one hundred second predetermined time, the current in the meter is calibrated in counts per second. The value for the coupling condenser C_c is obtained from the relation

$$q = \frac{N}{2} E_1 \cdot C_c = E_2 \cdot C_F$$

where E_2 is the change in voltage across C_F, E_1 is the voltage change of the scaler plate, and N is the number of counts.

The meter can also be used to read count rate by connecting in the resistor R-75 which allows the charge to leak off continuously.

Figure 4 shows the schematic diagram of the power supplies. The positive high voltage supply for the phototube is an ordinary shunt regulated voltage supply except that the positive voltage supply at two hundred and seventy volts is used for the reference voltage.

PERFORMANCE

The spectrometer was used to measure the spectra of I^{131}, Cr^{51} and Cs^{137} to check the operation of the spectrometer and also to show the effects of scattered radiation on measurements such as encountered in thyroid uptake work.

Chromium and cesium were chosen for these measurements, because they each have a single gamma

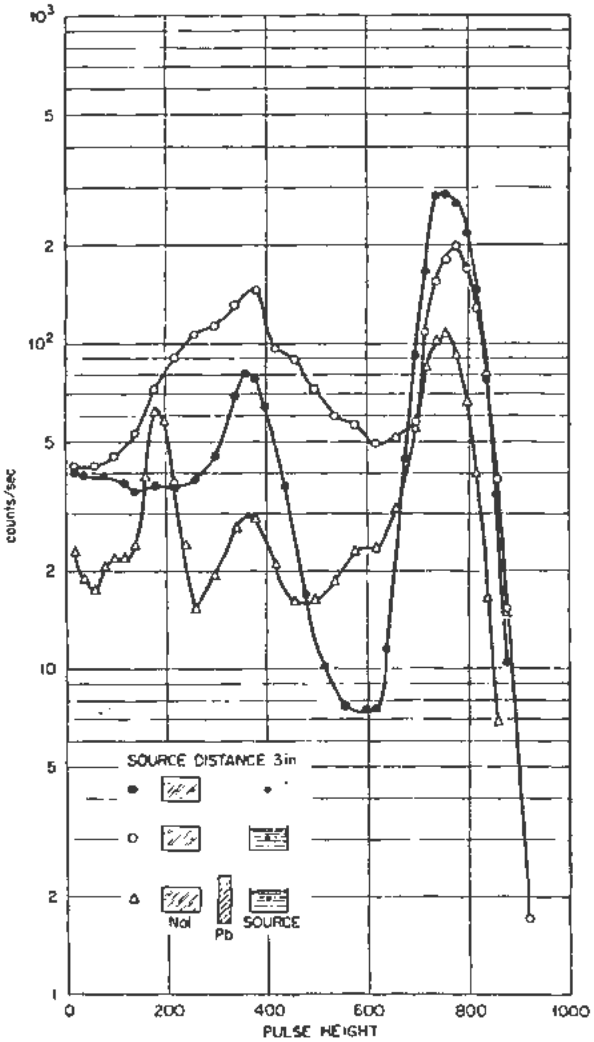

Figure 5. Pulse height spectrum of chromium with source alone and with source immersed in water showing absorption and scattering due to water and lead

TABLE I. Calibration Numbers for Source Strength Determinations

3″ × 3″ NaI Source in well ½″ × 1¼″ $\Delta E = 100$

Source	Pulse height	cps per μc
I^{131}(368)	220	16,900
(638)	500	882
Cr (320)	220	2,300

Weak Source Determination

Source	Pulse height	Counts	Time (sec)	cps − bg = cps
I alone	220	4096	291	14.05 − 1.915 = 12.13
Cr alone	220	4096	752	5.44 − 1.915 = 3.52
I + Cr	220	4096	238.9	17.15 − 1.915 = 15.23
I + Cr	500	4096	3085	1.328 − 0.705 = 0.623
Background	220	2816	1471	1.915
Background	500	512	714 avg.	0.705
			718	
			738	
			735	
Integral background	40 (60 kev)	4096	230	17.8
	220 (270 kev)	4096	500	8.2

Source strength

Assay of Cr alone by 320 kev peak $= \dfrac{3.52}{2300} = 1.53 \times 10^{-3}\ \mu c$

Assay of I alone by 364 kev peak $= \dfrac{12.13}{16{,}900} = 0.78 \times 10^{-3}\ \mu c$

Assay of I alone by 638 kev peak $= \dfrac{0.625}{882} = 0.71 \times 10^{-3}\ \mu c$

Calculating Cr and I from mixture

$(0.71 \times 10^{-3})\ 16{,}900 = 12$ cps calc. counts expected for I at 220 pulse height from 638 kev measurement.
$17.15 - 12 = 5.15$ cps due to Cr and background at 220 pulse height.
$5.15 - 1.92$ (Bg) $= 3.23$ cps due to Cr.
$\dfrac{3.23}{2300} = 1.41 \times 10^{-3}\ \mu c$ Cr, calculated value.
This value is to be compared to the assay value of $1.53 \times 10^{-3}\ \mu c$.

ray which closely approximates one of the two main gamma rays encountered in iodine.

Figure 5 shows the spectrum obtained under three different conditions from Cr^{51} which has a single gamma ray of 320 kev. First, the solid dots represent the spectrum obtained with the source at three inches from the crystal. The main peak at 780 pulse height units is the photoelectric peak, while the counts in the region 0 to 400 pulse height units are produced largely by Compton scattering in the crystal, although a few of them are due to detection of scattered radiation from the source and the crystal container. The source was then immersed in an 800 ml beaker of water to give a rough approximation of the conditions encountered in actual thyroid measurements. The spectrum shown by the circles was obtained under the above conditions. The main peak at 780 pulse height units is attenuated by absorption of the gamma rays in the water. It should be noted that the number of counts in the entire region from 0 to 600 pulse height units is increased due to the scattered radiation.

The triangles show the spectrum obtained when a $\frac{1}{16}$ inch lead shield is placed between the source in the water and the crystal. The entire spectrum is attenuated with the exception of the valley before the peak at 780 pulse height units and a peak at 180 pulse height units due to lead X-rays appears.

Figure 6 shows the spectra obtained when the integral counting method is used. That is, recording all pulses whose amplitude is greater than a given pulse height. Note that when the source is immersed in water, there is an apparent increase in the source strength for low pulse height settings. This is due to the scattered radiation. Even when a lead shield is used to minimize the effects of the scattered radiation, the attenuation measured varies greatly with different integral pulse height settings. Using a wide channel width (ΔE setting) arranged to accept the photo peak one obtains the same counting rate from Cr^{51} with a source alone as is obtained with the integral method using a lead shield. The differential method of counting will give the correct value for the attenuation of the gamma ray. In addition to obtaining the same counting rate using a wide channel width and the right answer for the attenuation the background will be cut down as much as a factor of ten using the differential method of counting. Integral and differential backgrounds are included in Table I to demonstrate this point.

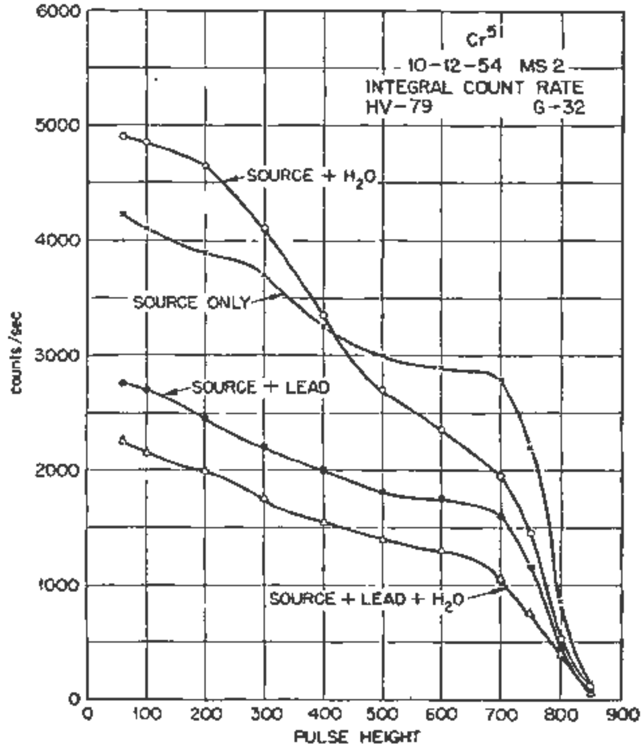

Figure 6. Integral pulse height spectrum of chromium showing effects due to scattering

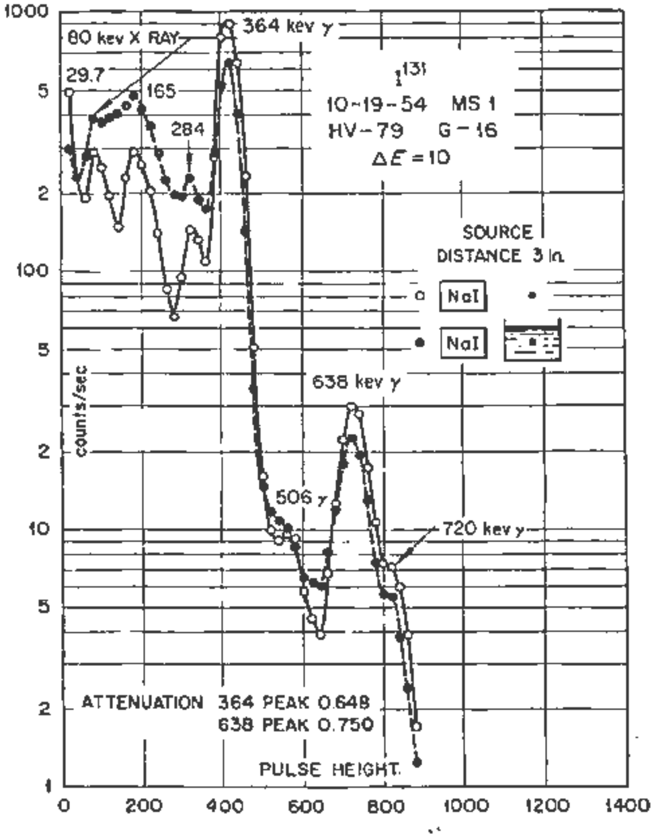

Figure 7. Pulse height spectrum of iodine with source alone and with source immersed in water to show absorption and scattering caused by water

Figure 7 shows the spectra of I^{131} under two different conditions. The circles show the spectrum for the bare source and the solid dots were obtained when the source was immersed in water. The detail of the spectrum for the bare source is good enough to permit detection of radioactive contaminants if any were present.

For the simultaneous determination of iodine and chromium in the same blood sample the following curves were obtained.

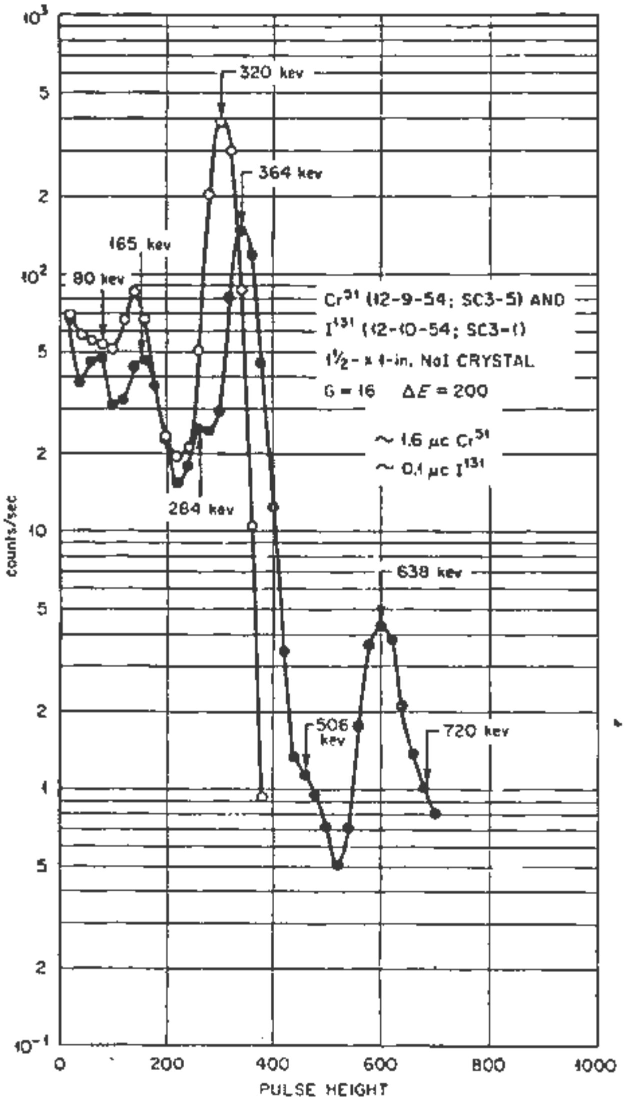

Figure 8. Spectrum of iodine and chromium under identical conditions

Figure 8 shows the spectrum of chromium and iodine alone taken under the same conditions.

Figure 9 shows the spectrum obtained when both sources are measured simultaneously. Although the 320 kev peak from chromium and the 364 kev peak from iodine are not resolved, by matching the 638 kev peak of Fig. 9 with the 638 kev peak of Fig. 8 it is possible to obtain the spectrum of chromium as shown in Fig. 8 by making a point to point subtraction.

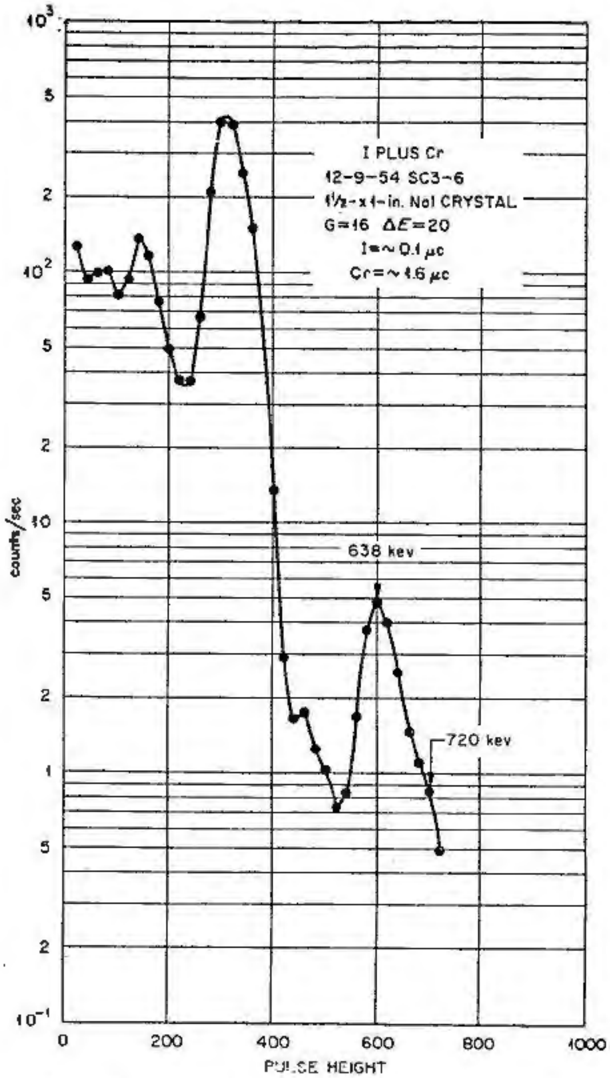

Figure 9. Spectrum of mixture of iodine and chromium

Using a narrow channel width and running a complete spectrum shows in detail the physics of the situation, and source strengths can be computed by measuring the area under the curve, but this is a time consuming process. For routine tests where the physics is understood and the sources known, the concentration of the two substances can be measured much more rapidly by using a wide channel width and proper calibration.

As an example the relative amounts of chromium and iodine in the same sample were determined by taking a measurement at only two points. Table II gives the intrinsic peak efficiencies for two crystal sizes and the counting rates expected in the peaks of the spectrum for a one microcurie source. Table I shows the experimental calibration numbers used and data obtained when measuring a weak source containing 1.5×10^{-3} microcuries of Cr^{51} and 0.7×10^{-3} microcuries of I^{131}.

TABLE II. Calculated Peak Counting Rates per Microcurie

	Efficiency	*Per cent gamma*	*Geometry*	*Counts per second per microcurie*
1 × 1½ inch crystal				
Cr(320 kev)	(0.33)	(0.1)	(0.5)	610
I (364)	(0.27)	(0.84)	(0.5)	4190
(638)	(0.13)	(0.078)	(0.5)	188
3 × 3 inch crystal, source in well				
Cr(320)	(0.747)	(0.1)	(1.0)	2765
I (364)	(0.642)	(0.84)	(1.0)	19,950
(638)	(0.381)	(0.078)	(1.0)	1110

Two collimators have been developed for use with the spectrometer. The first is a flat field collimator to be used for thyroid uptake studies. It accepts radiation only from an arc of 30 degrees. The response of this collimator as a source is moved in a circle around the collimator is shown for a source of Cr^{51} (320 kev) in Fig. 10 and for a source of Cs^{137} (661 kev gamma ray) in Fig. 11. The unique feature of this collimator is that while the transmission of the gamma rays to the crystal from the back direction is low, it was not necessary to extend the lead portion of the shield around the back of the photomultiplier tube. Instead a light piper made from a sodium iodide crystal which was not activated with thallium was used to obtain the necessary shielding for the back direction. This reduces the weight considerably while still giving the same shielding characteristics.

The second type of collimator is intended for scanning various organs of the body to map the distribution of a radioisotope within them. This collimator is called a focusing collimator and has nineteen tapering hexagonal holes in a two inch lead

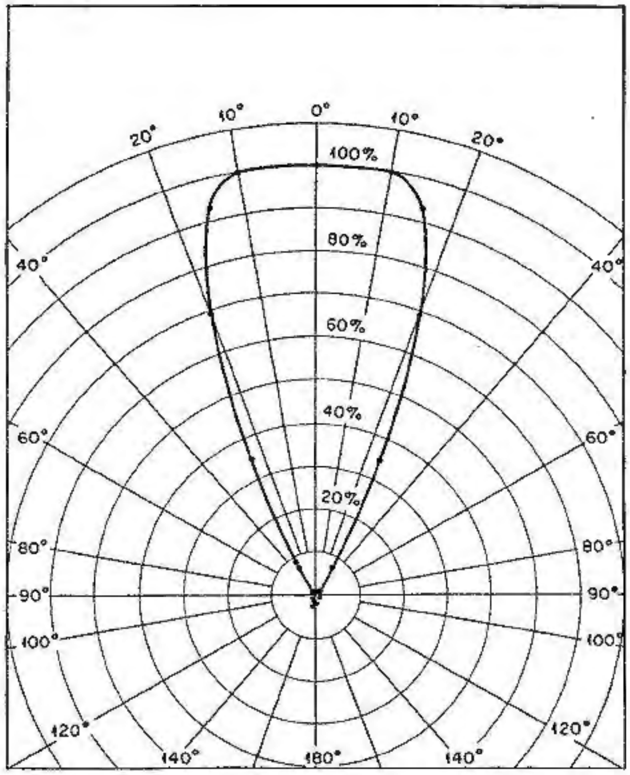

Figure 10. Polar diagram showing response of flat-field collimator to 320 kev gamma ray of chromium

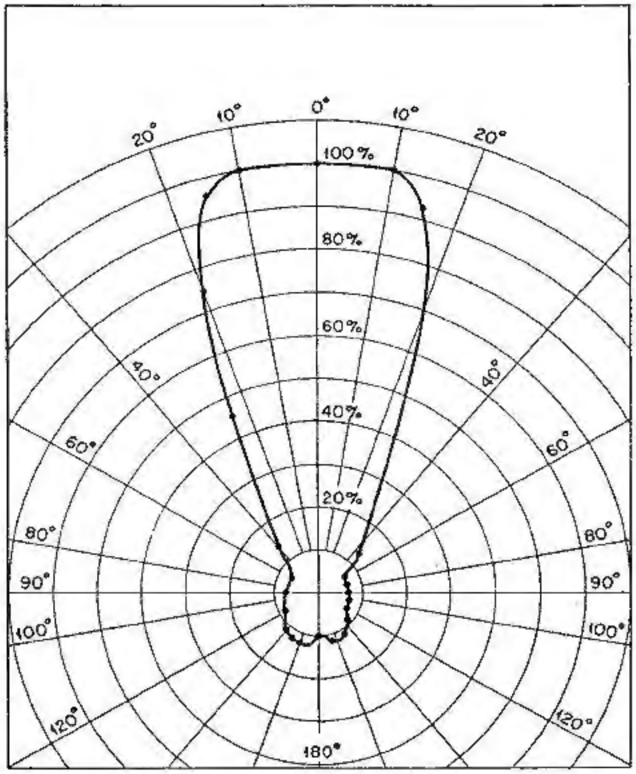

Figure 11. Polar diagram showing response of flat-field collimator to 661 kev gamma ray of cesium

shield. The axis of the holes meet at a point two inches from the face of the collimator. The transmission of the collimator is very large. At two inches the counting rate obtained is about 58 per cent of that obtained without the collimator.

Figure 12 shows a contour map of the response of this collimator to a 320 kev gamma ray point source. Note that a source on the axis gives a lower counting rate when in contact with the front face of the collimator than it does at 1.5 inches away which is the point of maximum sensitivity.

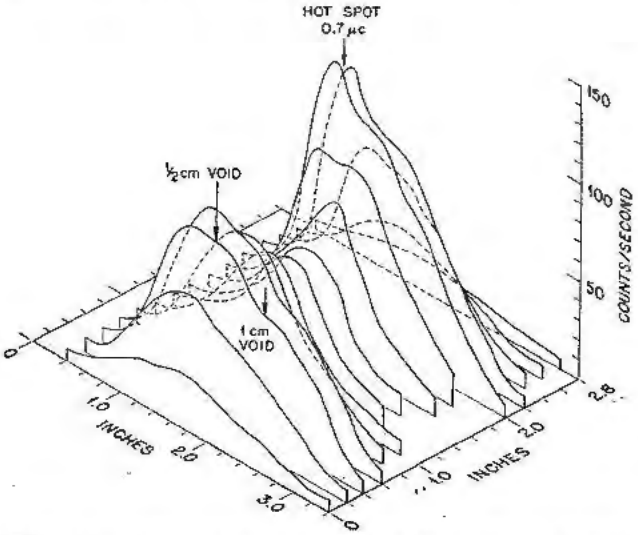

Figure 13. Isometric drawing of distribution of iodine in mock thyroid using focusing collimator

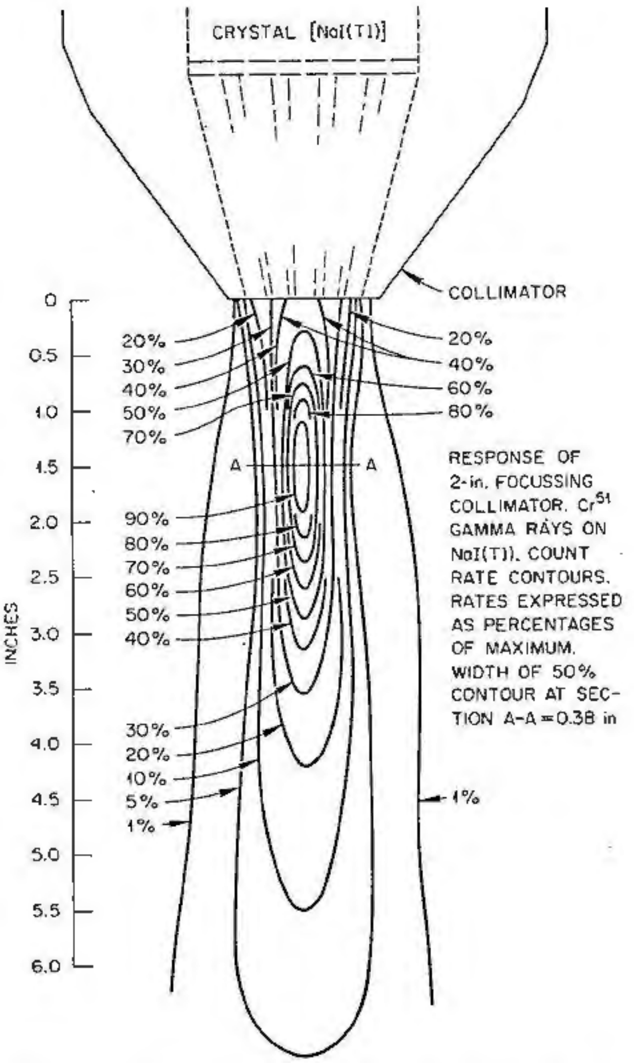

Figure 12. Contour map showing response of focusing collimator to 320 kev gamma ray of chromium

Using the focusing collimator it was possible to detect a 1 cm void at a depth of 0.75 inches in a solution tank 5 inches in diameter and 1.5 inches deep. The solution contained 0.07 microcuries per milliliter of iodine equivalent.

To demonstrate the value of this collimator with the spectrometer, a mock thyroid gland was measured. This gland contained 0.6 microcuries of iodine per cubic centimeter with an additional source 1 millimeter in diameter in one lobe containing 0.67 μc of iodine and two voids in the other lobe of 0.5 centimeter and 1.0 centimeter diameter. A map of the counting rate from the 364 kev gamma ray in iodine was made. This map (Fig. 13) definitely shows the location of the hot spot and the 1 centimeter void. The location of the 0.5 centimeter void while visible would not be definite enough to be used for a diagnosis without experience based on the shape of the contour to be expected from normal thyroid measurements.

CONCLUSIONS

We believe that the preceding curves and data show (1) that the spectrometer works satisfactorily as a radiation detector, (2) the integral method is not a precise method for measuring iodine uptake, (3) iodine and chromium in the same sample can be measured easily, (4) background counting rates with the spectrometer are much lower because only background pulses which occur within the channel being counted enter into background measurements, (5) that the focusing collimator will be of great assistance in locating abnormalities in various parts of the body, especially those that tend to absorb greater amounts of radioactive material than neighboring tissues, (6) the spectrometer can be used for source identification and assay of its radioactive content and for the identification of radioactive impurities.

Visualization of Gamma-Ray-Emitting Isotopes in the Human Body

By H. O. Anger, R. K. Mortimer, and C. A. Tobias,* USA

When a compound labeled with a radioactive isotope is administered to a human or animal, the material is distributed to various organs and sites in the subject according to the material administered and the metabolic state of the subject. It would be useful to be able to visualize on a map or photograph the distribution of the active isotope. This can be done for gamma-ray-emitting isotopes by means of the instruments described here.

THE GAMMA-RAY PINHOLE CAMERA

The gamma-ray pinhole camera is an instrument that provides a picture of the distribution of radioactivity. As described by Copeland and Benjamin,[1] it consists essentially of a lead shield, with a small aperture through which gamma rays can pass, and a gamma-ray-sensitive material such as a radiographic film located a few inches behind the aperture. Some of the gamma rays from the subject pass through the aperture and form an image of the gamma-ray-emitting areas of the subject behind the pinhole. The image can be made visible by exposing and developing the radiographic film.

The very low sensitivity of this instrument can be improved by replacing the radiographic film with a suitable phosphor and a light-sensitive photographic plate as described by Anger.[2] The phosphor converts the gamma rays to light and the resulting light exposes the photographic plate. A suitable material for this purpose is thallium-activated sodium iodide. When gamma rays impinge on this phosphor, recoil electrons are produced, which in turn produce scintillations of light. The high density of sodium iodide and the large thickness that can be used result in more efficient blackening of the photographic plate.

A working model of a gamma-ray pinhole camera using this principle is shown in Fig. 1. Some of the gamma rays from the subject below the camera travel through the pinhole and reach the large flat sodium iodide crystal at the top. The gamma rays cause the crystal to emit light, some of which reaches and exposes a photographic plate placed just above the crystal. The spreading of the light before it reaches the photographic plate and the consequent loss of definition are limited by the inverse-square law and by total reflection at the boundary between the crystal enclosure and the photographic plate.

Because thick transparent intensifying screens are used, it is not possible to obtain good definition, but this can be tolerated in a gamma-ray pinhole camera because the definition of the camera is inherently low. Even if the pinhole is made very small some gamma rays travel through the lead adjacent to the pinhole, and the effective size of the aperture is still fairly large.

The use of this technique allows the sensitivity of the camera assembly to be about 20 times as great as it would be if Kodak No-Screen X-ray film were used with the usual lead foil intensifying screens. Thus it is possible to obtain a faint image of a source containing 1 millicurie of I^{131} per square centimeter with an exposure time of 1 hour when the pinhole size is $\frac{1}{8}$ inch, the thickness of the thallium-activated sodium iodide crystal used as the intensifying screen is $\frac{5}{16}$ inch, and Kodak type 103a-0 spectrographic plates are used as the light-sensitive material.

Although this sensitivity is quite low, it has been possible to obtain an *in vivo* gamma-ray autoradiograph of a metastatic thyroid tumor containing 20 millicuries of I^{131}. It was possible to take this picture only because the patient required a therapeutic dose of I^{131}. The tumor was located at the patient's elbow.

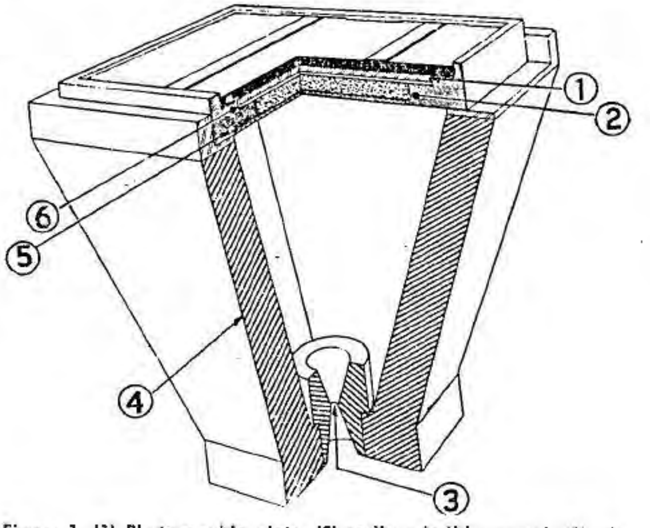

Figure 1. (1) Photographic plate, (2) sodium iodide crystal, (3) pinhole aperture, (4) lead shielding, (5) black bakelite, and (6) lucite or glass

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* Donner Laboratory of Biophysics and Medical Physics and Radiation Laboratory, University of California, Berkeley, California.

The radioactive iodine is shown to be taken up in two main areas in the tumor.

THE PINHOLE CAMERA AND IMAGE AMPLIFIER

The sensitivity of the gamma-ray pinhole camera can be increased by use of an image-amplifier tube similar to those used for the intensification of fluoroscopic X-ray images. A drawing of the instrument as described by Mortimer, Anger, and Tobias[3] is shown in Fig. 2. The subject is at the left of the camera. Gamma rays from the subject pass through the pinhole and form an image of the gamma-ray-emitting areas of the subject on the mosaic of fluorescent crystals. Each crystal is surrounded with a reflector which directs the light produced in the crystal to the photocathode of the image-amplifier tube. Light falling on the photocathode causes electrons to be emitted. The electrons are accelerated by a 25-kv potential and are focused by an electron lens on a small zinc sulphide-zinc selenide screen at the other end of the tube. The image formed here can be viewed through a magnifying lens system or it can be photographed.

The tube increases the light intensity by a factor of 600. The gain is due both to the acceleration of the photoelectrons by the 25-kv potential and to the reduction of the image size by the electron lens. The size of the image on the fluorescent screen is ⅑ the size of the photocathode, thus gaining by a factor of 81 in brightness because of the reduction in size of the electron image. With this apparatus it is possible, after dark-adapting the eyes, to view on the screen sources of radioactivity as small as about 200 microcuries per square centimeter.

The useful gain of the image-amplifier tube—and therefore the sensitivity of the system—is limited by the background glow of the viewing screen when no light falls on the photocathode. This glow is caused by thermal and field emission of electrons from the photocathode surface, and also by electrons knocked out by cesium ions migrating to the photocathode. The background due to the latter cause can be reduced by a factor of 5 by cooling the tube to 0°C.

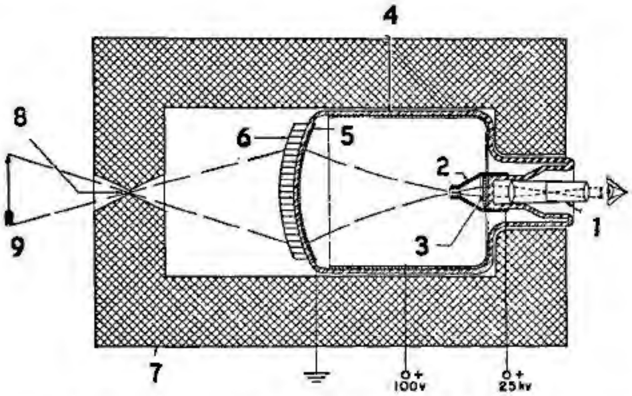


Figure 2. (1) Optical system, (2) electron lens, (3) fluorescent screen, (4) image amplifier tube, (5) photoelectric cathode, (6) mosaic of fluorescent crystals, (7) lead shield, (8) pinhole aperture, and (9) subject

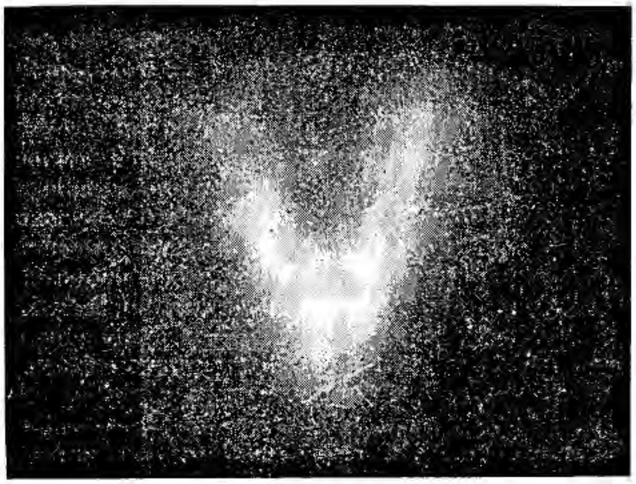


Figure 3

However, even with the image-amplifier tube cooled to 0°C and with the image photographically integrated, the sensitivity is about 10 microcuries per square centimeter with a 30-minute exposure time. This sensitivity is great enough for some *in vivo* tracer experiments. Greater sensitivity would increase the usefulness of the instrument considerably.

A photograph of an image obtained on the screen of the image-amplifier tube from a radioactive source is shown in Fig. 3. The source was a V-shaped groove in a lucite block filled with I^{131} solution. The activity of the source was $4mc/cm^2$. The diameter of the pinhole aperture was 3 millimeters and the exposure time to obtain this picture was 8 minutes with Super XX film at f/6.

The sensitivity would be considerably greater if the photographic film could be placed in contact with the fluorescent screen. In the tube used for these tests the screen was located at an appreciable distance inside the envelope, thus making it necessary to photograph it with a camera. This caused a considerable loss in light and sensitivity. The ultimate in sensitivity would be reached if each scintillation produced in one of the fluorescent crystals by a gamma ray were amplified so as to be visible to the eye and so that it would appear as a dot if a photograph were taken. This sensitivity might be achieved if a two- or three-stage image amplifier were used. Further work along these lines is indicated.

THE MULTIPLE-SCINTILLATION COUNTER SCANNER

Another method of determining the distribution of radioactivity in a subject is to scan over the subject with one or more directional gamma-ray counter and indicate by some means the relative counting rate over the area scanned. Scanners employing a single scintillation counter have been described by Cassen[4] and Mayneord.[5]

If more than one scintillation counter is used, the area that can be covered in a given time is proportionately increased. A scanner employing 10 scintillation counters is shown in Fig. 4. This instrument

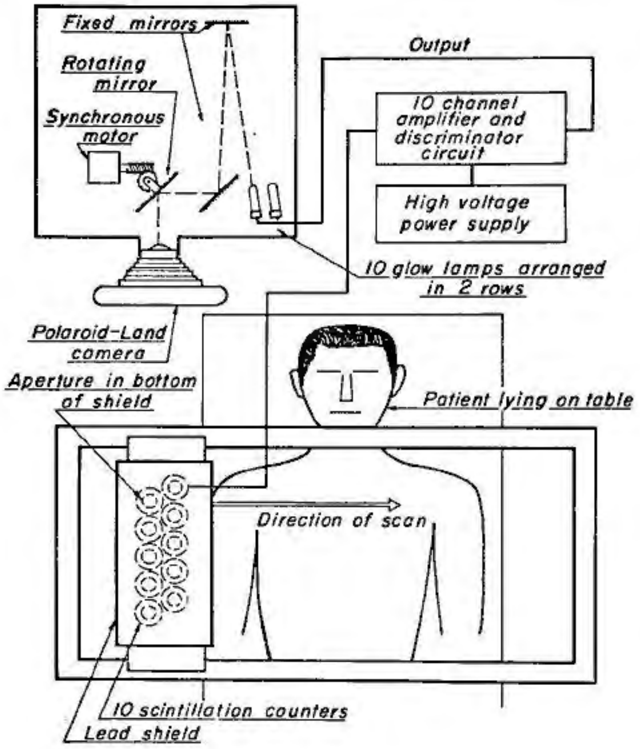


Figure 4

can produce a picture of the distribution of radioactivity in the whole body of a human subject in about 45 minutes and can detect less than ¼ microcurie per square centimeter. It is a modification of a 10-counter scanner described previously.[6]

The instrument consists essentially of 10 scintillation counters in a lead shield, 10 glow lamps connected to the counters through amplifiers so that a count from each of the counters produces a flash of light in the corresponding glow lamp, a mirror system, and a Polaroid Land camera that records the flashes from the glow lamps as spots on a photographic film.

Each scintillation counter consists of an end-window photomultiplier tube 1.5 inches in diameter which is coupled optically to a thallium-activated sodium iodide crystal 0.5 inch in diameter by 1 inch long. The counters are positioned along two straight parallel lines as shown in Fig. 4. Below each counter is an aperture in the lead shield which points straight down. Various sizes of apertures are used, depending on the sensitivity and definition required. To scan an area for gamma-emitting isotopes, the counters are moved slowly by means of a motor in a direction perpendicular to the two lines along which the counters are located. An area 8 inches wide and any desired length (usually 24 inches) is covered by each scan.

When a person is to be scanned, the subject lies on a table and the counters move over him from left to right for a distance of 24 inches and then return to the starting point. To scan the whole subject, about 10 or 11 scans are taken. The first scan covers the subject's head; then the table on which the patient lies is moved so that the second scan covers his neck; the third scan covers his upper chest; and so on. Four minutes are required for each scan and 45 minutes are required to scan in this manner from head to toe. The resulting separate pictures are joined together to form a composite head-to-toe picture.

The distribution of activity is recorded by means of the 10 glow lamps, the mirror system, and the camera. The glow lamps and 10-channel amplifier are connected so that each gamma ray detected by any of the scintillation counters produces a flash of light in the corresponding glow lamp. For instance, gamma rays detected by the first counter produce flashes in the first glow lamp, and so on. The glow lamps and mirror system are contained in a dark box, and are so arranged that while the counters are moving over the subject, the rotating mirror in front of the camera lens causes the glow lamps to appear to move in synchronism before the camera. During each scanning period a time exposure of the glow lamps is taken with the Polaroid Land camera. This camera develops and delivers a finished print one minute after each scan is taken.

Each flash of a glow lamp appears as a dot on the photograph. Where the activity is greatest in the subject, the greatest number of dots appears on the corresponding part of the photograph. Therefore, a map of the distribution of activity is obtained with concentrations of dots indicating where the activity is greatest. The natural background due to cosmic rays and stray radioactivity appears as a few dots randomly distributed over the print.

A single scan results in a picture with 10 rows of dots having what might be called 10-line definition. In practice, increased definition is obtained by interlacing scans in the following way. The counters first move from left to right over the subject. Then the shield is moved, in a direction perpendicular to the original direction of motion, just one-half the distance between the apertures. Then the counters scan from right to left over the same area of the subject. At the same time the images from the glow lamps are interlaced optically by an adjustment in the mirror system. The result is a scan with 20-line definition. This is the type of scan usually taken on all large subjects.

Test images obtained with the scanner are shown in Fig. 5. The scans were taken of a radioactive test pattern made by filling an X-shaped groove in a lucite block with I^{131} solution. The test pattern contained a total of 10 microcuries of I^{131} or about 0.25 microcurie per square centimeter. It was located one inch from the scanner shield, and apertures ⅜ inch in diameter were used. The thickness of the shield was 1 inch. A 10-line image made by a single scan is shown at (*A*), a 20-line interlaced image at (*B*), and a 40-line interlaced image at (*C*). The increase in definition obtained by interlacing is clearly demonstrated.

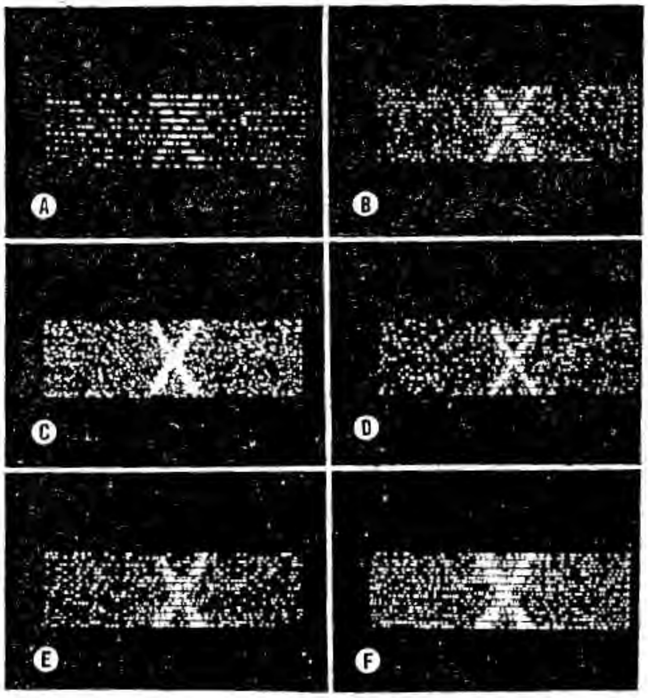


Figure 5

The same radioactive test pattern was scanned when it was located 0.25 inch away from the shield in (*D*) and 3 inches away in (*E*). These pictures show the decrease in definition obtained as the distance to the radioactive source is increased.

The result of increasing the scanning time without changing the interlacing is shown by comparing (*E*), which is a regular 4-minute, 20-line interlaced scan, with (*F*), which was made by recording two 4-minute scans over the same area on the same print. The repeated scan gives a more definite image because of the larger number of counts recorded.

The scanner is sufficiently sensitive to detect one microcurie of I^{131} if the activity is concentrated in an area 1 inch or less in diameter. This sensitivity is for a 20-line interlaced scan with apertures ⅜ inch in diameter and with material equivalent to 2 inches of tissue between the scanner and the radioactive source. Slightly weaker sources can be detected with no loss in definition by taking repeated scans of the same area, each scan being recorded on the same print.

Some examples of *in vivo* gamma-ray pictures are shown in Fig. 6. The four pictures show the liver and spleen of human subjects who have received an intravenous injection of 50 to 100 microcuries of colloidal gold-198. Nearly all of this material is taken up by the liver and spleen. In each case, the liver is at the left of the picture and the spleen, sometimes considerably enlarged over the normal size, is shown more faintly on the right. The radioactivity present in these organs was as little as 0.1 microcurie per square centimeter of area as seen by the scanner. Each picture covers an area 16 by 24 inches and the total scanning time for each was 16 minutes. From these pictures the size and shape of the organs can be estimated. Stirret, Yuhl, and Cassen[7] have used similar surveys of the liver to detect tumors large enough to displace an appreciable amount of liver tissue.

An example of head-to-toe scanning of a thyroid carcinoma patient is shown in Fig. 7. The patient's thyroid had been removed 2 years previously and he had a large metastatic lesion near his left elbow for which he had been given therapeutic doses of I^{131}. No other lesions were evident before the scanner pictures were taken. The patient was given an oral dose of 5 millicuries of I^{131} and a series of three head-to-toe scanner pictures was taken. The first was taken 1 hour after the dose was administered, and it shows that the I^{131} was distributed throughout his body. The second was taken 48 hours later and shows some points of concentration of iodine, but they are inconclusive except for the elbow because of the high body background. An outline of the patient's body is still visible because of the slow disappearance of iodine in this patient from the blood and intercellular space. The third picture was taken at 96 hours and shows 6 points where iodine was taken up in significant amounts. They are at the sternum, the right pelvic region, the lower right chest, the left arm near the shoulder, the jaw region near the front teeth, and the left elbow. The locations are indicated on the outline drawing at the right.

X-ray radiographs were taken of the points where iodine was taken up after they were found with the scanner. On the sternum near the third rib a calcified area was found, and in the right pelvic region a small spur of bone was found on the ilium. Neither of these artifacts as seen on an X-ray radiograph was abnormal enough to be considered as the site of a metastatic lesion on the basis of the X-ray evidence alone. However, on the basis of the findings with the scanner, all six points, with the possible exception of the jaw region, can be identified with relative certainty as the site of a metastatic thyroid lesion.

The use of gamma-ray scanning in combination with X-ray radiographs provides maximum information in locating thyroid lesions, since the X-ray radiograph, if the lesion is visible on it, shows with greater accuracy where the lesion is located, and also

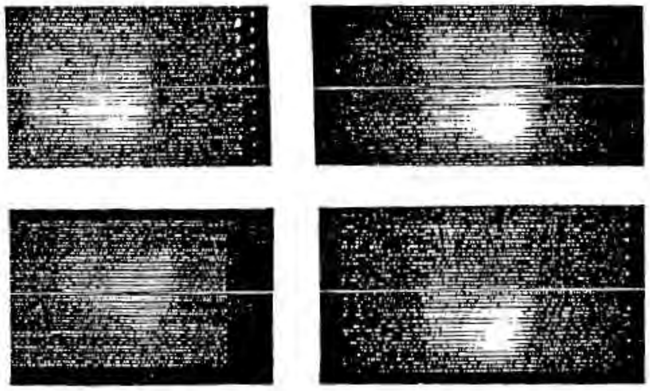
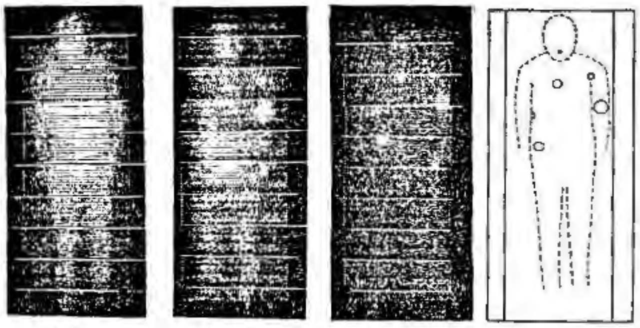


Figure 6



1 hour 48 hours 96 hours

Figure 7

indicates the depth. However, thyroid lesions that are not visible by means of X-ray radiographs can be detected and located by means of gamma-ray scanning.

CONCLUSION

The pinhole camera with an improved image amplifier and the scintillation-counter scanner both show promise of increasing usefulness in tracer research and in diagnosis involving gamma-ray-emitting radioisotopes. The scanner is useful at the present time for locating thyroid lesions and for outlining the liver and spleen. Other uses will be found as new tracer compounds and techniques are developed.

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Dosimetry of Reactor Radiations by Calorimetric Measurement

By D. M. Richardson,* A. O. Allen† and J. W. Boyle,* USA

Of basic importance in the study of the effect of radiations on matter is the quantity of radiation energy which is absorbed. With most sources of radiation this quantity can be readily estimated. For materials exposed to the radiations of a nuclear reactor, however, even a rough estimation of the quantity of energy absorbed is difficult. Materials placed in a reactor absorb energy by slowing down fast neutrons, by absorption of gamma-rays produced by fission and the decay of fission products, and by self-absorption of rays from the induced radioactivities in the materials themselves. Since neither the flux nor spectrum of neutrons and gamma-rays in a thermal reactor can be accurately calculated, the determination of total absorbed energy requires calorimetric measurement.

Such measurements in a reactor are subject to unusual difficulties. All parts of the calorimeter will absorb radiation and generate heat, so that special provision must be made for distinguishing between heat generated by the sample material and that being produced in the rest of the apparatus. A water-cooled hole in the Oak Ridge Graphite Reactor provided a favorable thermal environment for calorimetric work. The method chosen was that of isothermal calorimetry in which the total heat flow across a boundary surrounding the material was determined by the steady state temperature difference across the boundary. This method also provided the possibility of direct comparison of the heat generated by the sample with heat introduced electrically. Measurements have been carried out which are believed to represent the total heat production in samples irradiated in the water-cooled hole with an accuracy of a few per cent.

The problem remaining is the subdivision of the total absorbed energy into the portions resulting from neutron scattering, gamma-ray absorption and self-absorption. The self-absorption energy can be reasonably calculated for each material since thermal neutron reactions and radioisotope decay processes are well characterized. Further division of the remaining total energy can be achieved by solution of simultaneous equations of the total energy for two or more materials. By means of relative mass absorption coefficients for gamma-rays and relative neutron scattering integrals, using an assumed neutron spectrum, the final division of energy is possible which is consistent for the materials studied.

EXPERIMENTAL PROCEDURE AND RESULTS

The calorimeter consisted essentially of two coaxial aluminum tubes with a thermocouple embedded in each (see Fig. 1). The heat generated within the inner assembly was conducted mainly across the air gap between the two tubes, good thermal insulation being provided at the ends of the inner tube. Convection currents were minimized by a thin sleeve of polystyrene foam in the middle of the air space. A helical coil of fine insulated copper wire was embedded in the wall of the inner tube and the temperature difference between the tubes was calibrated by electrical heating. The electrical heat input was determined by simultaneous measurement of the current and of the heater resistance by means of the four lead potentiometer method of Mueller.[1]

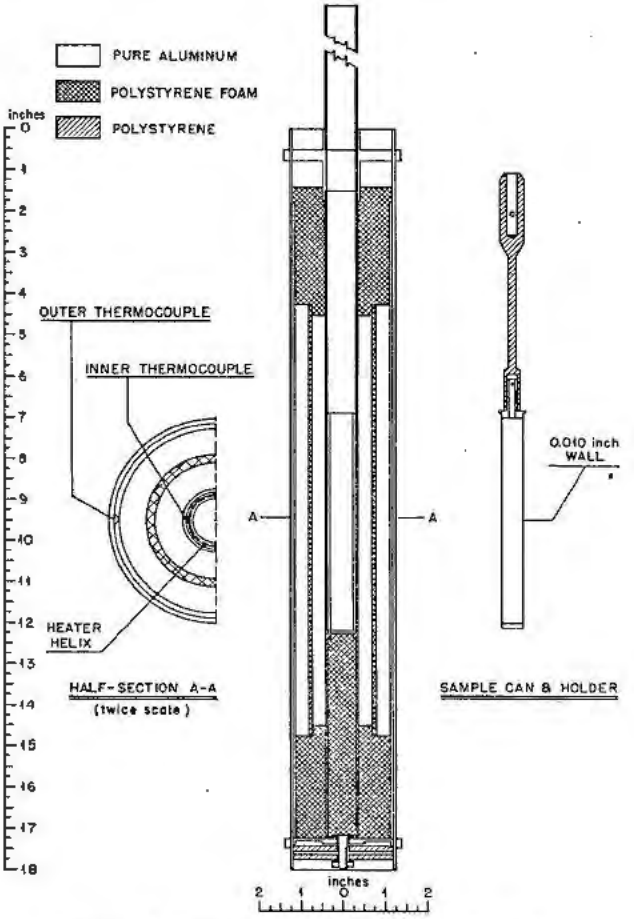

Figure 1. Reactor radiation calorimeter, sectional views

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The aluminum sample cans were identical in their dimensions and were uniform in weight within 0.6%. Since the reaction of water and aluminum is highly exothermic it was necessary to season the sample cans used for water before their use in the calorimeter. This required approximately 10 days at 90°C. The water sample cans were tightly closed to prevent evaporation during measurements. The sample materials used were: redistilled H_2O; D_2O of 99.85 mole-% isotopic purity; graphite of reactor quality, powdered by machining; c.p. granulated bismuth of 99.8% purity; and commercially pure aluminum.

During measurements the reactor power was held constant within 0.2%. An empty can was first placed in the calorimeter and left until the temperature difference between the thermocouples became constant at approximately 3°C. This temperature difference was due to the background radiation heating of the inner calorimeter assembly and was taken as the reference when additional heat was added, whether by radiation heating of a sample or by electrical heating. The calibration curve of electrical heat versus temperature difference was linear. The measured total rates of radiation energy absorption are presented in Table I. The probable error is estimated to be 4.5% for carbon and about 2% for the other materials.

INTERPRETATION

The heat generated in H_2O, D_2O and carbon arises almost completely from the elastic scattering of neutrons and the absorption of gamma rays. The gamma-ray contribution per mole is the same for D_2O and H_2O and the large difference in heat generation observed in these materials indicates that most of the energy in H_2O results from the slowing down of fast neutrons. The heat production per gram in carbon is smaller primarily due to its lower efficiency in moderating neutrons.

The ratios of the true mass absorption coefficients for gamma-rays (μ_T) of water to those of carbon are constant within 1% in the energy range from 0.2 to 2.0 Mev.[2] In contrast, the average fraction of kinetic energy transferred per neutron collision ($K = 2A/(A+1)^2$) varies by several factors from carbon to deuterium and hydrogen. Likewise, the neutron scattering cross sections (σ_s) of hydrogen, deuterium, oxygen and carbon are dissimilarly affected by changes in neutron energy.[3] These considerations, in addition to the insignificant amount of nuclear reaction experienced by these materials, indicate that accurate division of the total absorbed energy into two contributions is particularly favored in the case of light water, heavy water and carbon. Therefore the heating data for these materials were chosen for analytical treatment.

The cross section for neutron scattering varies with neutron energy in a widely different manner for these elements, so that a quantitative treatment of the results requires a knowledge of the neutron spectrum at the place where the materials were exposed. For the purposes of calculation, we have assumed the neutron spectrum shown in Fig. 2, which is of the sort to be expected at the point of exposure in the Oak Ridge graphite lattice.[3-6] Since only integrals are compared in the calculations, the details of the actual spectrum are not important and may differ greatly from the curve assumed.

In general, the number of collisions made by neutrons of energy between E and $E + d(\ln E)$ is given by $(nv)_{\ln E}\,\sigma_s N d(\ln E)$ where N is the number of atoms present. The average amount of energy transferred per collision is KE. The rate of energy loss to a material is obtained by integrating over all energies. Per mole of target atoms, the resulting integral is

$$I = N_A K \int_0^\infty (nv)_{\ln E}\,\sigma_s dE \qquad (1)$$

where N_A is Avogadro's number. In Fig. 2 are shown the graphical integrals of this equation for various numbers of H, D, C, and O atoms: number of H atoms in one gram of H_2O; number of O atoms in one gram of H_2O; number of D atoms in one gram of D_2O; and the number of C atoms in one gram of carbon. The values of the integrals, and those for one gram of H_2O and one gram of D_2O derived therefrom, are shown in Table II along with associated information.

The total heat produced by reactor radiation per mole of these materials is expressed in the following equation:

$$Q\,M_m = S\,(I) + G\,(M_m/M_{H_2O})\,(\mu_{T,m}/\mu_{T,H_2O}) \qquad (2)$$

where Q is the total rate of energy absorption in calories/gram/second; M_m is the molecular weight of material m; S is the empirical scattering constant, which is the combined factor (actual flux/assumed flux) × (calories/Mev); I is the scattering integral in Mev/mole; G is the empirical gamma-ray con-

TABLE I. Rate of Heating in the Oak Ridge Graphite Reactor at 3500 Kilowatts

Material	*Weight of sample in grams*	*Temperature difference produced, °C*	*Calories per second per gram*
H_2O	16.025	1.60	0.001217
H_2O	15.765	1.59	0.001226
D_2O	16.653	1.06	0.000774
Carbon	10.100	0.39	0.000467
Aluminum	43.691	2.10	0.000585
Bismuth	89.103	4.20	0.000573

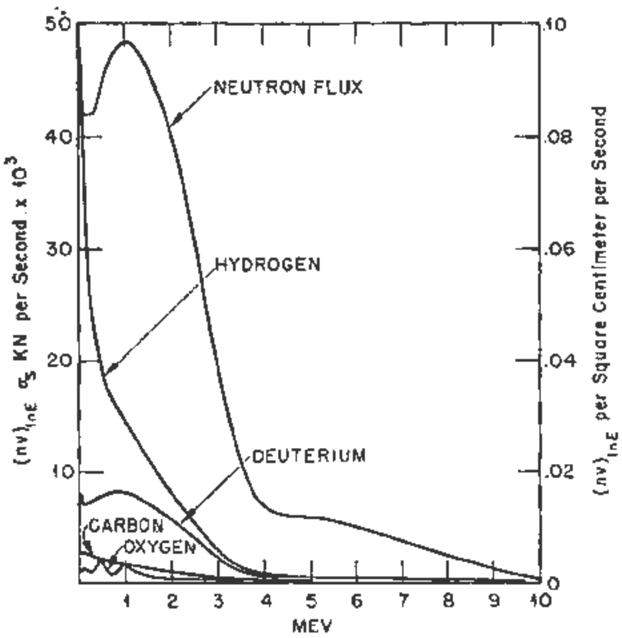

Figure 2. Assumed neutron spectrum in Oak Ridge Graphite Reactor and resulting energy integrals from neutron scattering

stant, which is the gamma-ray heat absorbed in calories/second by one mole of H_2O. The empirical constants of Equation 2 were evaluated by the method of least squares, using the experimental heating data for H_2O (two determinations), D_2O and C. The four equations and the resulting values of S and G are shown in Table III.

By use of the empirical constants of Equation 2 it is possible to calculate the neutron-scattering and gamma-ray absorption heat of these materials. The partial heats thus calculated are shown in Table IV. Agreement between the calculated total heats and the observed total heats is within experimental error. The application of these results to the cases of aluminum and bismuth requires special treatment and is discussed below. The estimated partial heats in aluminum are included in Table IV.

The total heat produced in aluminum results not only from absorption of reactor radiations but also from activation of aluminum by the (n,γ) reaction with thermal neutrons to produce Al^{28} which decays with a half-life of 2.4 minutes and is therefore in radiative equilibrium during measurement. Each

TABLE II. Neutron Scattering Energy Integrals for Assumed Flux*

Material	*Number of atoms or molecules*	*K*	*Scattering energy Mev per second*
H	6.69×10^{22}	0.5000	0.04304
D	6.02×10^{22}	0.4444	0.02215
O	3.34×10^{22}	0.1107	0.002768
C (1 gm)	5.01×10^{22}	0.1420	0.004817
H_2O (1 gm)	3.34×10^{22}	—	0.04580
D_2O (1 gm)	3.01×10^{22}	—	0.02464

* σ_s values obtained from reference 3.

TABLE III. Simultaneous Equations and Resulting Empirical Constants by Method of Least Squares

$$Q\,M_m = S\,(I) + G\,(M_m/M_{H_2O})(\mu_{T,m}/\mu_{T,H_2O}) \tag{2}$$

$$H_2O\ (0.001217)(18.016) = S\,(0.04580)(18.016) + G\,(1)(1)$$
$$H_2O\ (0.001226)(18.016) = S\,(0.04580)(18.016) + G\,(1)(1)$$
$$D_2O\ (0.000774)(20.028) = S\,(0.02464)(20.028) + G\,(20/18)(18/20)$$
$$C\ (0.000467)(12.011) = S\,(0.004817)(12.011) + G\,(12/18)(9/10)$$

$S = 1.749 \times 10^{-2}$ (calories) $(\text{Mev})^{-1}$

$G = 7.416 \times 10^{-3}$ (calories) $(\text{second})^{-1}$ (mole of $H_2O)^{-1}$.

TABLE IV. Energy Absorbed by Various Materials in Oak Ridge Graphite Reactor in Calories per Gram per Second, at 3500 kw

Material	*γ-Energy calculated*	*n-Scattering energy calculated*	*Total energy calculated*	*Total energy observed*
H	0.000738	0.006727	0.007465	
D	0.000369	0.001926	0.002296	
O	0.000371	0.000055	0.000426	0.001217
H_2O	0.000412	0.000801	0.001213	0.001226
D_2O	0.000370	0.000431	0.000801	0.000774
C	0.000370	0.000084	0.000454	0.000467
Al Reactor radiation	0.000358	0.000023		
$Al^{27} + n \rightarrow$	0.000042			
Al^{28} + 8 Mev				
$Al^{28} \rightarrow Si^{28} + \beta^-$	0.000162			
$Al^{28} \rightarrow Si^{28} + \gamma$	0.000013			
Sub-total	0.000575	0.000023	0.000598	0.000585

decay of Al^{28} produces a gamma-ray of 1.8 Mev[7] and a beta-ray having a maximum energy of 2.75 Mev[7] with an average energy of 0.4385 times the maximum.[8] The activation cross section for aluminum is 0.21 barns.[9] Using a value for the thermal flux of 0.772×10^{12} neutrons/centimeter²/second for the position of exposure, the saturation activity of the aluminum is calculated to be 95.085 millicuries/gram. This corresponds to an output of 0.000162 calories/gram/second in the form of beta-rays, and 0.000242 calories/gram/second in the form of gamma-rays. All of the beta-ray energy will be measured; however, only a fraction of the gamma-rays will be absorbed in the inner part of the calorimeter. A rough estimate of this fraction was made by assuming that all of the activity was concentrated in the form of a line source down the center of the specimen. The estimated fraction was 0.053 or 0.000013 calories/gram/second. It was assumed that all of the energy of the capture gamma-rays emitted when Al^{28} is formed appeared as one quantum of about 8 Mev. Using the same fraction for the percentage absorbed, it was estimated that capture gamma-rays contribute 0.000042 calories/gram/second.

The neutron-scattering heat in aluminum was a relatively small contribution and was estimated by taking 2.5 barns as the average scattering cross section for fast neutrons and multiplying the neutron heat per gram of carbon by the ratio 2.5/2 (since the average cross section for carbon is about 2 barns), by the ratio of the values of K for aluminum and carbon, and by the ratio of the atomic weights of carbon and aluminum. The neutron-scattering heat thus estimated was 0.000023 calories/gram/second.

The heat produced by absorption of reactor gamma-rays in aluminum was estimated by multiplying the gamma-ray heating of carbon per gram by the ratio of the true mass absorption coefficients of aluminum and carbon. This contribution was 0.000358 calories/gram/second.

The heat produced in bismuth by neutron scattering is negligible compared with the other materials, due to the small value of K. The heating in bismuth arises almost entirely from gamma-rays, and may be estimated by multiplying the gamma-ray component of the carbon heat by the ratio of the true absorption coefficients of bismuth and carbon. This ratio is 1.085 for 1 Mev gamma-rays and 2.88 for 0.5 Mev gamma-rays. The ratio between the measured heat per gram of bismuth and the calculated heat produced by gamma-ray absorption in carbon is 1.55. Thus the effective energy of the reactor gamma-rays would appear to be less than 1 Mev.

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Methods of Measurement of Neutron Flux at Low Levels

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Many methods have been developed for evaluating neutron flux levels and energy spectra for research purposes. However, they are often either too insensitive or too complicated for use in evaluating radiation hazards. This paper will describe a number of the methods available for such use with emphasis on the determination and improvement of sensitivity.

THRESHOLD DETECTORS

Certain neutron-induced nuclear reactions, occurring only above a characteristic energy threshold, may be used to obtain a general idea of an unknown spectrum of neutrons. The method has the advantage of being applicable to pulsed accelerators as well as to continuously operating neutron generators. Although foils used as threshold detectors are usually too insensitive for health physics applications, a great improvement in sensitivity can be realized by the use of scintillation techniques. Seven such detectors evaluated by the authors are listed in Table I, together with pertinent descriptive and performance data.

Values of minimum detectable flux in Table I are arbitrarily chosen such that the statistical uncertainty at the 90% confidence level is equal to the flux, for instance 2.3 ± 2.3 in the case of the anthracene detector. This corresponds to a 95% probability that a measured flux of the stated minimum detectable value is indeed greater than zero.[1] Flux levels of 5 to 10 times minimum detectable are required before reasonably accurate measurements can be made. In computing values of minimum detectable flux, background counts of 30 minutes are assumed. A variety of irradiation times are used, depending on the half-life of the product activity and the inherent sensitivity of the detector. A five-minute interval is allowed between irradiation and counting, except where a larger value is required to eliminate an unwanted activity of shorter half-life.

THE ANTHRACENE DETECTOR—20 MEV THRESHOLD

The $(n,2n)$ reactions may be utilized for several threshold detectors. Of the three shown, the C^{12} $(n,2n)$ C^{11} reaction leads to the most sensitive de-

TABLE I. Descriptive and Performance Data for Threshold Detectors

Reaction used	*Threshold in Mev*	*Detector material*	*Induced activity*	*Half-life*	*Detector dimensions*	*Interfering reactions*	*Photo multiplier type*	*Irradiation time*	*Cooling time*	*Counting time*	*Min. detectable flux ($n/cm^2/sec$)*
$U^{238}(n,f)$	1.1	Uranium nitrate crystals	—	—	12.7 cm dia. thin coating	—	6364	30 min	—	30 min	6.6
$P^{31}(n,p)Si^{31}$	2.0	$NH_4H_2PO_4$ in "phoswich"	1.5 Mev beta	160 min	7.6 cm × 4.4 cm "phoswich"	Several (see text)	6363	180 min	30 min	30 min	200
$S^{32}(n,p)P^{32}$	2.0	Fused sulphur	1.7 Mev beta	14.5 days	10.2 cm dia. 0.32 cm thickness	none	6364	15 hrs	5 min	30 min	230
$Ag^{107}(n,2n)Ag^{106}$	9.6	Silver orthophosphate in "phoswich"	2.0 Mev positron	24.5 min	7.6 cm × 4.4 cm "phoswich"	$Ag^{107}(n,\gamma)Ag^{108}$ $P^{31}(n,p)Si^{31}$	6363	60 min	5 min	30 min	350
$I^{127}(n,2n)I^{126}$	10	NaI(Tl) crystal	0.87 Mev beta	13 days	3.8 cm dia. 2.68 cm thickness	$I^{127}(n,\gamma)I^{128}$ $Na^{23}(n,\gamma)Na^{24}$	6292	15 hrs	6 days	30 min	20
$C^{12}(n,2n)C^{11}$	20	Anthracene	0.97 Mev positron	20.3 min	7.62 cm dia. 1 cm thickness	$C^{12}(\gamma,n)C^{11}$ $C^{12}(p,pn)C^{11}$	6363	30 min	5 min	30 min	2.3
$Bi^{209}(n,f)$	50	Bismuth nitrate	—	—	16 cm dia. 38 cm high	—		—	—	—	—

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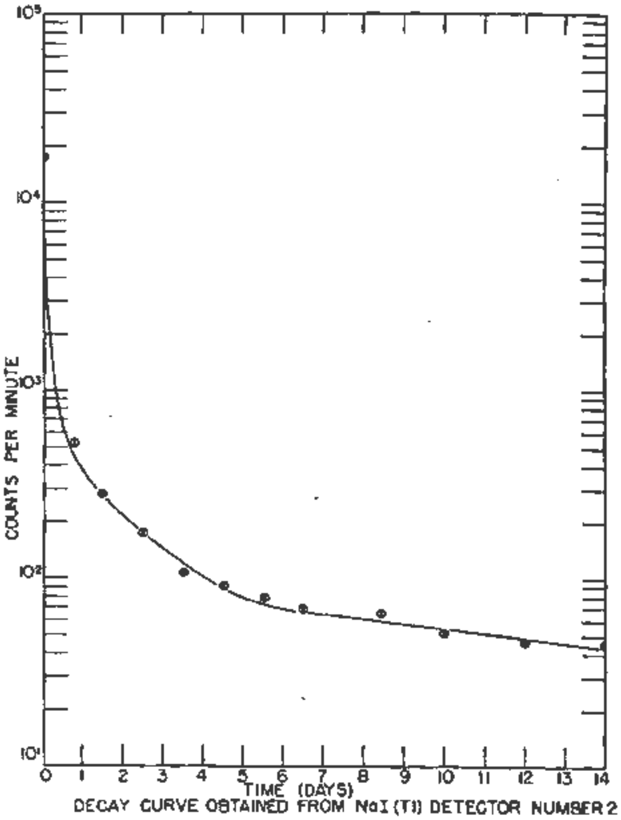
tector. The technique is that of Sharpe and Stafford.[2] A photomultiplier tube is used to count the positrons emitted by C^{11}, produced during previous irradiation of an anthracene crystal mounted on the tube. Except for a 78 mb peak at 42 Mev, the C^{12} $(n,2n)$ C^{11} reaction has a cross section of about 25 millibarns above its threshold which occurs at 20 Mev.[3] We utilized a large anthracene crystal and a discriminator bias that gave a counting efficiency of 95%. Efficiency was determined by calibrating the discriminator in Mev per volt with the internal conversion line of Cs^{137} (625 kev) and then evaluating graphically the fraction of the C^{11} spectrum rejected by the bias used. This detector shows a single decay period of convenient value. The competing (γ,n) reaction cited in Table I is generally unimportant. It has a threshold of 20 Mev but a cross section peak with a height of only 11.6 barns and a half-width of 4.25 Mev. The competing (p,pn) reaction has a variation of cross section with energy similar to that of the C^{12} $(n,2n)$ C^{11} reaction but the value of cross section is only one-sixth that of the $(n,2n)$ reaction. Hence, it too will not usually cause appreciable error in a measurement of neutron flux. It should be emphasized, however, that most threshold detectors have interfering reactions of one sort or another and must, therefore, be used with care. A 30-minute irradiation period suffices for a minimum detectable flux of 2.3 n/cm²/sec. By irradiating and counting for a longer time, fluxes of the order of 1 n/cm²/sec can be detected and measurements can be made in the region of 5 to 10 n/cm²/sec.

THE SODIUM IODIDE DETECTOR—10 MEV THRESHOLD

Another common scintillator material worth evaluating as a threshold detector is sodium iodide,[4] since the I^{127} $(n,2n)$ I^{126} reaction has a threshold of 10 Mev. I^{126} has a half-life of 13.0 days and emits 0.87 Mev betas, 1.26 Mev betas and 1.21 Mev positrons. The use of this scintillator is complicated by the presence of two competing reactions, I^{127} (n,γ) I^{128} with a 25-minute half-life, and Na^{23} (n,γ) Na^{24} with a 14.9-hour half-life. The former can be greatly reduced by covering the crystal with cadmium during irradiation, but the Na^{24} activity must be allowed to decay before a correct count of the I^{126} activity can be obtained. It is essential to plot a decay curve since the cooling time will depend on irradiation time and the neutron spectrum. Such a decay curve is shown in Fig. 1 for a NaI(Tl) crystal irradiated at the cosmotron. A cooling period of 6 days was required and the I^{126} activity was only about 1% of the total activity measured shortly after the end of the irradiation period. The counting efficiency was the same as for the anthracene detector described above and was measured in the same manner. A long period of recovery is necessary between measurements unless an increased background can be tolerated. This detector is of limited health physics value when irradiated for a short time such as 30 minutes, since the minimum detectable flux would then be about 590 n/cm²/sec for a cooling time of 6 hours and a counting time of 30 minutes. However, where averages over a long period of time are of interest, it may be quite useful. The data in Table I are for such a case, where an accelerator ran steadily for 15 hours and it was possible to realize a minimum detectable level of 20 n/cm²/sec.

THE SULPHUR DETECTOR—2.0 MEV THRESHOLD

Sulphur has long been used as a threshold detector but is relatively insensitive as normally used in small quantity with a Geiger counter. The reaction is S^{32} (n,p) P^{32} and the resultant activity has a half-life of 14.5 days. In order to improve the sensitivity, 50 grams of sulphur were cast into the form of a disc, 10.2 cm in diameter and 0.32 cm thick. A photomultiplier tube (Dumont 6364) with a 5 in. diameter face was covered with a mosaic of anthracene chips and used to count scintillations caused by the P^{32} betas. The mosaic was constructed with anthracene chips held together with a transparent glue and was ground to a flat outer surface. Its thickness varied from 1½ mm at the center to 3 mm at the edges. Use of such a thin detector resulted in a background counting rate of only 97 cpm. The efficiency for detecting disintegrations in the 50 grams of sulphur was found to be 5% by comparing the observed counting rate due to a Ra-Be source, with the P^{32} activity computed from the known sulphur cross section and Ra-



DECAY CURVE OBTAINED FROM NaI (Tl) DETECTOR NUMBER 2

Figure 1

Be spectrum. Even with the large disc and counter, the sulphur detector turns out to be rather insensitive for hazard evaluation purposes. The case shown in Table I was for 15 hours irradiation, the minimum detectable flux being 230 n/cm^2/sec based on a cross section of 230 mb for the S^{32} (n,p) P^{32} reaction.[5] The sulphur detector has the virtue of being free from disturbing reactions. Figure 2 shows the sulphur disc and photomultiplier tube. The anthracene mosaic is covered with a 1 mil thick aluminum foil to exclude light.

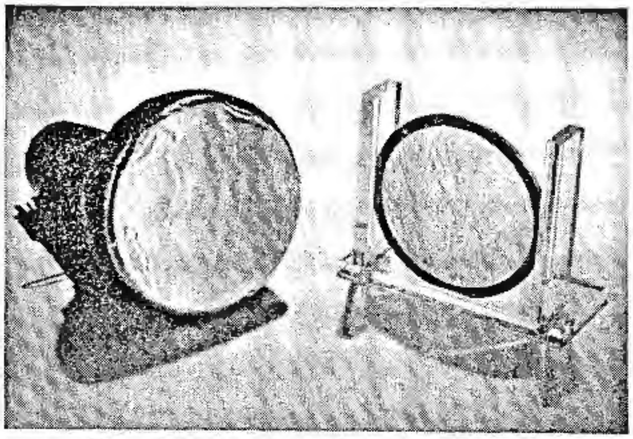


Figure 2. Sulphur disc and photomultiplier tube

THE PHOSPHORUS DETECTOR—2 MEV THRESHOLD

In order to test a phosphorus threshold detector it was desirable to use a compound in the form of a powder. This was done by fabricating a slotted plastic scintillator as shown in Fig. 3. The scintillator material used was p-terphenyl in polystyrene. The ends of the slots were covered with transparent tape, an aluminum cap was fitted over the detector and the whole assembly was wrapped with black tape to exclude light. The term "phoswich," i.e., phosphor sandwich, is used for this device after Wilkinson[6] who used the term for a sandwich-like assembly of fast and slow scintillators. A picture of one of our "phoswiches," before being filled and covered with tape, is shown in the foreground of Fig. 3.

The substance used for the phosphorus detector was di-H ortho-ammonium phosphate, chosen because it was white and not deliquescent. The product activity, Si^{31}, has a half-life of 160 min. A cross section of 70 mb[7] and a background counting rate of 150 cpm were used in computing the minimum detectable flux which was found to be 200 n/cm^2 per sec. Possible interfering reactions are $P^{31}(n,2n)P^{30}$, $N^{14}(n,2n)N^{13}$, $C^{12}(n,2n)C^{11}$ and $P^{31}(n,\alpha)Al^{28}$. However, these are all of considerably shorter half-life than Si^{31} and will decay out if a cooling time of an hour or so is allowed. A decay curve for irradiation at the cosmotron is reproduced in Fig. 4 and shows a pure 160-minute decay after 30 minutes.

OTHER "PHOSWICH" DETECTORS

The "phoswich" was also used for sulphur with the expectation that increased sensitivity could be achieved. In this case, molded slabs of sulphur were inserted in wells machined in a cylinder of plastic scintillator as shown next to the photomultiplier tube in Fig. 3. The thickness of the "phoswich" was ¾ in. and was chosen so that the weight of sulphur would be about the same as was used in the sulphur disc described above. After correcting for the differences in diameter used for disc and "phoswich," it was found that the "phoswich" was about 20% more sensitive than the disc. Experiments on sulphur with a thicker "phoswich" have not been carried out but it is clear that a considerable additional improvement in sensitivity is possible.

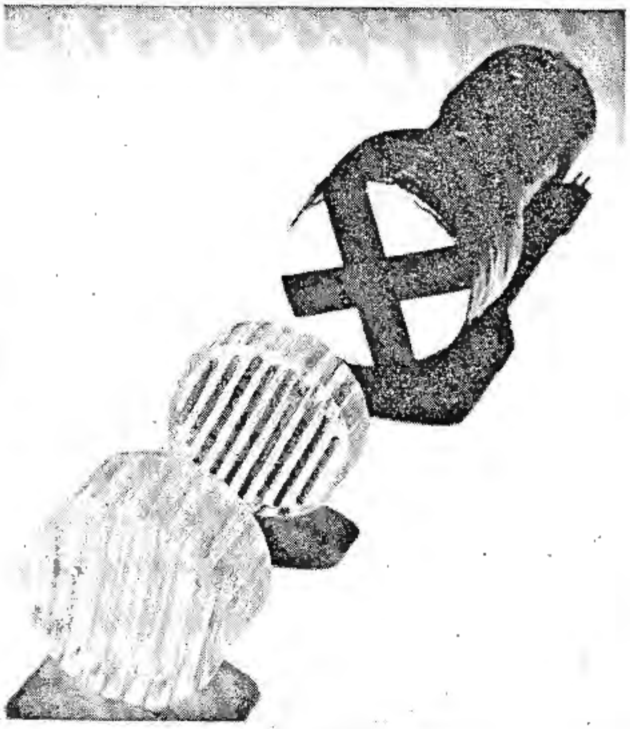


Figure 3. "Phoswich" detectors

Silver orthophosphate in a "phoswich" was considered as an alternative to NaI(Tl). It has a threshold of 9.6 Mev for the Ag^{107} $(n,2n)Ag^{106}$ reaction and a very convenient half-life of 24.5 minutes. The detector was not actually calibrated but, based on experience with other detectors, should have an efficiency of 8–10% and a minimum detectable flux value of 350 n/cm^2/sec. The Ag^{107} $(n,\gamma)Ag^{108}$ interfering reaction is of no concern since it has a half-life of only 2.3 minutes and can be reduced by a cadmium shield. However, the $P^{31}(n,p)Si^{31}$ interfering reaction may be expected to result in more activity than the reaction of interest. A correction for this could be made by plotting a decay curve since its half-life is 160 minutes. This detector would recover much more rapidly than the NaI(Tl) detector and can serve a dual purpose in that values of flux above both 2.0 and 9.6 Mev can be derived from a single detector by proper interpretation of the decay curve.

We have not yet investigated the possibilities opened up by use of the "phoswich" as a container for powders and liquids not otherwise amenable to scintillation counting but feel that the device will prove to be useful in many circumstances.

THE URANIUM DETECTOR—1.1 MEV THRESHOLD

A very sensitive threshold detector was obtained by utilizing the 1.1 Mev threshold for the fission of U^{238} by fast neutrons. The method was similar to that described by Koontz, *et al.*[8] A thin layer, consisting of equal parts of uranium nitrate and ZnS (Ag) mixed with zapon, was painted on the end window of a Dumont type 6364 photomultiplier tube. Pulses due to the uranium alpha particles were biased out and only those due to uranium fission were counted. In this case, of course, counting was done while the detector was being irradiated. Calibration with a Ra-Be source gave an efficiency of 0.001%. If we assume a background counting rate of 1 count per minute, the minimum detectable flux, for a 30-minute period, amounts to 6.6 n/cm²/sec. For a pulsed accelerator, background can be reduced by gating the counter. Thus, we have a rather sensitive detector, although the counting rate is still rather low for ratemeter service where one needs at least 50 counts per minute even with a fairly long time constant. For natural uranium the error due to thermal fission will be low in many cases but should be considered where there is an appreciable thermal flux component. Thorium can be used instead of uranium, in which case thermal fission will be even less important. The reader should consult a paper by Rohr, *et al.*, in regard to the use of uranium coatings in proportional counters for fast neutron detection.[9]

THE BISMUTH DETECTOR—50 MEV THRESHOLD

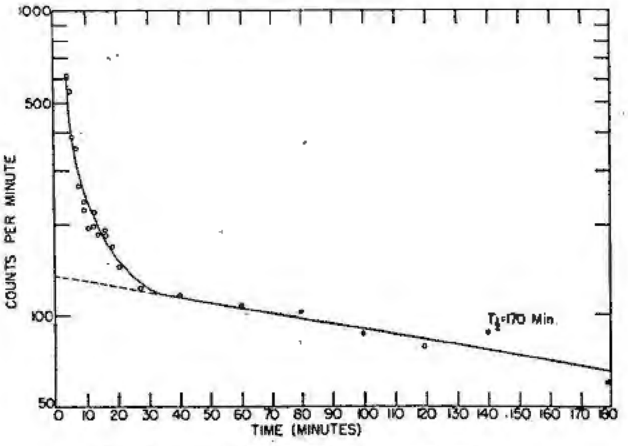
A large fission chamber for measuring neutron flux above 50 Mev has been described by DeJuren.[10] Using the published values of efficiency and plate area, we obtain a counting rate of 0.04 cpm per n/cm²/sec. Since a very low background can be obtained with such a counter, flux levels of a few neutrons per cm²/sec can be detected by counting for an hour or so. Our definition for minimum detectable flux density breaks down at such low counting rates so no value is given in Table I. A flux of 100 n/cm²/sec would be determined within about 10%, at the 90% confidence level, by a 1-hour count.

In order to improve efficiency, it has been decided to utilize an available 36-liter tank with four type-5819 photomultiplier tubes at each end, and to fill the tank with liquid scintillator. This scintillator will consist of three grams of α-napthylphenyloxazole per liter of solvent consisting of 70% xylene, 24% napthalene and 6% triphenylbismuthene by weight, as utilized by Kallmann and Furst.[11] The solution will contain 1340 grams of bismuth in a geometry such that a high percentage of the bismuth fissions can be detected by the photomultipliers. A scintillation detection efficiency of 50% or better is anticipated for fission pulses. This would give a counting rate of 70 cpm per n/cm²/sec. Background has not been determined but will be very low, particularly in a situation where the counter can be gated. An extremely sensitive detector of high energy neutrons should result. For practical survey applications, a much smaller tank would suffice. A 10-liter spherical container with 5 in. photomultiplier tubes on opposite radii has been constructed and will be tested along with the larger apparatus.

FAST NEUTRON SURVEY METERS

In recent years, techniques for making fast neutron surveys have been greatly improved, especially where it is desired to measure neutron flux in the presence of gamma radiation.[12] A proportional counter, developed by Hurst, *et al.*,[13] has the great advantage of a meter deflection proportional to the dose from 0.2 to 10 Mev. However, it is directional and not as sensitive as is desirable. The most sensitive range reads full scale for 50 mrem/hr, or about 350 n/cm²/sec at an energy of 1 Mev. However, an excessively long time constant is required for this range so that conveniently usable sensitivity is considerably less than this value. Metal-walled proportional counters, filled with methane, have been widely used and show a response roughly independent of neutron energy over a range from 0.5 to 3 Mev.[14] Sensitivity is somewhat low for ratemeter application, but good data at low levels can be obtained by using a scaler and counting for 5 or 10 minutes. Counters of this type used at this laboratory have a volume of 410 cc and give a counting rate of 60 counts per minute for a flux of 20 Po-Be n/cm²/sec. Somewhat greater sensitivity can be realized by use of proportional counters with uniform hydrogenous walls. Moyer and his associates have worked out the theory of such counters and have developed two practical instruments.[12] The response is highly dependent on neutron energy but closely approximates proportionality to neutron energy flux in the range from 0.5 to 20 Mev.

The scintillation type meter developed by Hand-



DECAY CURVE OBTAINED WITH THE $NH_4H_2PO_4$ PHOSWICH

Figure 4

loser and Higinbotham[15] is compact, reliable and quite sensitive. A molded zinc-sulphide-in-lucite scintillator described by Hornyak[16] is used on a 2 in. diameter photomultiplier tube and gives full scale deflection for 300 1-Mev n/cm^2/sec. This sensitivity is realized with a conveniently short time constant so that measurements can be made at levels of 20–30 n/cm^2/sec corresponding to the 40-hour per week maximum permissible exposure limit for neutrons of a few Mev energy. Similar scintillators have been made by Seagondollar, *et al.*[17] using self-curing bioplastic liquid. Emmerich has achieved a similar result with a paraffin-ZnS(Ag) mixture and lucite strips as light pipes.[18] Recently, Handloser has developed an instrument similar to that described in Reference 15 using a 5 in. diameter photomultiplier tube with a 5 in. × 1¼ in. scintillator. The larger scintillator results in a 5-fold increase in sensitivity. In designing the final instrument however, he chose to improve instrument stability somewhat at the expense of sensitivity so that full-scale deflection for 1-Mev neutrons corresponds to 100 n/cm^2/sec. This provides the sensitivity needed for measurements well below the maximum permissible level. Energy dependence is similar to that of the hydrogenous walled counters referred to above.

THERMAL NEUTRON DETECTORS

The problem of providing sensitive detectors for thermal neutron measurements is relatively easy and has found a variety of solutions. Not only is the maximum permissible flux for 40-hr-per-week exposure 50 or 60 times the fast neutron value, but there are a number of substances such as boron and lithium with very large cross sections for thermal neutron induced reactions of practical utility. Proportional counters lined with boron compounds or filled with boron-containing gas have been used for many years and are capable of great sensitivity, for instance 1800 cpm per n/cm^2/sec for a large counter filled with enriched BF_3 under pressure.[19] For practical health physics survey work, much lower sensitivity is required and ratemeters using smaller tubes filled with unenriched BF_3 may be utilized.

Recently, attention has been focused on the use of phosphors for thermal neutron detection.[20, 21] Detailed studies have been made of the use of lithium iodide crystals,[22, 23] a solution of boron esters in liquid scintillator,[24] a mixture of BN or B_2O_3 with ZnS(Ag)[25, 26] fused discs of boric oxide and scintillator,[27] and cadmium plates in scintillator.[28] All of these are quite sensitive and a number of them could presumably be utilized in constructing useful survey instruments.

We should not leave the subject of thermal neutron detection without mentioning lithium borate loaded films[29] and indium foils.[30] The former are extremely sensitive while the latter can be used to detect fluxes as low as 5 n/cm^2/sec if 2-in. diameter foils are utilized and they are counted with good efficiency.

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A Gamma-Ray Insensitive Semi-Conductor Fast Neutron Dosimeter Using Single Crystal Germanium

By B. Cassen,* USA

Studies of the biologically interesting depth dose of fast neutrons in animals or phantoms is not at present readily accomplishable by any simple method. This is especially so when the fast neutron flux is contaminated with gamma radiation and slow neutrons. Tissue equivalent or near tissue equivalent ionization chambers, although of great value in rep standardization calibrations, become difficult to use and interpret in depth dose studies. They cannot be made small enough for use with smaller animals except in a crude sort of way. They frequently exhibit electrical insulation leakage and polarization problems, and associated complexities of electronic equipment.

Radioactivation detectors are insensitive to gamma radiation. The most commonly used fast neutron radioactivation detector is sulfur. It has a threshold in the neighborhood of 3 Mev, and accordingly would give no response to a fast neutron spectrum with a high energy cut-off below 3 Mev. Other threshold detectors, both of the (n, p) reaction type and the fission reaction type, have been used. They usually have some disadvantage for ordinary dosimetry, such as short half life or difficult or restricted availability.

The work of Lark-Horowitz[1] and collaborators has shown that the electrical conductivity of certain semi-conductors is permanently modified by easily attainable doses of fast neutrons. The effect is large in highly purified (intrinsic) single crystal germanium. This material is now commercially available from several companies engaged in transistor development. It is not appreciably affected by exposures to very large doses of gamma radiation. A program was initiated at UCLA about three years ago to determine whether the change in conductivity effect could be used in dosimetric studies of fast neutrons, especially in a mixed neutron-gamma radiation field. Initially, the practical limits of sensitivity were unknown. They obviously depended, among other factors, on the precision and reproducibility of resistance measurements, the purity and properties of the available germanium, and the resistance measurement bath temperature selected for optimum results.

After much trial and error, and a surprisingly large amount of effort to eliminate erratic effects, a practical dosimeter has been developed and utilized in a series of tests. They are still subject to improvement and possible increases in sensitivity. It has been found that the major problem is that of quality control and specification of the germanium. Different lots presumably prepared in the same way have been found to differ considerably in their behaviour and reproducibility. Some lots behave perfectly and have been re-used several times; others are unuseable on account of an erratic behaviour, the nature of which is still not fully understood. It is probably connected with so-called "surface channeling."

The present form[2] of the dosimeter consists of a wafer of intrinsic germanium 4 mm long, 2 mm wide, and 0.75 mm thick. Short lengths of copper wire 0.15 mm in diameter are soft soldered to the ends. This small unit can be enveloped in plastic and inserted in an incision or body cavity of even a small animal. However, it is usually encapsulated in a thin-walled fluorinated hydrocarbon tube 2.5 cm long and 2.8 mm inside diameter. Hollow rivets 6 mm long are pressed in the ends of the plastic tube, the wire leads are brought out and soldered in the rivet. The solder closes the ends of the rivets, making a fairly moisture-proof unit.

In the simplified semi-conductor model widely used, the effect of fast neutrons in changing the electrical conductivity can be described as follows: The fast neutrons produce germanium atom recoils which dislodge neighboring germanium atoms from their normal lattice sites, producing lattice defects which act as electron traps. The holes in the filled Fermi band act as positive carriers, giving the crystal additional conductivity. At ordinary temperatures, this additional conductivity is masked by the normal conductivity of the semi-conductor produced by electrons put in conduction levels by thermal excitation. By lowering the temperature of resistance measurement to minimize the thermal excitation, more sensitivity can be obtained for measuring the effect of interest. In practice, it has been found most convenient to measure resistance before and after fast neutron exposure in a bath at −78°C, the temperature attained by mixing solid carbon dioxide with a low viscosity electrical insulating fluid. A silicone fluid has been found quite suitable (Dow Corning 200, 1 centistoke). The fluid bath is contained in a wide mouthed Dewar flask. However, there is enough heat transfer to keep the bath bubbling slightly from the evapora-

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tion of carbon dioxide. The crystals that we have so far found to have the best performance have a resistance at —78°C of from 3000 to 8000 ohms before exposure. So far, exposures have been made on five different sources of fast neutrons. The results have been expressed tentatively in rep based upon a calibration obtained on the Oak Ridge cyclotron. The calibration of the cyclotron target neutron output was that obtained with the proportional counter method of G. S. Hurst. The neutron spectrum of this target was originally supposed to resemble a fission spectrum, but later was shown to be of higher energy. This throws our original, presumably fission spectrum calibration in some doubt, as the energy dependence of the germanium cross section does not parallel that of hydrogen, upon which the definition of a rep essentially depends. However, on the basis of the tentative calibration, gradual improvements have enabled the lower level of definitely measurable dose to get well down into the biologically most interesting range (below 1000 rep). The response of the change of conductivity to exposure was found to be linear from about 150 rep up to tens of thousands of rep. The line, however, did not go through the origin, but intersected the exposure axis at about 150 rep. At lower doses, evidence was obtained of a slight increase in resistance instead of decrease. This effect is easily explainable as arising from the originally, slightly n-type material going over to p-type. The observation suggests that linearity can be obtained in the lower dose range by sufficient pre-exposure of the crystals. This has been found to be the case. In fact we have used crystals over and over again without annealing, as long as they did not receive a tremendous dose. Some other batches of germanium go up in resistance in the usual exposure range and do not start decreasing in resistance until very large doses have been received. Why the germanium varies so remarkably is now being studied.

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Fast Neutron Dosimetry

By G. S. Hurst, R. H. Ritchie and W. A. Mills,* USA

Three methods of measuring the tissue dose due to fast neutrons have been developed at the Oak Ridge National Laboratory. All three methods have yielded practical instruments, capable of accurately measuring a fast neutron dose, even if a gamma dose rate much greater than the neutron dose rate is present. This paper will discuss the three methods, after a brief discussion of the fundamental physical effects of fast neutrons on tissue.

The principal interactions of fast neutrons with living tissue are the elastic collisions with hydrogen, carbon, nitrogen and oxygen atoms. The energy imparted to these atoms is spent by ionization and excitation. An important quantity for the radiobiologist and health physicist to measure is the amount of energy transferred per gram of tissue. In the case of mixed fields of neutron and γ-radiation, the energy per gram of tissue contributed by each radiation source should be separately measured, since both the biological effects and the shielding considerations are different for each radiation.

The accepted unit for measuring tissue dose is the roentgen equivalent physical (rep) and is defined as that amount of radiation which is absorbed in tissue to the extent of 95 ergs per gram. As a result of a decision of the Units Committee of the International Commission on Radiological Protection at its meeting in Copenhagen in July 1953, a new unit was defined in terms of 100 ergs/gm of any medium and called the "rad."

For fast neutrons, where the dose is due entirely to elastic collisions with the medium elements, the dose per neutron per cm^2 may be computed for any medium from the formula,

$$D(\text{Mev/gm}) = E\, \Sigma_i \sigma_i f_i Q_i \qquad (1)$$

where E is the neutron energy in Mev, σ_i is the cross-section (cm^2) of the i^{th} type of atom, f_i is the average fraction of energy that is lost by the neutron during its collision with the i^{th} kind of atom, and Q_i is the number of atoms of element i in a gram of the medium. Assuming isotropic center of mass scattering,

$$f_i = 2\, mM/(m + M)^2 \qquad (2)$$

where m is the neutron mass and M is the mass of the recoil atom.

Specifying the medium as average wet tissue[1] having a composition by weight of H = 10 per cent, N = 4 per cent, O = 73.6 per cent and C = 12 per cent, the dose has been calculated. The results are shown in Fig. 1 where a comparison is made with the ethylene (C_2H_4) dose curve (cross sections taken from Adair).[2]

PROPORTIONAL COUNTER METHODS OF MEASURING DOSE

The rep does not lend itself readily to measurement. In principle it may be measured by the heating effect produced in tissue; however, the temperature rise is usually too small to be measured. This leaves one with the problem of inferring the energy deposited from ionization measurements. Utilization of the Bragg-Gray cavity principle[3] leads to an instrument which measures the number of ion pairs produced per gram of gas filling a cavity which is completely surrounded by thick, solid material having an atomic composition identical with that of the gas. If the gas and wall are both "tissue equivalent" then the number of ion pairs per gram of tissue is known. One may then relate the amount of ionization produced to the amount of energy absorbed; however, this is not easy to do since in some gases the value of W (number of electron volts per ion pair) depends on particle velocity,[4] and it certainly varies with type of particle.[5] The variations with particle velocity are small, but the change with particle type is sometimes significant.

It is noted (Fig. 1) that tissue and ethylene have the same energy dependence essentially and for this reason it can be said that ethylene is tissue equivalent for fast neutrons. From a practical viewpoint the result is important, since ethylene may be surrounded by polyethylene to achieve a perfect Bragg-Gray design.

In order to discriminate against gamma radiation, the ionization is measured with a proportional counter. If the dimensions of the counter are small compared with the range of high-energy electrons, the atom recoil pulses may be counted above the γ-level. Under special conditions, which will be discussed later, the output pulse height of a linear amplifier is proportional to the number of ion pairs causing the pulse, regardless of the position or orientation of the ionizing track. An addition of pulse heights then measures the dose due to fast neutrons.

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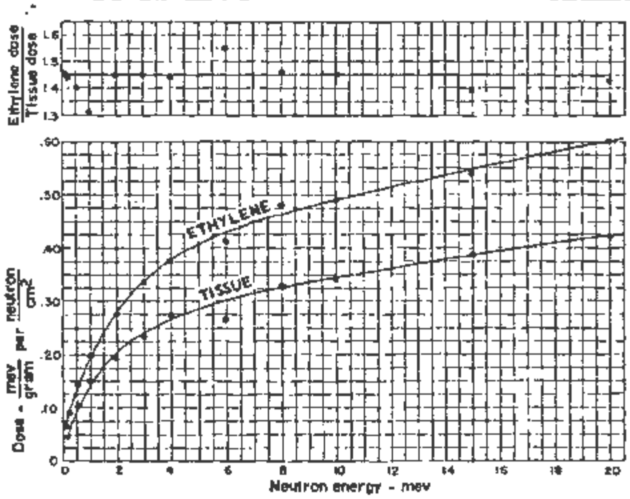

Figure 1. Fast neutron first collision curves for tissue and for ethylene

There are two reasons why the pulse height in a proportional counter depends on the point where a single ion pair is formed. The first reason is that the gas amplification can vary with length along the wire. This may be prevented by a field tube arrangement developed by Cockcroft and Curran.[6] The other reason is that the electron can become attached to form a heavy ion, so that it can never gain enough energy between collisions to produce gas amplification even when very near the wire. Attachment is most severe for contaminating gases such as oxygen or water vapor.

Even if the pulse height does not depend upon the point of creation of a single ion, it may depend on the angle that the ionizing track makes with the wire unless the time constant of the linear amplifier is of the correct value. If the track makes an angle with the wire (track extension), electrons arrive at the wire over a period of time (τ_0), and the counter pulse is the resultant of pulses due to single electrons and thus has a rise time which increases with the collection time of the outermost electrons. The results[7] shown in Fig. 2 are for the case where the amplifier is of the pure RC-RC type with equal differentiating and integrating time constants t_1. The output does not change appreciably for a ratio of τ_0/t_1 as large as unity. Therefore the height of the amplifier output pulse is proportional to the number of electrons, independent of track orientation, so long as the collection time of the outermost electrons is not much greater than the amplifier time constant.

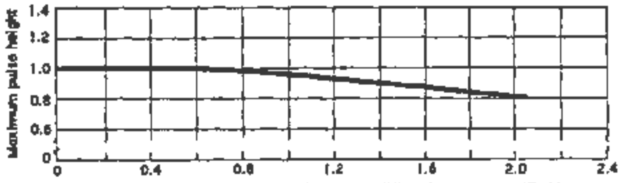

Figure 2. Variation of amplifier pulse height with the ratio τ_0/t_1

In order to discriminate against γ-radiation it is necessary to apply a bias on the instrument. In cases where the γ-intensity is so low that pulse "pile-up" does not occur, the bias is low, but when the intensity is high the bias may be as much as 100 to 200 kev. This sets a lower limit to the neutron energy that can be measured, and even for energies above the bias some of the recoils due to neutrons are lost. Energy is lost under the bias for two reasons. (1) For neutrons of energy E, recoil protons are produced with energies from 0 to E. Those with energy less than the bias B are not counted. (2) A proton having energy greater than B can leave the sensitive volume before losing all of the energy B. In practice the loss due to (1) far exceeds the loss due to (2), unless the neutron energy is large in which case both (1) and (2) are small. Therefore only the loss due to (1) need be calculated.

Hydrogen scattering is isotropic in the center of mass system, hence the energy of the recoil leaving at angle θ (Fig. 3) is $E \cos^2 \theta$. The recoils that are counted are those defined by the cone formed by θ_1, where $B = E \cos^2 \theta_1$. Also, the number of recoils in the cone θ_1 is $N(\theta_1) = \sin^2 \theta_1$, and the number of recoils having energy less than B is therefore $(1 - \sin^2 \theta_1) = B/E$. The average energy of these is $B/2$, and the average for the entire number of recoils is $E/2$; hence the energy lost under the bias goes as $(B/E)^2$ which is small if B is only moderately less than E.

The analogy between the proportional counter and the ionization chamber is then complete provided the gas amplification of the counter is measured. This is easily accomplished by means of an α-source built into the counter. Therefore one can calibrate the neutron dose in terms of the energy of the α-source if the sensitive volume of the counter is known.

Many kinds of proportional counters following these principles have been developed. Two examples will be given. A design[8] that is useful for measuring the ethylene dose in absolute units is shown in Fig. 4. The active volume is determined by means of field tubes. Alpha particles from Pu^{239} are admitted into the active volume by removing the source cover with an external electromagnet. The results in Fig. 5 were

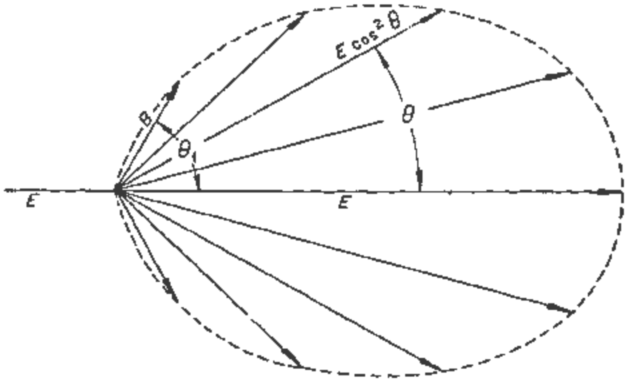

Figure 3. Neutron scattering in the laboratory system

obtained by flowing the ethylene through the counter without purification. Medical grade ethylene seemed to be pure enough if the counter is used as a flow device. The integral α curve has a maximum when differentiated at 165 volts, thus this point is taken as 5.14 Mev, the energy of the α-particle. The dose rate due to neutrons is given by the area under the integral pulse height curves and this area may be calibrated in terms of Mev per second by means of the α calibration. In the case of the Po-boron source the area under the curve is 14.1 Mev/sec. If this is corrected for scattering (by means of a shadow cone measurement), the number becomes 13.1 Mev/sec. Similarly the area under the Po-Be curve is, after correcting for scattering, 4.0 Mev/sec.

The mass of ethylene in the counter's active volume is 0.061 gm, and the flux (computed by the inverse square law, assuming that the neutron strength is known) corresponding to the above energy dissipation is 718 n/cm^2/sec for Po-B and 172 n/cm^2/sec for Po-Be. Thus, we have 0.30 Mev/gm ethylene/n/cm^2 for Po-B, and 0.38 Mev/gm ethylene/n/cm^2 for Po-Be.

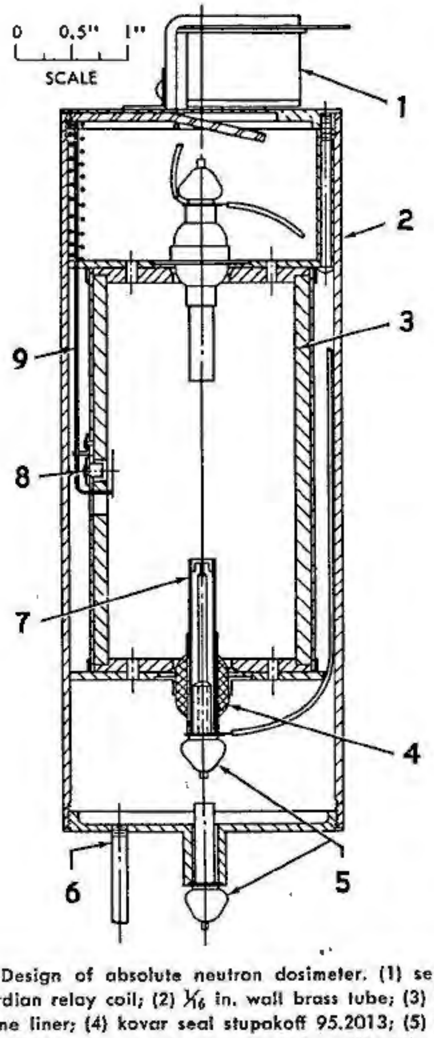

Figure 4. Design of absolute neutron dosimeter. (1) series 200 6 v.d.c. guardian relay coil; (2) $\frac{1}{16}$ in. wall brass tube; (3) $\frac{1}{8}$ in. wall polyethylene liner; (4) kovar seal stupakoff 95.2013; (5) kovar seal stupakoff 95.2040; (6) gas inlet; (7) field tubes; (8) alpha source; (9) alpha source shutter

If the Po-B neutron spectrum[14] is weighted by the first collision dose curve for ethylene (Fig. 1), it is found that one neutron per cm^2 dissipates 0.30 Mev/gm ethylene for Po-B. Weighting the spectrum[15] for Po-Be by the first collision curve gives 0.34 Mev/gm ethylene for Po-Be. Thus, the agreement between the measured dose rate and calculated dose rate is good for both sources.

Figure 6 shows a small counter which is useful in making phantom measurements. The volume of ethylene is completely surrounded by a polyethylene liner; hence the conditions for application of the Bragg-Gray principle are fulfilled. Since the active volume is not known and it contains no α source, this counter is calibrated by using a "standard" neutron source or by comparison with the standard counter just described. The outstanding advantage is that it may be sealed off and used as a compact unit.

PULSE INTEGRATION

With the above proportional counters, the fast neutron dose is obtained by integration of pulses due to recoil atoms, after discriminating against the pulses due to electrons. Figure 7 illustrates the principle of an integrator[9] designed such that the number of pulses recorded is proportional to the summation of pulse heights. *A*, *B*, *C*, and *D* are discriminator tubes biased so that the pulse heights required to make the tubes conduct are, for example, 5, 10, 20, and 40 volts, respectively. The discriminators feed into the one, two, four, and eight counts stages of a usual binary type scaling system. Every time the *A* tube conducts, because of the arrival of a five-volt pulse, one count is added to the scaling unit. Each time a ten-volt pulse arrives, both *A* and *B* conduct. This pulse adds three counts if the number one indicator is not on. If the number one indicator is on, it will be erased while the number two indicator records the pulse from *B*, giving a net count of only one. Thus, the average is two counts for each ten-volt pulse.

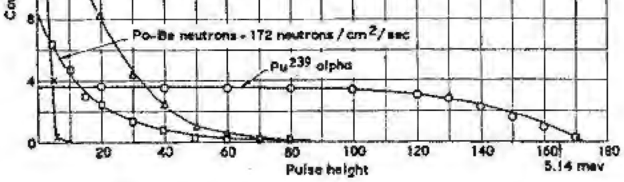

Figure 5. Pulse distribution curves for the absolute counter

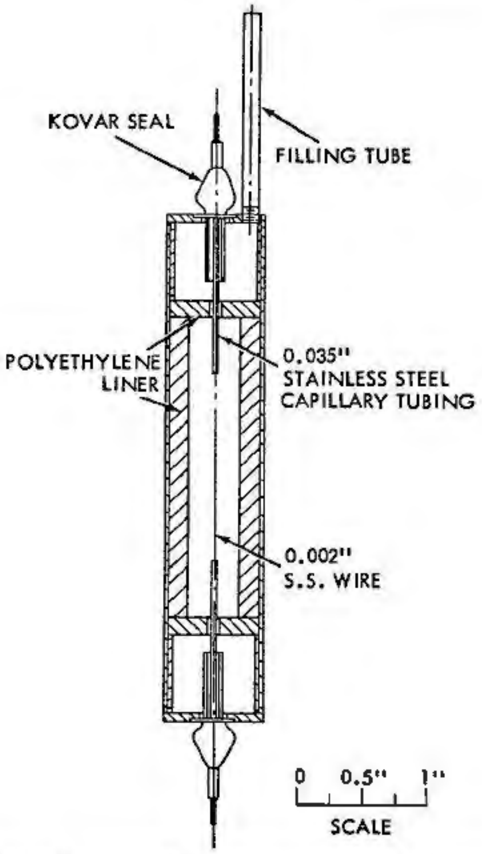

Figure 6. Design of the phantom neutron dosimeter

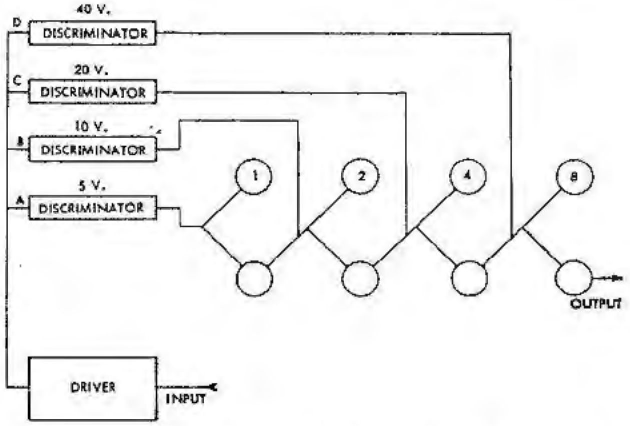

Figure 7. Pulse integration with one binary scaling unit

Similar analysis shows that each 20-volt pulse gives an average of four counts, while the 40-volt pulses give an average of eight counts. Then, the only discriminator that is effective is the one whose bias is less than and nearest to the height of the pulse. The discriminators of lower bias levels merely add and subtract counts from the system which cancel for either a regular distribution of equal height pulses or for a random distribution of any assortment of heights. Therefore, it is not mandatory to use anticoincidence circuits to prevent the lower level discriminators from adding counts to the system.

In an actual instrument two scaling units were used so that the consecutive discriminator voltages differed by smaller amounts. This is showo schematically in Fig. 8. Here tubes *A*1 and *B*1 are set at the same voltage so that the first interval will give two counts. When *A*2 conducts three counts are obtained. When the last tube to conduct is *B*2, *A*4, *B*4, *A*8, or *B*8, the counts recorded are 4, 6, 8, 12, or 16, respectively. In order to obtain linearity between the numbers of counts and pulse height, the average voltage for the various channels can be set as shown in Table I. The table also gives values for the minimum and maximum voltages necessary to obtain these averages. The discriminators are set at the voltages shown in the column of minimum voltages.

The instrument described above was used to integrate the pulse spectrum shown in Fig. 9. This is a typical spectrum obtained by irradiating a recoil proton proportional counter with fast neutrons. Unless there is a gamma-radiation field of 2 r/hr or more, very few electron pulses will be equal to or greater than 6 volts. Therefore, to discriminate against gamma radiation, the first discriminator is set at 6 volts, and the remaining discriminators are set according to column 4 of Table I. These are indicated by the vertical lines on the curve. The dose represented by the spectrum is proportional to the area under the solid curve obtained by using an A1 linear amplifier and integral pulse-height selector.[10] The dose measured by the automatic integrator is proportional to the area under the dashed line, obtained by joining the points corresponding to discriminator settings with straight lines. For this spectrum, the integrator should overestimate by about 6 per cent.

A more elaborate design uses four binary scalers (Fig. 10), with discriminator voltages set as shown in Table II. In Fig. 10 the numbers 1 through 13 refer to the discriminators as listed in Table II. The accuracy of the four-scaling-unit integrator may be

TABLE I

Disc. no.	*No. of counts*	*Voltage*		
		Average	*Minimum*	*Maximum*
*A*1*B*1	2	8	6	10
*A*2	3	12	10	14
*B*2	4	16	14	18
*A*4	6	24	18	30
*B*4	8	32	30	34
*A*8	12	48	34	62
*B*8	16	64	62	—

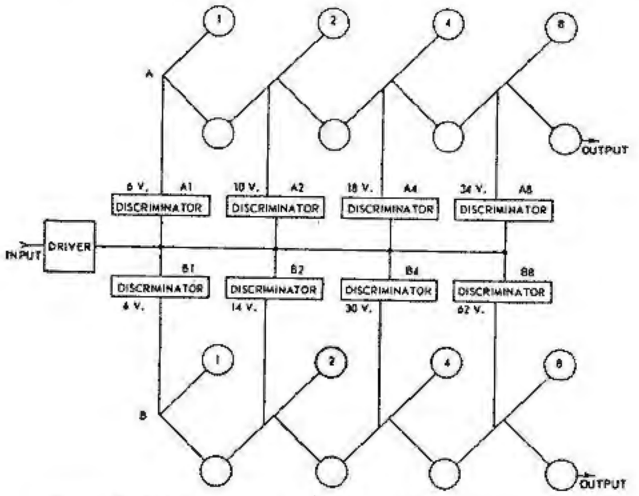

Figure 8. Pulse integration with two binary scaling units

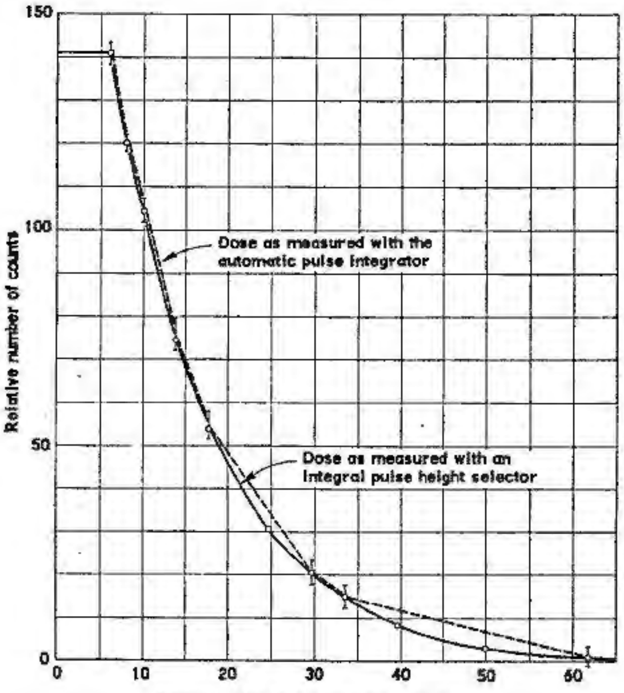

Figure 9. Test of the two unit pulse integrator

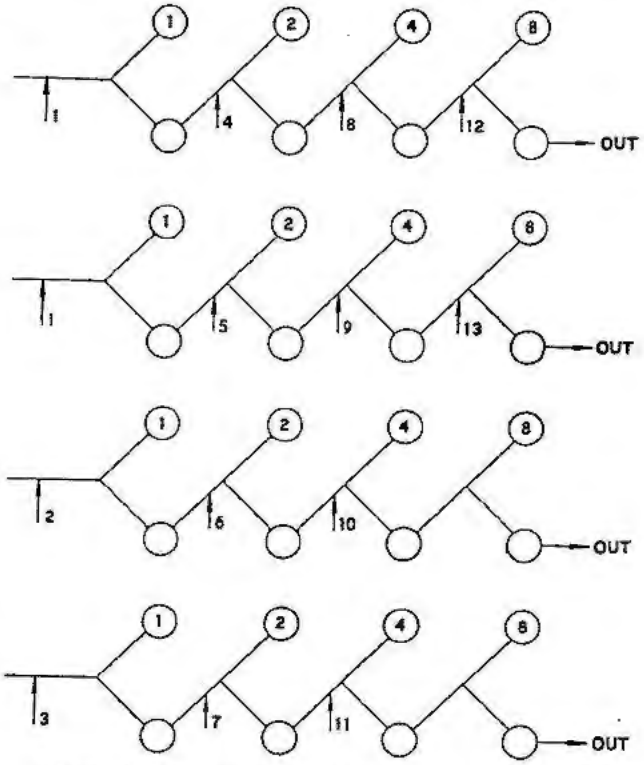

Figure 10. Pulse integrator with four binary scaling units

judged from Fig. 11, where the same pulse spectrum as in Fig. 9 is used. Here the area under the dotted curve is essentially the same as the area under the solid curve.

Area 1 is proportional to the neutron dose which is measured above the gamma discriminating bias. As mentioned previously, some energy is lost under the bias, and area 2 represents this energy. The new integrator is capable of automatically extrapolating the curve to zero volts, hence including area 2, and thus correcting for the energy lost under the bias. Success of the method depends on the empirically established fact that the count rate vs pulse height curves are linear in the region of low pulse heights for a wide range of neutron energies. If the curves are linear, a separate measurement of the number of pulses with heights greater than V_1 but less than

TABLE II

Discriminator no.	*Counts*	*Voltages*		
		Average	*Minimum*	*Maximum*
1	2	8	6	10
2	3	12	10	14
3	4	16	14	18
4	5	20	18	22
5	6	24	22	26
6	7	28	26	30
7	8	32	30	34
8	10	40	34	46
9	12	48	46	50
10	14	56	50	62
11	16	64	62	66
12	20	80	66	94
13	24	96	94	—

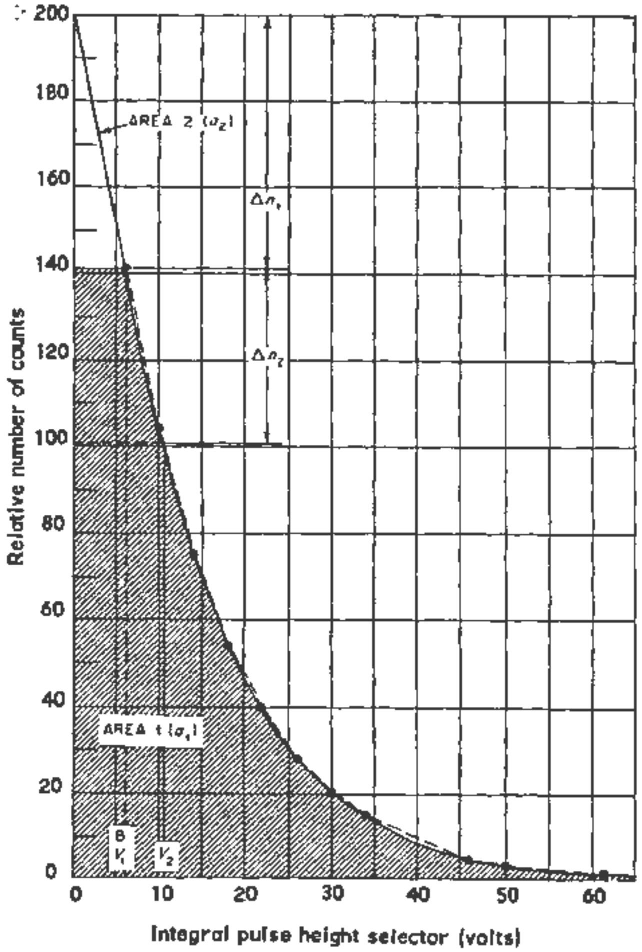

Figure 11. Test of the four unit pulse integrator

V_2, can be used to measure the energy lost under the bias, B. The area representing the energy lost is a_2 (Fig. 11) where $a_2 = B/2.\Delta n_1$ (volts). As seen in Table II, each count on the integrator represents 4 volts. Thus the area a_a corresponds to $B/8.\Delta n_1$ integrator counts. Let us require that the number of counts Δn_2 have a weight of unity, so that extrapolation can be made by simply setting a single channel pulse analyzer so that it counts Δn_2. Thus, $\Delta n_2 = B/8.\Delta n_1$. But $\Delta n_2 = \Delta n_1 (V_2 - V_1)/B$, therefore, $V_2 - V_1 = B^2/8$, gives the required value for V_2 and V_1. As an example, set $B = 6$ volts, and put $V_1 = B = 6$ volts. The required value for V_2 is 10.5 volts, therefore a single channel analyzer set over the range 6 volts to 10.5 volts, and connected to a separate scaling unit serves as an extrapolator. Since both V_1 and V_2 are set with the condition $V_2 > V_1 \geqslant B$, the instrument does not respond to gamma rays.

COUNT RATE DOSIMETER

Another method[11] developed utilizes a proton recoil proportional counter of special design whose count rate response to a collimated beam of neutrons is proportional to the tissue dose. The energy response of the counter is determined by the three sources of recoil protons shown in Fig. 12. Calculations were made of the probability that neutrons of energy E could cause a recoil proton to lose energy greater than B (the bias energy needed to discriminate against gamma radiation) in the counting volume. The energy response curves for the three sources of protons are such that when added for the illustrated proportions of hydrogenous materials, the tissue first collision dose curve is approximated. The response has been checked experimentally with monokinetic neutrons and agrees well with the theoretical curve. Gamma-ray interactions in the counter are easily discriminated against since the ionization in the counter due to secondary electrons is much less than the proton ionization. A portable instrument employing this counter is available commercially.

THRESHOLD DETECTORS

A third method of dosimetry[12] developed for intensities greater than 10^9 n/cm^2 delivered in a few minutes or less is based on the use of threshold detectors. The following elements have cross sections which rise rapidly at the threshold energy and remain essentially constant for higher energies:

Detector	*Threshold energy*
Pu^{239} shielded with B^{10}	100 ev
Np^{237}	0.7 Mev
U^{238}	1.5 Mev
S^{32}	2.5 Mev

Employing these detectors in addition to gold for the thermal region, a given neutron spectrum may be established in reasonably good detail. An example is shown in Fig. 13, where both the flux spectrum and the dose spectrum are given. From this information, the total dose contained in the spectrum may be determined. One outstanding advantage of this method of dosimetry is that it permits a calculation of how the energy deposited is distributed with respect to

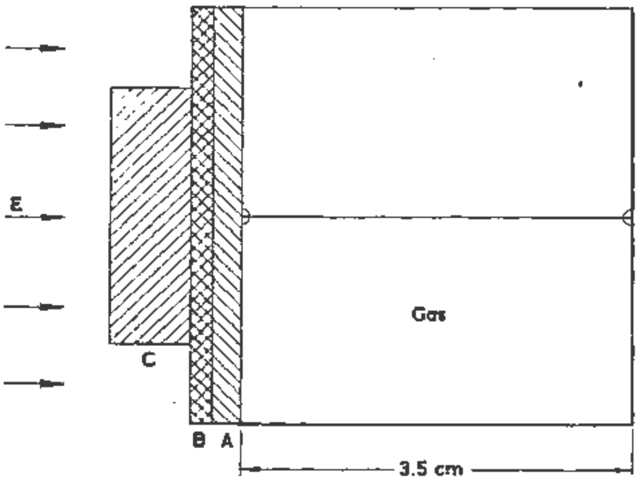

Figure 12. Diagram of the count-rate neutron dosimeter. A, 13 mg/cm^2 polyethylene; B, 29 mg/cm^2 aluminum; C, 100 mg/cm^2 polyethylene; gas is methane at 30 cm Hg pressure

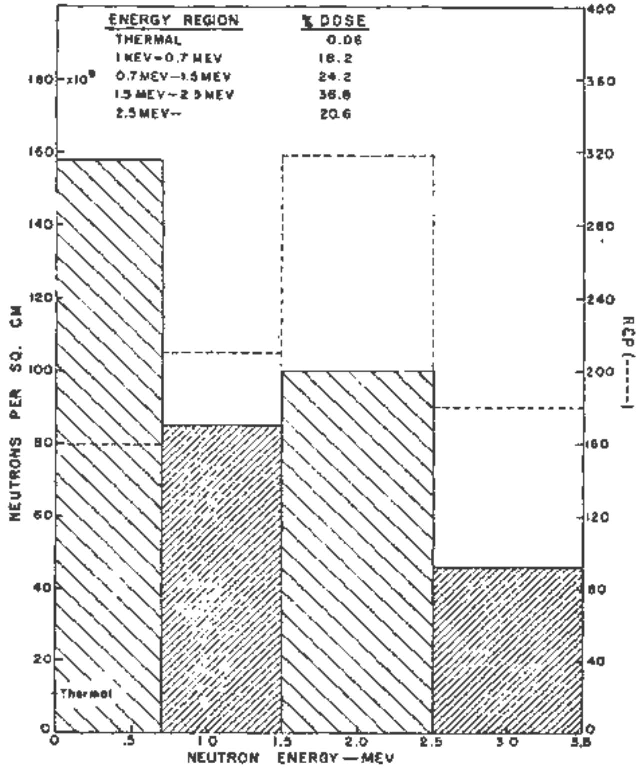

Figure 13. Typical neutron histogram, showing flux and dose distribution

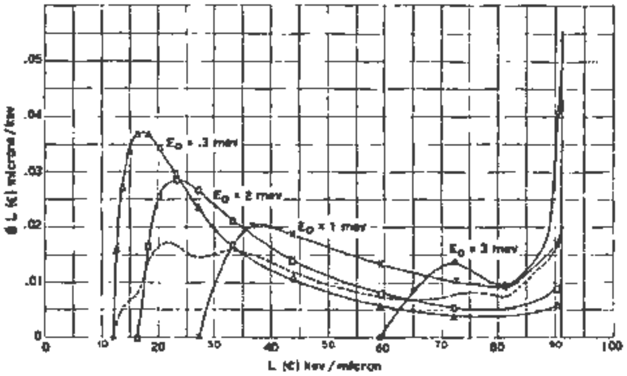

Figure 14. Dose versus LET curves for monoergic neutrons (0.3, 1, 2, and 3 Mev) and for a mixture of these energies

linear energy transfer (LET). In Fig. 14 the curves for $E_0 = 0.3$, 1, 2, and 3 Mev are the dose (or energy deposited) distributions as a function of LET, based on the work of Boag.[13] The dashed curve is the LET distribution corresponding to the neutron energy spectrum shown in Fig. 13. A comparison of measurements with the proportional counter dosimeter and with threshold detectors in the region in which both methods are applicable has shown agreement within 10 per cent. The threshold detector method has been used very successfully at certain of the nuclear weapons tests, and in other experiments where biological materials were exposed to reactor radiation or neutrons produced by cyclotrons.

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Development of Direct-Reading Chemical Dosimeters for Measurement of X, Gamma, and Fast Neutron Radiation

By George V. Taplin,* USA

The purposes of this paper are: (1) to describe the methods of preparing and calibrating chemical dosimeters of three main types, (2) to present data which demonstrate their radiation characteristics, and (3) to discuss the applicability of these dosimeters in the measurement of large doses of X- or gamma-rays and in the dosimetry of mixed neutron-gamma beams.

The three types of chemical systems which have been developed during the past six years are based on the same principle. Acid production from irradiation of chlorinated hydrocarbons, such as chloroform, trichloroethylene, or tetrachloroethylene, is a linear function with radiation dose over a broad range. The acid products may be determined directly by color changes in the pH indicator dye used or indirectly either by simple acidometric titrations or by determining the accompanying changes in pH or conductivity with readily available equipment.

The development of nuclear reactors, cyclotrons, and high energy X- and gamma-ray sources for radiation therapy has created a need for dosimeters which register large doses of X- or gamma-rays and/or mixtures of neutron-gamma radiations. To measure the gamma-ray contamination in the neutron beams from cyclotrons or in the fast neutron ports of reactors, the dosimeters must be totally neutron insensitive, or, two dosimeters could be used which respond equally to gamma radiations but differently to fast neutron exposures.

The first type of chemical dosimeter to be described is made from a chlorinated hydrocarbon overlayered with a pH indicator dye. The second dosimeter is prepared by saturating an aqueous pH indicator dye with relatively small amounts of a chlorinated hydrocarbon. Both of these systems may be adjusted to respond equally to gamma radiations; whereas their responses to fast neutron irradiation are considerably different (factor of about five) due to the relatively high hydrogen content in the second type of dosimeter. Thus, by exposing both types of instruments in a mixed neutron-gamma field, it is possible to estimate either the neutron- or gamma-ray component in the beam. A third type of gamma-ray sensitive system which is devoid of hydrogen has been made using tetrachloroethylene. Dosimeters prepared with this agent have practically no sensitivity to fast neutrons. Thus, they are capable of registering gamma contamination directly in mixed fields.

The applicability of these chemical dosimeters includes calibration of large cobalt sources, depth dose measurements in phantoms, tumor dose determination in teletherapy with high energy gamma-ray sources, dosimetry of food and drug samples being sterilized by radiation, and the measurement of fast neutrons and gamma radiation from nuclear reactors and cyclotron beams.

RADIATION EQUIPMENT

The radiation sources included a 250 kv Picker Industrial X-Ray Unit, a 70 curie Co^{60} source, a 300 mg radium source, use of the 23.5 Mev Chicago Betatron, the 60-inch cyclotron at the University of California, Berkeley, operated with 20 Mev deuterons and a beryllium target, and the 86-inch cyclotron at Oak Ridge, Tennessee, operated to give a neutron beam simulating a fission spectrum (0.5 to 3.0 Mev).

CHEMICAL REAGENTS AND METHODS USED FOR THEIR PURIFICATION

All reagents employed were analytical grade. Water was purified by triple distillation in permanganate, sulfuric acid, and silica vessels, finally collecting the condensed steam in a closed Pyrex vessel. Chlorinated hydrocarbons were purified by water washing and by fractional distillation. Only the mid-fraction was used in these studies.

The pH indicator dyes used included recrystallized bromocresol purple, phenol red, and chlorophenol red, obtained from the National Aniline Company. Stock solutions were prepared in conductivity water, dissolving 0.8 gram per 100 ml for bromocresol purple and chlorophenol red, and 0.4 gram per 100 ml for phenol red. Complete solution was obtained by heating the concentrated acid dye solution for 20 to 30 minutes at 90°C, and then adding alkali (NaOH) slowly before bringing the solution to volume with conductivity water.

PRIMARY CONTAINERS FOR DOSIMETER SYSTEMS

Pyrex and neutraglass brands of borosilicate glass flasks and/or ampules were used exclusively. They

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were cleaned prior to use by live steam under high pressure. Their inner surfaces were then coated with silicone to reduce the phenomenon of alkali leaching. Ampules were siliconed by filling them with a 2.0% solution of Dow Corning 200 silicone, 350 centistokes, in methyl chloroform. After emptying and drying, the silicone coating was cured by baking the ampules in a drying oven at 300°C for four hours.

FLAME-SEALING PROCEDURES

When glass ampules, partially filled with dosimeter systems containing chloroform-dye solutions or related hydrocarbons, are sealed directly in a gas-oxygen flame, trace amounts of hydrocarbon or dye, adhering to the glass at the point of sealing, are decomposed with the release of acids into the system. If the neck of each ampule is flushed for two to three minutes through a hypodermic needle with pure nitrogen gas just prior to flame sealing, the sealing process may be accomplished without detectable decomposition.

ADJUSTMENT OF RADIATION SENSITIVITY

The radiation sensitivity of two-phase systems may be regulated by varying the dye concentration, or its pH, by altering the amount of stabilizer (resorcinol), and by varying the hydrocarbon/dye ratios, as previously described.[1] Similar principles are employed with single-phase systems.

METHODS FOR ESTIMATING THE EXPOSURE DOSE

Dose may be estimated (± 10 to 15%) by direct visual color comparison with irradiated controls, and more accurately by titration with $10^{-3}N$ NaOH to the pre-exposure color, or by determination of pH changes in the dye indicator.

RADIATION CHARACTERISTICS

Two-Phase, Resorcinol-Stabilized Chloroform-Dye Systems

The acid yield from two-phase chloroform-bromocresol purple systems has been shown to be a linear function of radiation dose over a broad range (0 to 10^6 r). Although the exact mechanism of the radiolysis is incompletely understood, it is known to involve a chain reaction.[2] Chloroform as obtained commercially is thermostabilized by the addition of 0.5 to 1.0% ethyl alcohol. When chloroform is purified by distillation and the alcohol is removed, its sensitivity to irradiation is increased many times, indicating that the alcohol inhibits the chain reaction.[1] If resorcinol is substituted for alcohol, greater thermal stability is provided, and the length of the chain reactions is reduced.[3] However, sufficient sensitivity may be maintained to permit measurement of gamma-ray doses as low as 25 to 50 r. Previous studies have demonstrated that resorcinol-stabilized chloroform systems employing resorcinol in concentrations of 0.2 to 2.0% in the aqueous phase are capable of registering gamma radiation accurately and independently of wide variations in dose rate and/or temperature during irradiation.[3] Because of the high Z values of this system, its response to radiation has a spectral dependence similar to, but less than, that of photographic film.[3] Data presented in Table I demonstrate that if this type of dosimeter is shielded with about 0.5 mm of lead, the spectral dependency is greatly reduced, particularly in respect to the measurement of relatively soft X-rays compared with its response to gamma-rays from radium and Co^{60}.

Single-Phase, Resorcinol-Stabilized Chlorinated Hydrocarbon-Saturated Aqueous Dye Systems

Chemical dosimeters of this type contain less than 0.5% of chlorinated hydrocarbon, and therefore have Z values similar to that of water. One of their main advantages over similar two-phase systems is their relatively flat spectral response when exposed to radiations having mean effective energies varying from 35 to 1200 kv (see Table II). In addition, they exhibit very little temperature dependency. Data summarized in Table III demonstrate that their

TABLE I. Reduction of Spectral Dependency in Chemical Dosimeters by Shielding Them with Various Thicknesses of Lead Sheeting

Radiation sources	*Filters (thickness in mm)*			*Effective energy of the beam, kv*	*Acid yield in μ Meq HCl/r in dosimeters shielded with lead sheeting (mm)*				
	Cu	*Al*	*Pb*		*None*	*0.25*	*0.50*	*0.75*	*1.00*
250 kvp X-rays	0.25	1.0	0.52	190	0.24	0.125	0.075	0.055	0.045
250 kvp X-rays	0.25	1.0	—	100	0.34	0.11	0.06	0.045	0.04
150 kvp X-rays	0.25	1.0	—	65	0.45	0.10	0.05	0.045	0.04
100 kvp X-rays	0.25	1.0	—	40	0.55	0.09	0.05	0.045	0.04
Radium	—	—	—	700	0.065	0.064	0.063	0.062	0.06
Co^{60}	—	—	—	1200	0.065	0.064	0.063	0.062	0.06

NOTES: Chemical system used was the two-phase, 0.2% resorcinol-stabilized chloroform-bromocresol purple, prepared in flame sealed 8 mm diameter neutraglass ampules.

Each value is the mean, computed from exposing 10 separate dosimeters.

All instruments were exposed around the circumference of the radiation field, and were given a total dose of 800 r.

Acid yields were determined by titration with 10^{-3} N NaOH.

TABLE II. Effect of Varying the Effective Energy of X and Gamma Radiation on Acid Formation from Single-Phase Chloroform-Dye Systems

Dosimeter systems	*Radiation yield in Meq acid × 10^{-4}/ml/r at different mean effective energies*			
	35 kv	*80 kv*	*160 kv*	*1200 kv*
1. Chloroform-saturated aqueous bromocresol purple	0.025	0.024	0.023	0.025
2. Same as above, but stabilized with 0.002% resorcinol	0.018	0.018	0.018	0.018

NOTES: Radiation rates varied from 30 r/min to 300 r/min. Each value is the mean from titrating 20 separate dosimeter ampules.

Radiation factors: a) 35 kv effective, 85 kvp X-ray unit operated at 75 kvp, 5 ma, using 60 mm Al filtration. b) 80 kv effective, 250 kvp X-ray unit operated at 15 ma, 150 kvp, using 0.28 mm Cu and 1.0 mm Al filtration. c) 160 kv effective, 250 kvp X-ray unit operated at 250 kvp at 15 ma, using 0.28 mm Cu and 1.0 mm Al filtration. d) 1200 kvp effective, Co^{60} source.

acid yields remain constant when exposures are made at temperatures varying between 2°C and 50°C during irradiation. The main limitation of single-phase, chloroform-dye dosimeters is their relatively low sensitivity. However, this deficiency is overcome if trichloroethylene is substituted for chloroform and a pH dye which has color end points bracketing neutrality (phenol red) is used. To prepare sensitive systems with phenol red, it is essential that the entire procedure be accomplished in a CO_2-free atmosphere. The apparatus must maintain the system under nitrogen at all times. The dosimeter ampules must be flushed with nitrogen prior to filling and again immediately before flame sealing. An additional feature which increases the sensitivity of this system is to use a weak concentration (0.025%) of the stock dye solution of phenol red and 4½ in. long glass ampules. Distinct color changes following 20 r doses of Co^{60} radiation are readily detected visually by viewing the dosimeter lengthwise. The sensitive, single-phase, CO_2-free dosimeters made of trichloroethylene-saturated solutions of phenol red have the additional advantage over similar chloroform dosimeters of exhibiting a high degree of thermal stability. They withstand exposures in water baths at 50° to 60°C for at least a month, and for two weeks at 70°C without evidence of acid decomposition. They withstand boiling for at least seven hours. Two-phase systems of similar sensitivity decompose within a few minutes at 100°C, last only a few hours at 70°C, two weeks at 50°C. Both types, however, are stable for many months at ambient temperatures below 37°C.

FAST NEUTRON RESPONSE OF CHEMICAL DOSIMETERS

The relative fast neutron response of single- versus double-phase chlorinated hydrocarbon systems is shown in Table IV. It is apparent that single-phase dosimeters are approximately five times more sensitive to fast neutrons than the two-phase dosimeters used. This difference in neutron sensitivity is attributed to the high hydrogen content in the single-phase dosimeters. Similar studies using the 86 in. cyclotron at Oak Ridge, operated to give a neutron beam, simulating a fission spectrum (0.5 to 3.0 Mev) demonstrate that single-phase dosimeters respond the same as they do to higher energy (~ 8 Mev) fast neutrons. However, the exact difference in response of the two types of dosimeters in a mixed neutron-gamma field is critically dependent upon the exact amount or percentage of gamma-ray contamination. The problem of measuring gamma contamination with a high degree of accuracy under these conditions has not been solved completely.

Data presented in Table V show that under similar conditions, where three types of chemical dosimeters were exposed simultaneously along with National Bureau of Standards film pack dosimeters, the third type of chemical dosimeters made from tetrachloroethylene registered the lowest readings. These findings further demonstrate the relation between hydrogen content and fast neutron sensitivity of chemical systems of dosimetry. These data also indicate that hydrogen free tetrachloroethylene dosimeters are less sensitive to fast neutrons than film and/or the other two types of chemical dosimeters.

TABLE III. Effect of Varying the Temperature During Irradiation on Acid Production from Single-Phase Chloroform-Dye Systems

Dosimeter systems	*Radiation yield in Meq acid × 10^{-4}/ml/r when irradiated at different temperatures*			
	2°C	*20°C*	*37°C*	*52°C*
1. Chloroform-saturated aqueous bromocresol purple	0.020	0.025	0.027	0.027
2. Same as above, but stabilized with 0.002% resorcinol	0.017	0.018	0.020	0.019

NOTES: Samples flame-sealed in 2 ml, 10 mm diameter Kimble glass ampules selected spectrophotometrically at pH 6.0. Each value represents the mean from titrating 20 separate samples, using standard 10^{-3} *N* NaOH. Radiation factors: (*a*) 250 kvp X-rays operated at 15 ma. (*b*) dosage rate of 300 r/min. (*c*) HVL of beam 1 mm copper. (*d*) total dosage 5000 r. Dosimeters irradiated while half submerged in a water bath held at constant temperature.

TABLE IV. Neutron-Dose Distance Response of Chemical Dosimeters Upon Exposure to the University of California 60 in. Cyclotron

Neutron dose in rep	*Axial distance from Be target in inches*	*Estimated gamma contamination in roentgens*	*Neutron response of chemical dosimeters in r corrected for 10% gamma contamination*			
			Single-phase systems		*Two-phase systems*	
10,100	10		$ChCl_3$-BCP	4625	TCE-BCP	900
3950	15		$ChCl_3$-BCP	2125	TCE-BCP	400
2100	20		$ChCl_3$-BCP	1100	TCE-BCP	250
935	30	100 r	$ChCl_3$-BCP	550	TCE-BCP	85
10,600	10		$ChCl_3$-BCP	4737		
4090	15		$ChCl_3$-BCP	2280		
2390	20		$ChCl_3$-BCP	1000		
985	30	100 r	$ChCl_3$-BCP	425		

NOTES: Values shown for chemical dosimeters represent the averages from 3 to 5 separate dosimeters of each type exposed simultaneously along with sulphur dosimeters (see notes for Table III).

Gamma contamination estimated to be about 100 r, or 10% of the neutron dose at 30". Neutron dose in rep based on $2.0 \times 10^8/cm^2 = 1.0$ rep.

$ChCl_3$-BCP: single-phase systems made from chloroform-saturated solution of aqueous bromocresol purple.

TCE-BCP: two-phase systems made from resorcinol-stabilized tetrachloroethylene, overlayered with aqueous bromocresol purple.

APPLICABILITY OF CHEMICAL DOSIMETERS

General

Dosimeters of the types described have wide applicability and are adaptable to the practical solution of many special problems in radiation measurement. The applications include the calibration of large Co^{60} sources, depth dose measurements in phantoms, direct tumor dose determination in radiation therapy with high energy X- and gamma-rays, dosimetry of food and drug samples being sterilized by radiation, and measurement of fast neutrons and/or gamma rays from nuclear reactors and cyclotron beams.

For many of these laboratory applications, it is not necessary to prepare permanent type dosimeters from these systems, particularly in the case of the single-phase systems. For example, in the calibration of large Co^{60} sources, one can use either the two-phase resorcinol-stabilized chloroform-bromocresol purple system or the trichloroethylene-chlorophenol red single-phase system, in 2 ml volumetric glass-stoppered Pyrex flasks, or in similar glass bottles, sealing the reagents by use of Teflon gaskets and screw cap closures. These simple methods of preparing dosimeters for short term use avoid the more difficult problems involved in flame sealing the same reagents in specially prepared glass ampules (acid washed, siliconed). The latter methods are necessary only when the dosimeters are needed for long term (months) use, or when the application requires measurement of total exposures in the dose range below a few hundred r. Furthermore, if the application involves measurement of doses exceeding 500 r over a period of a few hours, it is possible to use single-phase systems in polyethylene or Teflon containers. External containers of this type (tissue equivalent) are useful in performing depth dose measurements in phantoms exposed to relatively low energy X-ray beams; e.g., 200 to 500 kvp.

Medical Applications

Single-phase, trichloroethylene-saturated chlorophenol red systems, prepared in 5 mm diameter, 40 mm long glass ampules or Teflon tubes, make suitable dosimeters for direct tumor dose measurement in radiation therapy. Dosimeters of this size may be inserted into various body orifices and left adjacent

TABLE V. Comparative Readings in r from Three Types of Chemical Dosimeters and from National Bureau of Standards Film Pack Dosimeters, All Exposed in Mixed Fast Neutron-Gamma Radiation Fields

Exposure number	*Chemical dosimeter readings in r*			*Film dosimeter readings in r (NBS)*
	Single-phase	*Double-phase*	*TCE*	
1	50,000	44,500	17,000	27,300
2	>20,000	20,000	15,000	19,000
3	> 7000	6000	3000	4000
4	> 5000	4500	2000	2600
5	150	95	~50	95
6	50	25	25	25

NOTES: Each value for chemical dosimeter readings represents the average from 3 to 5 samples.

Single-phase dosimeters made from trichloroethylene-saturated aqueous chlorophenol red.

Double-phase dosimeters made from 0.5% resorcinol-stabilized chloroform, overlayered with aqueous bromocresol purple.

TCE dosimeters made from di-isobutyl resorcinol-stabilized tetrachloroethylene.

NBS (National Bureau of Standards) film pack dosimeters, employing high and low range film emulsions.

to tumor sites during irradiation. The sensitivity of the system is readily adjusted to register total accumulated doses in the ranges desired (500 to 5000 r or 1000 to 10,000 r). Tumor dose may be estimated ± 10 to 15% by color comparison with irradiated controls, or more accurately (± 5 to 10%) by simple titration procedures using $10^{-4}N$ NaOH. The specific applications of these dosimeters are in the treatment of tumors of the upper and lower gastrointestinal tract, the cervix, the uterus, and the urinary bladder.

Another medical application is in demonstrating the importance of physical factors and geometry in the multiple port treatment of deep seated tumors. For teaching purposes, colorimetric dosimeters placed at a tumor site in a transparent phantom provide a directly visible method for demonstrating the necessity for meticulous care in alignment and direction of the radiation beams.

Chemical Dosimetry in Food and Drug Sterilization by Radiation

Both single- and double-phase dosimeters of the types described have considerable potential applicability in this field of high level gamma-ray dosimetry. When the dose ranges are established, it is possible to prepare pH indicator dye dosimeters which have extremely sharp color end points, thereby registering large doses with a high degree of accuracy. By adjusting the pH of these systems around 10, and with the use of a dye having color end points around neutrality (phenol red or cresol red) the final one per cent of a 100,000 r dose induces a gross color change from bright red to yellow (pH 8.0 to 7.0). Dosimeters registering gamma-ray exposure in high dose ranges can be made by using extremely insensitive reagents such as monochlorobenzene in either single- or double-phase systems.

Chemical Dosimetry of Cyclotron Beams and Mixed Neutron-Gamma Radiations from Nuclear Reactors

The three types of dosimeters described are useful in the dosimetry of mixed neutron-gamma radiations because of their difference in response to fast neutrons versus high energy gamma rays. Under laboratory conditions and high level exposures it is entirely possible to obtain sufficiently accurate data from two or more types of chemical dosimeters to determine gamma contamination as well as fast neutron doses, simultaneously. Furthermore, with extended study it seems quite possible to develop a true rep dosimeter from a single-phase, water equivalent system. The accomplishment of this goal appears possible by adjusting the oxygen content of the system and by preparing it in a tissue equivalent plastic container. The single-phase, trichloroethylene-saturated phenol red system, containing about one per cent oxygen, is about twice as sensitive to Co^{60} gamma rays than it is to high energy (3 to 8 Mev) fast neutrons. By reducing the oxygen content its sensitivity to gamma rays is lowered, but its response to fast neutrons is not altered. Thus, by finding the optimal oxygen content, it appears possible to prepare dosimeters which have equal sensitivity to fast neutrons and gamma radiation.

SUMMARY

Three types of chemical systems, prepared from chlorinated hydrocarbons stabilized with resorcinol and aqueous pH indicator dyes, have the radiation characteristics required for measurement of large doses of X, gamma, and fast neutron radiations. These systems are applicable to the laboratory preparation of dosimeters useful in the calibration of large sources of high energy X- and gamma-rays, and for performing total tumor dose measurements in radiation therapy. They are also applicable in the dosimetry of food and drug samples being sterilized by gamma irradiation. Finally, methods have been described for using these three types of chemical dosimeters in measuring gamma-ray contamination and/or fast neutron exposures in cyclotron beams and in fast neutron ports of nuclear reactors.

ACKNOWLEDGEMENTS

The author wishes to express his gratitude to the following individuals who have performed a large part of the technical work and/or have made valuable suggestions during the course of these investigations: Benedict Cassen, Ph.D., Atomic Energy Project, UCLA; Norman S. MacDonald, Ph.D., Atomic Energy Project, UCLA; Clayton H. Douglas, Cedars of Lebanon Hospital, Los Angeles; Sanford C. Sigoloff, US Air Force, Austin, Texas; Butler Sanchez, Abbott Laboratories, Radioisotope Division; S. Wayne McFarland, Atomic Energy Project, UCLA; Donald E. Paglia, US Navy; Charles Heller, Wake Forest School of Medicine, Winston-Salem, North Carolina; Melvin Martin, Atomic Energy Project, UCLA; C. William Beckner, Atomic Energy Project, UCLA; Peggyann Campbell, Atomic Energy Project, UCLA; Adele Houx, Atomic Energy Project, UCLA; and Harvey Steinberg, Atomic Energy Project, UCLA.

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Neutron and Gamma-Ray Dosimetry of a Thermal Neutron Irradiation Facility

By F. P. Cowan,* L. Lewis* and F. B. Oleson,† USA

One of the facilities[1] for biological experiments at the Brookhaven reactor consists of a cubical chamber, 12 in. on each side, within which experimental animals can be exposed to high fluxes of thermal neutrons. This chamber is located in a column of graphite projecting upward from the graphite moderator of the reactor. There are very few fast neutrons present, the cadmium ratio being of the order of 5000. However, there is a very substantial level of gamma radiation in the chamber, along with the thermal neutrons. This gamma radiation arises mostly from the interaction of neutrons with the surrounding carbon since the fission gammas from the pile itself have been attenuated by a bismuth shield in the base of the column.

A proper separation of the dosages due to thermal neutrons and due to gamma rays is of great importance when using this thermal column for animal irradiations since there are marked differences in the biological effects of neutrons and gamma rays. This paper has to do with a detailed study of thermal neutron and gamma-ray dose rates in the thermal column irradiation chamber when empty and when used for irradiation of a group of rats. Emphasis will be placed on the methods used rather than on the magnitudes pertaining to this particular facility.

MEASUREMENT OF THERMAL NEUTRON FLUX

Values of thermal neutron flux were obtained by means of small gold foils (25.2 milligrams), counted with an end-window type Geiger counter. The procedure was standardized by counting foils exposed to a known flux of thermal neutrons in the Brookhaven sigma pile. The usual corrections were made for the disturbing effect of epithermal neutrons present in the sigma pile and for the depression of the standardized neutron flux by the gold foils.[2] It was determined that one cpm corresponded to 23.8 n/cm^2/sec by comparing the observed counting rate of the Geiger counter with the computed disintegration rate. An independent standardization was made by determining the absolute disintegration rate of a foil with a coincidence counter available at the Chemistry Department. This counter detects the 0.411 Mev Au^{198} gamma ray with a sodium iodide crystal, and detects the 0.96 Mev Au^{198} beta particles with an anthracene crystal. It also registers the coincidence rate between them. The Au^{198} disintegration rate is equal to the product of the gamma and beta counting rates divided by the coincidence counting rate. Good agreement was obtained. The thermal neutron flux in the empty facility for typical pile operating conditions is 9.8×10^8 n/cm^2/sec.

Since a number of rats were to be irradiated, it was of interest to know the variation of flux with position in the chamber. Accordingly, foils were exposed at a variety of locations in the empty facility. No significant variations over the bottom of the chamber were found. However, a drop of 12% was noted in going up 5 inches from the bottom.

It was also of interest to know how much the flux varied with direction. To determine this, discs of cadmium were affixed to the six sides of a lucite cube, a gold foil was mounted on the outside surface of each disc and the whole assembly was irradiated at the center of the chamber. Thus, each foil was irradiated only from one side. The activity of the top foil was 31% less than that of the bottom foil, indicating an appreciable transport flux. The four side foils agreed with each other and were 27% less active than the bottom foil.

MEASUREMENT OF GAMMA-RAY DOSE RATE

The gamma-ray dose rate in the empty facility was determined by the lithium extrapolation method of Brennan, *et al.*[3] A Landsverk thimble chamber was enclosed in a 1-inch thick shield of lithium. This is thick enough to reduce the thermal flux at the chamber to a negligible value without significantly increasing the gamma exposure. However, the lithium shield depresses the neutron flux somewhat and thereby reduces the gamma radiation since, as noted above, the gamma radiation originates largely from neutron interactions with the surrounding graphite. This gamma reduction is corrected for by adding ⅛ in. thick lithium shells and obtaining a plot of gamma-ray dose rate, measured by the chamber, as a function of lithium thickness for thicknesses between 1 in. and 2 in. Extrapolation of this plot to zero thickness of lithium gives the gamma-ray dose rate for the empty facility. The plot is linear on semilog paper but the extrapolation is a rather wide one and some uncertainty in the gamma level obtained is inherent in the method. The value of gamma-

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* Brookhaven National Laboratory.

† Federal Civil Defense Agency.

ray dose rate corresponding to the thermal neutron flux mentioned above was found to be 125 r/hr.

Recently, a small carbon chamber with a long neck has been constructed for measuring gamma-ray levels in the irradiation chamber directly. Provision is made to flow carbon dioxide gas through the chamber, thereby minimizing its response to thermal neutrons. The value obtained should set an upper limit to the gamma-ray dose rate since radiation-induced leakage in the insulators or thermal neutron interactions in the graphite or filling gas will add to the ionization current caused by gamma rays. Measurements with this chamber have just started but appear to be in reasonably good agreement with the lithium extrapolation data.

DESIGN AND CALIBRATION OF FILM DOSIMETER

For measurements of gamma-ray dose rates during rat irradiation it was necessary to have a dosimeter that could be used in the thermal column without a bulky shield. It was decided to use a film dosimeter and to correct for its response to thermal neutrons by means of a thermal neutron calibration. This calibration was obtained by exposing the dosimeter in the empty facility to a measured flux of neutrons and subtracting the response due to the gamma component which was known from the lithium extrapolation measurements. There was no appreciable depression of the thermal neutron flux caused by the presence of the film dosimeter in the irradiation chamber.

The Bureau of Standards film dosimeter[4] was chosen because of its freedom from any appreciable dependence on gamma-ray energy. However, since no gamma rays above 5 Mev were expected, the thickness of plastic in the badge was reduced from 8.5 mm to 6 mm. A further reduction in size was made possible by the use of a circular film $\frac{9}{16}$ in. in diameter, such as is used locally in film rings for personnel monitoring. The resulting dosimeter was considerably smaller than the Bureau of Standards model but presumably had similar characteristics. A small size dosimeter is desirable for this application both because of the limited space and our desire to disturb the existing neutron flux as little as possible. Figure 1 shows the comparative sizes of the two dosimeters. It was found that a neutron flux of 1.56×10^{10} n/cm^2 gave the same darkening of the film as 1 r of gamma radiation. In practice it was found that the dosimeters became only slightly activated during the usual 30-minute period of irradiation.

DOSIMETRIC PROCEDURES FOR RAT IRRADIATIONS

During previous experiments a number of rats had been placed in an uncompartmented lucite box during irradiation. There had been considerable uncertainty about the uniformity of dose from rat to rat since it was possible for one rat to be shielded by others in an unpredictable fashion. It was highly desirable that all rats should receive the same exposure and with this in mind the irradiation box shown in Fig. 2 was designed. Spaces are provided for 8 rats and the box is compartmented in such a way that each rat has an identical geometrical relationship to the other rats and to the pile. Thus, all 8 will receive the same exposure. The box is 8 in. × 8 in. in cross section and 6½ in. high.

It was decided to specify rat exposures to neutrons and gamma rays, in terms of the neutron and gamma-ray levels in air at the location where an individual rat was to be placed. These levels were to be determined, however, in such a way as to take

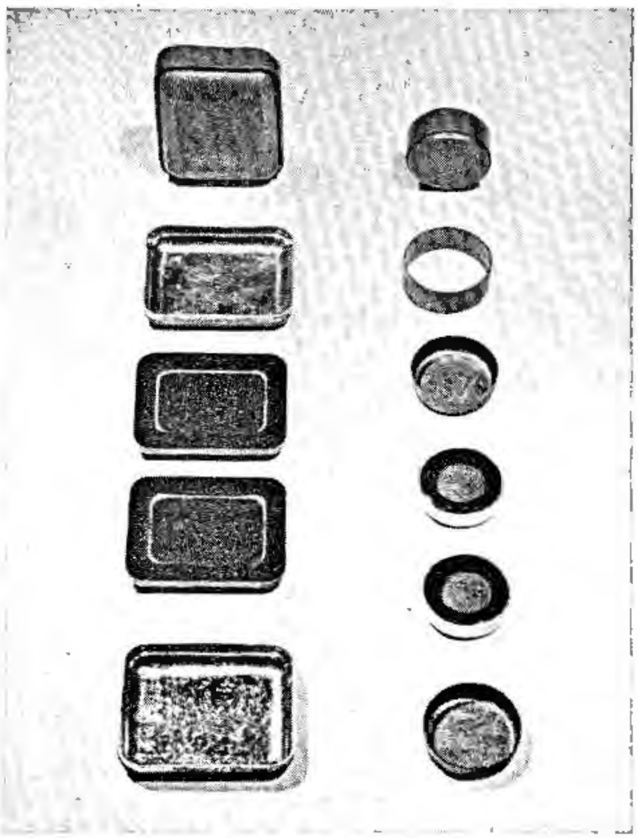

Figure 1. Film dosimeters

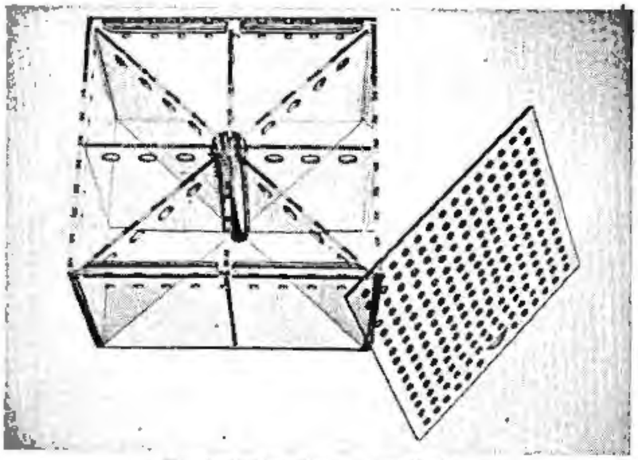

Figure 2. Box for rat irradiations

account of the effect of the rats on the general neutron and gamma-ray levels in the chamber and to include extra gamma rays originating from neutron interactions in the other rats. Accordingly, measurements were made of neutron flux and gamma-ray dose rate in one compartment of the irradiation box of Fig. 2 with the other 7 compartments occupied by rats. It was found that the neutron flux in the empty compartment was determined by the total weight of rats in the other compartments and was insensitive to the geometrical distribution of that weight. Figure 3 shows a plot of the neutron flux in the empty compartment against the total weight of rats in the other compartments. In determining the neutron flux for the irradiation of 8 rats the total weight of all 8 is used for obtaining the neutron flux from this curve. Thus, the effect of the 8th rat in depressing the general neutron flux level is accounted for.

It was found that the gamma-ray level in one compartment was essentially independent of the weight of rats in the other compartments. Presumably, the reduction in gamma-ray level caused by depression of the neutron flux is roughly balanced by the extra gamma rays originating in the rats.

For determination of dosage during individual irradiations, a gold foil was always exposed at a fixed location at the top of the lucite box. The ratio between the neutron flux at this location and that in one of the compartments was carefully determined and was found to be 1.65. This ratio was found to be independent of the weight of the rats being irradiated within the range of weights of practical interest. Such a monitor for individual exposure is of great importance since there are bound to be fluctuations in pile power even during a single run and the relation between pile power and neutron flux in the thermal column is subject to change from time to time. Exposure times were chosen on the basis of pile power at the time of irradiation but the exact value of exposure was always based on the simultaneously exposed foil just mentioned.

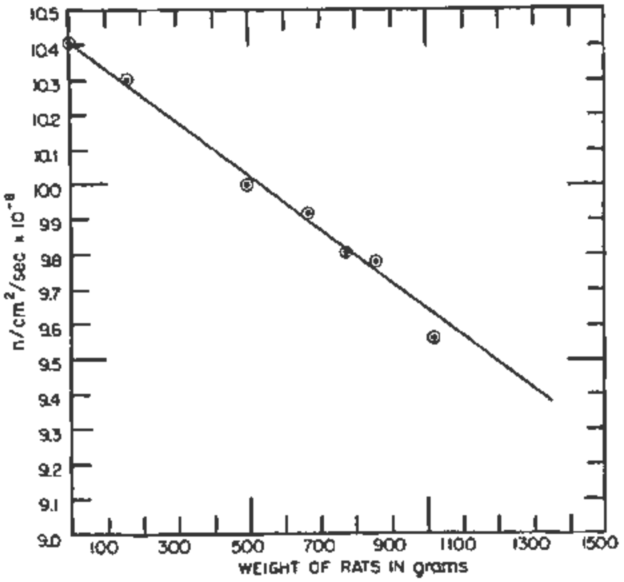

Figure 3. Effect of rats on neutron flux

The above procedure was used for a study of the "Thermal Neutron Equivalence of Whole Body X-Irradiation,"[5] and was found to be very satisfactory. It may be of interest to note that for a neutron exposure rate of 8.6×10^8 n/cm²/sec the total gamma dose rate was 215 r/hr of which about 120 r/hr came from the facility itself and 95 r/hr originated in the surrounding rats and may be referred to as inter-rat gamma radiation.

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A Neutron Device for Measuring the Moisture Content of Soil

By Jan Urbanec, Czechoslovakia

In the building and construction industry, it is very important to know the moisture content of the soil, for instance in the foundations of buildings. A great many methods have been developed for the purpose. The one which resorts to evaporation is the most accurate and most often used. Nevertheless, it is too slow and, in some cases, does not give an answer fast enough. The "fast" methods, for instance, are those based on measuring the electrical conductivity and permitivity of the soil. These methods are being tried in our country with good results, but, under complicated conditions, the interpretation of the results is not simple, and the accuracy lower than that of the evaporation method.

Noticing the special properties of water or of hydrides during neutron thermalization and scattering, several authors took advantage of this effect to determine the hydrogen content in soils. This method was used and described in 1951 by V. Gardner and Don Kirkham. The neutrons were supplied by a Po-Be source, the detector being a BF_3 filled neutron counter. These authors analyzed the method and the experimental results, on the basis of which one may accept that it has been shown that it is possible to use this method for rapid measurements.

We have improved the method by using Ra-Be as a source of neutrons, and a slow-neutron scintillation counter. An analysis has been made of the correlation between the measured neutron flux and the water content of the soil.

The purpose of the next part of this paper is to describe the experiments carried out and to give the results of our measurements.

PRINCIPLES OF THE METHOD USED

The neutrons leaving a source of fast neutrons such as Ra-Be, upon reaction with atomic nuclei of the medium in which the source is located, are either slowed down or captured by a nucleus.

The slowing down depends on the atomic number of the nuclei with which the neutron collides, and reaches its highest value upon collision with the hydrogen nucleus (proton). The average logarithm of the ratio of neutron energy before collision to neutron energy after impact, for the hydrogen atom, is several times larger than what it is for collision with the next most effective element found in soil in any significant quantity. The fact must also be taken into account that, of all the elements which are commonly found in the soil, hydrogen has the largest cross section. These facts account for the property of selective sensitivity to the presence of hydrogen in soils.

Once thermalized, the neutrons are scattered and detected.

In order to present the factors by which the change in the slow neutron flux (as registered by the detector at a constant distance from the fast neutron source) is determined, an elementary analysis of the problem is in order.

The following is a basic outline of the solution:

1. First, let us determine the distribution of slow neutrons in the space surrounding the point source of fast neutrons under the given test conditions. The distribution found for the slow neutrons will be the solution of the age equation for the deceleration density.

2. We will then substitute the known slow neutron density into the diffusion equation and will solve it for the boundary conditions.

In the first approximation for moist soil, only the number of atoms per unit volume changes with the moisture content. This assumption does not hold for the macroscopic absorption cross section, since the latter varies more in accordance with:

$$\Sigma_a = (\Sigma_a)_0 - (\Sigma_a)_1 S$$

where S is the relative water content per unit volume. (Quantities with a subscript 0 pertain to $S = 0$, and quantities with a subscript 1 pertain to $S = 1$.)

In the case of a point source, the solution of the age equation is

$$2\,(r, \tau) = \frac{e^{-\frac{r^2}{4\tau}}}{\sqrt{(4\pi\,\tau)}}$$

The value of τ, for a given fast neutron energy E_0, and energy E_{th} of the thermal neutrons, in the first approximation, is only a function of $\lambda_S{}^2$ in our case and, therefore $\tau = \tau_1 S^{-2}$

With the same value of $D_a\ \Sigma_a$ the functions of S also are the same:

$$D = D_1 \cdot S^{-1} \qquad \Sigma_a = (\Sigma_a)_1 \cdot S$$

Original language: Russian.

Then the diffusion equation may be rewritten as follows:

$$D_S^{-2} \Delta \phi (r) - \Sigma_a \phi (r) + S^2 2^+ (r, \tau_{th}) = 0$$

Solving this equation we find that the neutron current density is a function of S^2.
If the solution of the equation when $S = 1$ is

$$\phi (r) = \phi_1 (r)$$

then for an arbitrary value of S

$$\phi (r,S) = S^2 \phi_1 (S \cdot r)$$

Therefore, if the detector is placed very close to the source, where $\phi_1 (r) = \phi_1 (o)$, then the number of pulses is a quadratic function of the moisture content.

This result constitutes a satisfactory approximation for the cases found in practice. At the present time, attention is paid to the analysis of the solution for the case of space limited by a medium, so as to be able to determine to what distances this method of measurement is sensitive with different soil moisture contents.

SCINTILLATION DETECTOR FOR SLOW NEUTRONS USING A SCINTILLATION LAYER

Under field conditions, it is possible to use neutron sources Po-Be, Ra-α-Be, Ra-γ-Be. For practical utilization under field conditions the Ra-α-Be source is the most useful since its long half-life enables one to consider the intensity of the source as being constant.

A disadvantage of this source is the strong background of γ-rays emitted together with the neutrons. This means that slow neutrons from such a source can be measured only with detectors which are insensitive to γ-rays, but very sensitive to slow neutrons. In order for such a detector to be practical, its sensitivity must be higher than the sensitivity of a BF_3 counter, taking into account the geometrical fact that a large-size counter is located out in the space where the thermal neutron flux is small. This reduces its effectiveness as compared to a BF_3 counter when a point source of neutrons is used. It must also be remembered that use of the Ra-α-Be source precludes the use of the simple method with silver-cathode Geiger counters.

We attempted to design a sensitive scintillation detector which would meet the above requirements. As a first model, we tried a multi-layer detector with a transparent layer of B_2O_3 and a 0.5 mm thin naphthalene scintillation layer. This was chosen in order that, after testing a single-layer detector, it would be possible to use a multi-layer detector of greater sensitivity. Such detector construction, however, turned out to be too ineffective to register slow neutrons. With a single-layer detector using ZnS(Ag), the sensitivity also remained low.

During the next stage we tested a detector with a molten mixture $ZnS(Ag) + B_2O_3$. Such scintillation material had considerable sensitivity for the experimentally established 1:1 ratio of mixture. In order that the scintillation properties of ZnS(Ag) crystals be as little affected as possible, the mixture was melted at a temperature slightly higher than the melting temperature of boron oxide. The size of the ZnS(Ag) grains was about 20 μ.

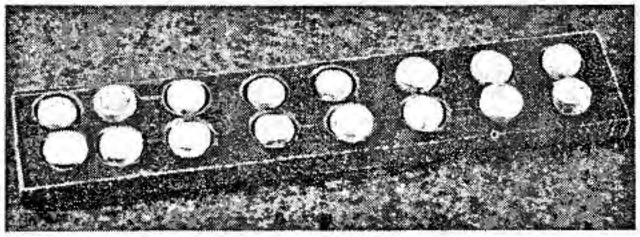

Figure 1

The maximum of the ZnS(Ag) radiation shifted from the violet region to the blue and green regions. Sixteen samples (Fig. 1) were prepared under identical conditions, but with varying amounts of component substances (with a diameter of 1 cm) for the purpose of determining the correlation between the sensitivity and content of the mixture; from this the best ratio of the component substances was determined. In preparing the mixture, it was necessary to maintain the operating temperature very accurately, as well as the time of heating, since the properties of the scintillation substance prepared in this manner strongly depend on the processing method and time. This is probably due to the fact that, in this process, the scintillation substance is activated by boron. This postulate must be verified with larger crystals.

The samples containing different proportions of component substances were measured, and the relative values were plotted (see Figs. 2 and 3).

The sensitivity of the detector to γ-rays was investigated by measuring the number of pulses registered for the adjustment of the discriminator while the detector was being irradiated by a 5 millicurie source of Co^{60} (Fig. 4). Photomultiplier tubes RCA 5819 and FEU 19 were used for testing the substances.

In order to produce slow neutrons, a 25 cm cube of paraffin was used, with a round channel 30 mm in diameter drilled through to the center (Fig. 5). A 50 millicurie Ra-α-Be source supplied the fast neutrons. A 25 cm square cadmium plate was used as a slow neutron screen.

The absolute determination of neutron current at the place where the scintillation substance was located was carried out by activating a layer of V_2O_5. By this means, the efficiency of the mixture giving the most sensitive ratio was shown to be 4%.

PROBE FOR FIELD MEASUREMENTS

For measurements under field conditions a probe was designed (Fig. 6), having an effective detecting surface of 12 cm², with photomultiplier tube RCA 5819. The 50 millicurie neutron source was located at

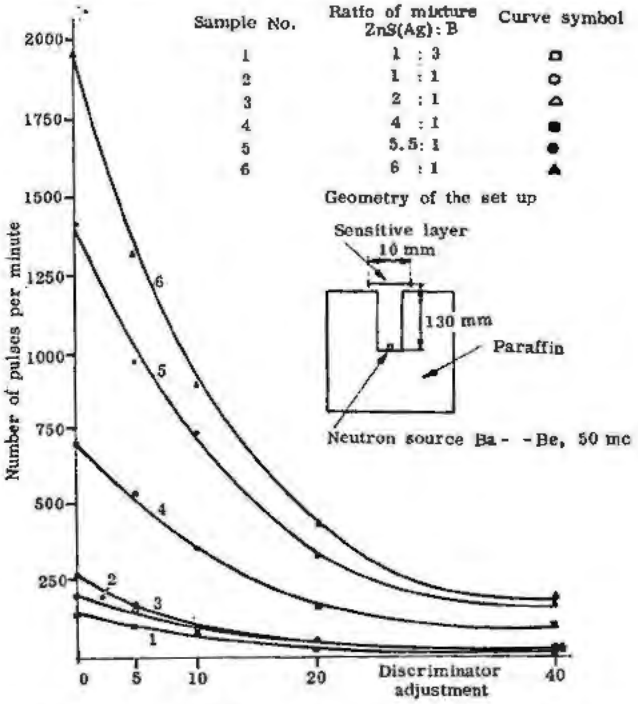

Figure 2. Dependence of cpm on discriminator adjustment (cpm during screening with a cadmium plate)

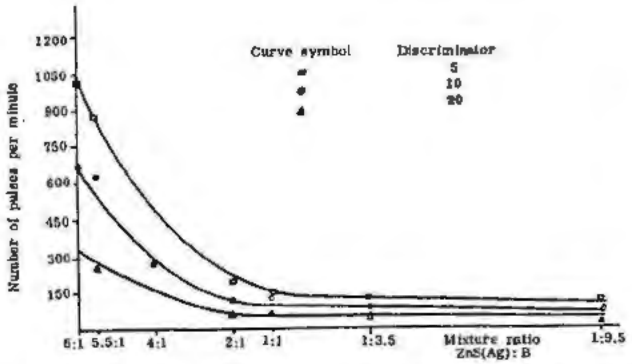

Figure 3. Detector sensitivity as a function of mixture ratio

some 30 mm from the scintillation layer; the γ-rays were screened out by a lead diaphragm in the cylinder 25 mm in diameter 30 mm long. The scintillator used in this probe did not give optimal efficiency, since a 1 :1 ratio of component substances was chosen. This non-optimum ratio was chosen before the study of the properties of the scintillator had begun, since it was necessary to start measurements under field conditions, even before obtaining the results from the laboratory. The probe has a conical tip, so that it may be pushed into the soil.

This probe was used in the laboratory on sand kept in a container (container diameter 42 cm, height 25 cm): the dry sand was gradually moistened with water. The resulting ratio between water content and pulse number is shown on Figs. 7 and 8.

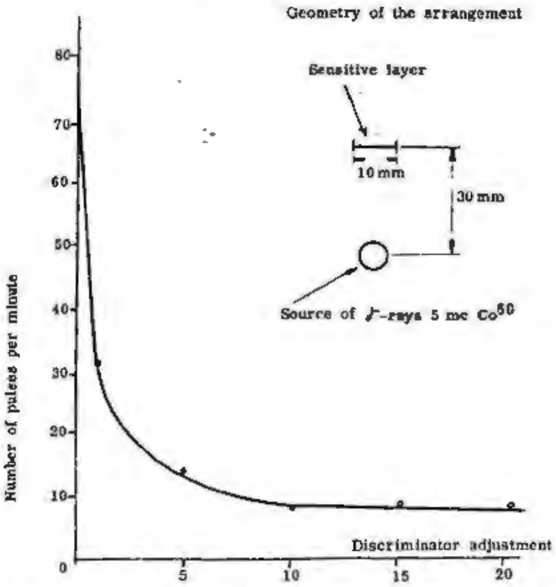

Figure 4. Sensitivity of sample No. 5 to γ-rays vs discriminator adjustment

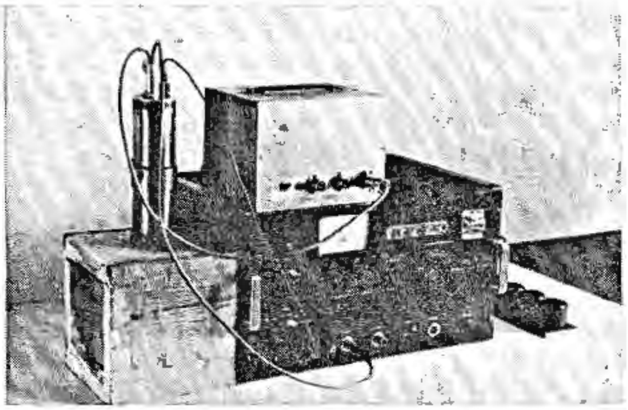

Figure 5

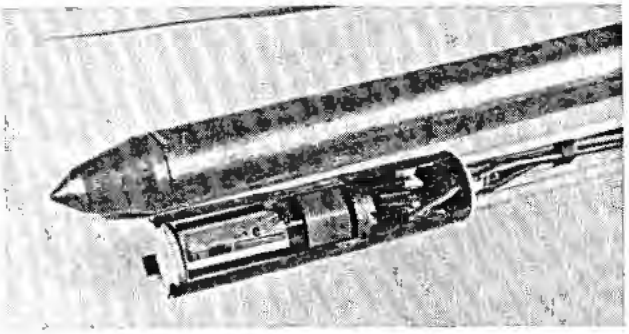

Figure 6

This coincides with the measurements made by other authors.

CONCLUSION

On the basis of the experiments carried out, it has been shown that it is of value, in order to measure the moisture content of the soil, to use a probe containing a scintillation detector and a Ra-α-Be source. The probe is sturdy (mechanically speaking): it can stand rough handling under field conditions. The associated electronic equipment is simpler and more compact than that used with BF_3 counters. The neutron current is used more efficiently, which leads to the reduction of the required neutron sources, and thus to minimizing effects on the operator. The probe contained a 50 millicurie Ra-α-Be source with a low efficiency scintillator. If a scintillator having a better mixture ratio is used, the required source intensity will be substantially reduced.

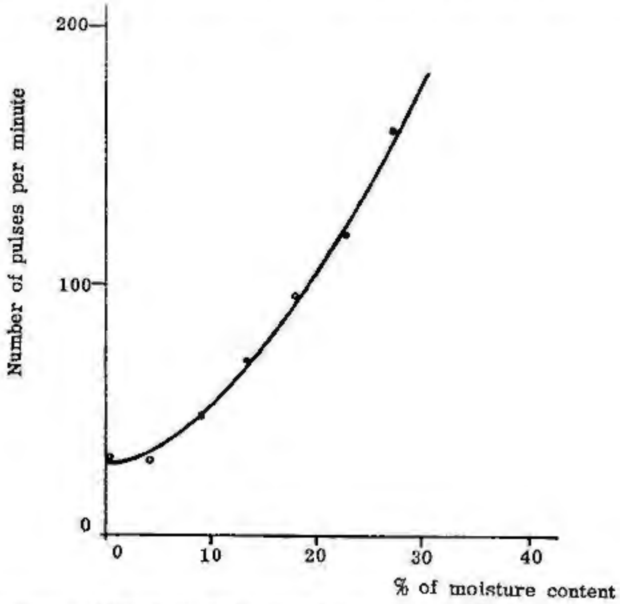

Figure 7. Number of pulses vs moisture content (laboratory measurement)

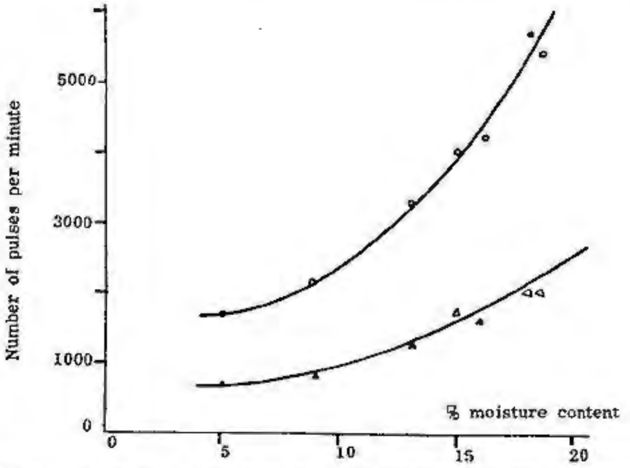

Figure 8. Number of pulses vs moisture content (measurements under natural conditions)

The scintillation detector using a fused mixture of ZnS (Ag) + B_2O_3 also will be useful for other neutron physics applications. Further improvements on the detector are being carried out.

Dosimetry of Ionizing Radiations

By G. Failla,* USA

In 1937 Gray[1] proposed a system of dosimetry based on energy absorbed in tissue and showed how this can be determined by suitable ionization measurements. Because of the great interest in the measurement of dose in roentgens, the attention of radiological physicists for many years subsequently was focused mainly on the application of the Bragg-Gray principle to such measurements. For this purpose the appropriate conditions under which ionization measurements were to be made had to fulfill also the requirements imposed by the definition of the roentgen. Since most of the publications on this subject have dealt with this problem the distinction between the conditions of measurement imposed by the Bragg-Gray principle and by the definition of the roentgen, has been obscured. Advances in radiobiology, as well as the great increase in the use of other types of ionizing radiation in recent years, have shifted the emphasis to the measurement of dose in rads. Also, the conditions of exposure encountered today are much more diversified than formerly. It is well, therefore, to re-examine the situation.

We shall assume at the outset that the objective is to determine by ionization measurements the absorbed dose at a "point" in a tissue no matter how it is irradiated and no matter how the dose may vary from point to point in the tissue. By absorbed dose is meant the dose in terms of the local specific energy absorption (that is, the energy absorbed per unit mass of tissue) in rad.† Dose at a "point" means the dose in a small element of volume surrounding the point, in terms of the average specific energy absorption in this volume—the linear dimensions of the element of volume being large in comparison to the effective diameter of the track of the ionizing particle in the tissue. Explicitly, we are excluding, for example, the specific energy absorption in an element of track of an alpha particle, which is extremely high in comparison with the average in the element of volume envisaged here.

The energy absorption by the tissue with which we are concerned refers to the locally effective portion of the energy imparted to the tissue by the radiation. In essence this means that we are dealing with the energy locally transferred in small lumps to the atoms and molecules of the tissue by ionizing particles traversing it, no matter where and how these particles originate. Let us consider the local transfer of energy from an ionizing particle to matter. In a gas it is possible by suitable means to determine the average energy lost by a particle per ion pair produced. This quantity is generally called *W*. The experimental procedure may be to measure the energy of the particle before and after traversing a layer of gas in which the ion pairs produced are counted. It has been found that in general‡ the value of *W* is about twice the ionization potential of the gas, which means that a considerable portion of the energy lost by the ionizing particle is absorbed initially by the gas by processes other than ionization (excitation, thermal agitation and possibly other processes). In special cases large lumps of energy may be lost by the particle without appreciable ion production in the gas layer and this would lead to an abnormally high value of *W*. Thus in a radiative collision of a high energy electron, the energy of the photon would be "lost" by the electron but it would contribute little to the measured ionization. This energy, then, would not be locally effective and should not be included in the energy absorption under consideration.

From the foregoing it follows that the energy locally absorbed in a volume of gas traversed by ionizing particles can be determined by counting the number of ion pairs produced and multiplying it by the appropriate value of *W*. The appropriate value of *W* is the one that applies to the gas, kind of ionizing particle and energy spectrum of these particles—and any other factor that may affect it.

The next step is to go from a gas to a solid. If the rate of loss of energy of the ionizing particles per unit mass traversed (mass stopping power) is independent of the physical and chemical states of aggregation of the atoms of the material, the energy absorbed from the same ionizing particles is the same in a solid as in a gas of the same atomic composition. In this case, therefore, by measuring the ionization produced in the equivalent gas and using the appropriate value of *W*, it is possible to determine the local energy absorption in a solid, provided the ionizing particles acting on the gas and the solid are identical in every respect. In most practical cases the mass stopping power is essentially independent of the state of aggregation of the atoms of the material and, therefore, the problem is to measure the ionization in a tissue equivalent gas under proper conditions and to determine the appropriate value of *W*.

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† One rad equals 100 ergs per gram.

‡ That is, excluding noble gases.

Homogeneous Medium with Uniform Flux of Ionizing Particles

Fano[2] has shown that "in a medium of given composition exposed to a uniform flux of primary radiation (such as X-rays or neutrons) the flux of secondary radiation is also uniform and *independent of the density* of the medium as well as of the density variations from point to point." In this case the presence of a gas cavity (of any size) within the solid does not alter the flux of ionizing particles so long as the gas has the same atomic composition as the solid. Hence, the energy absorption in the solid (tissue) can be determined by measuring the ionization per gram of gas and applying the appropriate value of W. It should be noted, however, that the constancy of the flux of secondary particles is predicated on the existence in the medium of a uniform flux of primary radiation. In practice this condition is not readily fulfilled when the medium is exposed to a single beam of primary radiation. Accordingly, the size (and at times the shape) of the gas cavity must fulfill certain requirements that will be discussed later. A very close approximation to a uniform flux of primary and secondary radiation may be found in practice in the central portion of a (large) mass of a chemical compound in which one atom of the molecule is radioactive—or a certain fraction of the molecules contain such atoms. In this case the gas cavity in which the ionization is measured may be of any size and shape, provided the gas contains the same proportion of radioactive atoms as the solid medium. When the radioactive material emits only beta particles the flux is uniform throughout the mass except in a peripheral layer of a thickness equal to the range of the fastest beta particles. In any case if the gas is not radioactive like the solid, the cavity must be very small for reasons that will be apparent later.

EXTRAPOLATION IONIZATION CHAMBER

In 1937 the writer described[3] an ionization chamber designed particularly to determine the back scatter contribution to the skin dose in X-ray therapy. It is generally known as an "extrapolation chamber" because the final result of measurements made with it is obtained by extrapolation. Actually in the measurement of skin dose two extrapolations were involved: one to eliminate the influence of the gap between the two parallel electrodes, and the second to correct for X-ray absorption in the first electrode of the chamber. Since then many different types of extrapolation chamber have been constructed for special purposes, having plane, cylindrical or spherical electrodes. The fundamental principle is the same in all cases. The ionization is measured in the gas in a narrow gap of uniform thickness between two electrodes (Fig. 1). Readings are taken with different gaps and the results are plotted. Since when the gap is zero there is no gas in which ionization can take place the curve thus obtained should pass through the origin. The slope of this curve at the origin represents the limiting value of the ionization current per unit length of gap as the electrodes are brought closer and closer together—that is, when they are in contact. Thus the influence of the gap *per se* may be eliminated. In practice, of course, there are many precautions to be taken, of which the following three are most important: (1) readings should be taken with small enough gaps to permit extrapolation to zero gap with the desired accuracy; (2) readings should be taken with positive and negative voltages applied to the chamber of sufficient magnitude to insure saturation current—the average of the positive and negative saturation current readings for each gap should be used in plotting the curve; (3) the spacing should be measured accurately. Experience has shown that for gaps of a few tenths of a millimeter the magnitude can be determined most accurately by measuring the electrical capacitance of the system and calculating the spacing from its geometry. This requires that the chamber and current measuring device be especially designed for this purpose. By suitable provisions it is possible to make the capacitance measurement while the chamber is exposed to radiation. (This is necessary for instance when the source of radiation is incorporated in the walls of the chamber.)

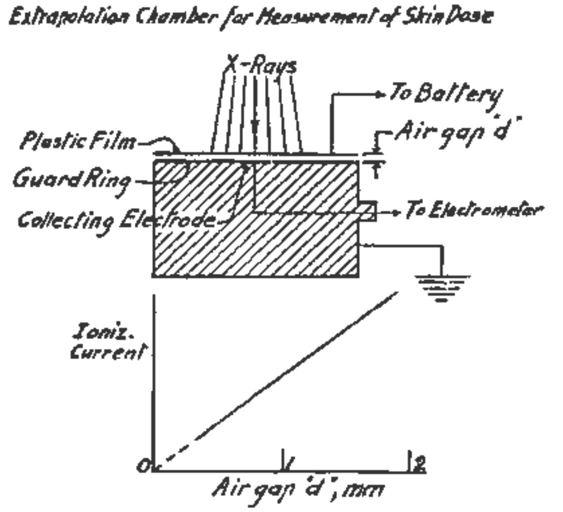

Figure 1

Homogeneous Medium with Non-Uniform Flux of Ionizing Particles

It will be shown elsewhere that in this case, changes in *density* of the medium disturb the flux of ionizing particles. Therefore, the introduction of a gas cavity of appreciable dimensions in the medium to measure the ionization, introduces errors in the determination of the local energy absorption. As shown above, the extrapolation chamber method in essence does away with the gas cavity. Therefore, at the limit the flux of ionizing particles responsible for the ionization in the gas is identical with the flux in the solid across the surface of virtual contact of the electrodes. Assuming again that the gas has the same atomic composition as the solid (and with the same qualifications as before) the number of ion pairs produced per gram of gas (extrapolated value) multiplied by the appropriate W gives the energy absorbed per gram of solid in a very thin layer along the surface of contact. It is taken for granted here that the geometry of the chamber is such as to insure uniformity of flux over the area of virtual contact. If this is not the case the result will represent an average for a thin layer of the given area. However, in principle the area can be made very small and thus the specific energy absorption "at a point" may be determined even when the flux of ionizing particles varies rapidly from point to point in the medium.

An important practical problem in which the flux of ionizing particles varies rapidly with tissue depth, in spite of the fact that the flux of primary radiation is nearly constant, is the measurement of absorbed dose in the skin (and deeper tissue layers) of patients treated with multi-million-volt X-rays, when initially the radiation does not contain the full complement of secondary electrons. The problem may be solved readily by the use of a simple parallel plate extrapolation chamber, in which the thickness of the front electrode may be varied to represent the tissue depth of interest.

Point Source of Radiation in a Homogeneous Medium

The practical problem in this case is to determine the tissue dose at different distances from the source. The flux is very non-uniform but its spherical symmetry simplifies the problem. Obviously, use of a spherical extrapolation chamber, with the source at the center, is indicated. However, if we are interested in the dose distribution very near the source the chamber must be very small and the scheme becomes impractical.

It will be shown elsewhere that a *uniform* change in the density of the medium alters the flux of ionizing particles in a systematic and predictable manner. Consider two homogeneous media of the same atomic composition, both of uniform density but differing in density by a factor n. A particle originating from the point source will cross a concentric spherical surface of radius r in the denser medium with a certain orientation and a certain residual energy. The same particle in the lighter medium will cross a corresponding spherical surface of radius nr with the same orientation and the same residual energy. (The amount of matter, in mg/cm^2, traversed by the particle in the two cases is the same.) Therefore, the total flux of ionizing particles through the two spherical surfaces is the same in number, directions and residual energies. Hence, by using a tissue equivalent material of lower density than tissue, the linear dimensions of the spherical extrapolation chamber may be increased in the ratio of the two densities. If the point source emits only beta rays (or beta rays and high energy gamma rays), balsa wood is a sufficiently good approximation to soft tissue insofar as its interaction with the radiation is concerned. Balsa wood with a density of 0.1 gm/cm^3 is readily obtainable and, therefore, the dimensions of the extrapolation chamber can be increased by a factor of ten. This means that extrapolation with a balsa wood sphere 5 mm in radius makes possible the determination of the absorbed dose at a distance of 0.5 mm from the point source in a tissue of density 1 gm/cm^3.§ (Fig. 2.)

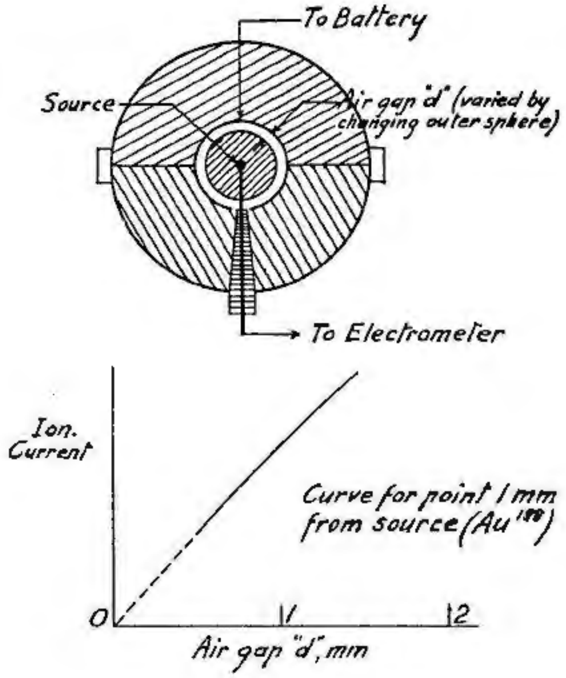

Figure 2

It is important to note that the sphere must be in a balsa wood block of the same density of sufficient size to provide the same complement of scattered electrons to the gas shell in which the ionization is measured that would obtain in the corresponding shell in the tissue. Differences in density within the sphere and in the surrounding block will upset the correspondence. (The gas gap will disturb the flux too but this is eliminated by extrapolation.) This method is being used by Miss P. McClement[4] in the writer's laboratory to determine the tissue dose rate in rads per hour at different distances from a "point" source of beta rays (P^{32}) of known radioactive content. For each distance an inner sphere of the proper radius and at least three outer spheres of different radii are required to permit extrapolation to zero gas gap.

Balsa wood spherical extrapolation chambers of small dimensions are very difficult to construct and to use. The one just described is about the smallest

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§ It will be seen later that the gas need not be "balsa-wood equivalent".

practical one. Yet in some cases it is of interest to know the dose at much shorter distances from the point source, or the integral dose in a living cell containing radioactive material at the center. Such problems can be attacked experimentally by further expansion of the tissue equivalent medium, that is, by using a gas instead of a solid. So long as the density is uniform throughout the medium (now of much larger dimensions) the total flux of ionizing particles through a concentric spherical surface is the same as through a correspondingly smaller spherical surface in the tissue. To maintain this correspondence no solid material must be introduced in the region where the ionization is to be measured. The writer has developed "wall-less" ionization chambers for this purpose and Miss McClement has made measurements with them[5] (Fig. 3). The essential principle of these chambers is that the gas volume (nearly spherical) in which the ionization is measured is defined by the lines of force of the applied potential rather than by solid walls. The source of radiation is suspended on a thin film and the influence of this solid material is substantially eliminated by extrapolation (provided the energy of the beta rays is not too low).

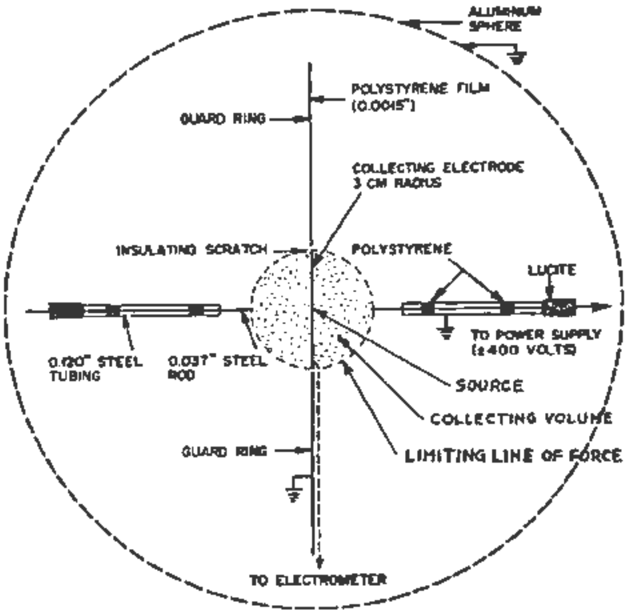

Figure 3. Wall-less ionization chamber model I

Measurements with Ionization Chambers in Which the Gas and the Wall Do Not Have the Same Atomic Composition

It has been pointed out above that when (1) the solid medium and the gas have the same atomic composition and (2) the flux of ionizing particles is uniform, the cavity (i.e., ionization chamber) can be of any size. However, when the flux of ionizing particles is not uniform the cavity *per se* disturbs the local flux at the point of interest. The difficulty can be overcome by using a suitable extrapolation ionization chamber whereby the influence of the cavity due to geometric factors is eliminated. When the gas in the cavity has a different atomic composition from that of the solid medium, complications arise because of the difference in the interaction of the ionizing particles with the atoms of the solid and those of the gas. If the ionizing particles are secondaries, there will also be a difference, in general, in the interaction of the primary radiation with the solid and the gas.

The Bragg-Gray principle is generally applied to situations in which both the primary flux and the flux of ionizing particles are uniform in the region of the cavity. (The gas in the cavity is usually air.) Under these conditions the main requirement is that the ionizing particles originating in the solid lose only a negligible proportion of their energy in crossing the cavity.¶

Although Gray has emphasized the point, it is not generally realized that to fulfill this requirement with air at atmospheric pressure the linear dimensions of the cavity must be very small indeed, especially when heavy ionizing particles are involved. Obviously, according to this requirement, the cavity may be made larger by reducing the pressure of the gas. In fact, use is sometimes made of this relation to see whether the conditions of measurement meet the requirements of the Bragg-Gray principle. If the ionization current is proportional to the gas pressure, it is assumed that the cavity is sufficiently small. This is a satisfactory test for measurements made in a medium in which the flux of ionizing particles is essentially uniform, but it is not sufficient in general. The *essential* requirement substantially as stated by Gray[6] is that the number, energy distribution and directional distribution of the ionizing particles crossing any element of surface of the cavity be the same as they were through the same area before the cavity was introduced. The gas pressure test shows simply that the ionizing particles lose a small fraction of their energy in crossing the cavity—in the range in which there is a linear relation between pressure and ionization current. In a non-uniform radiation field the presence of the cavity does change the ionizing particle flux (number, energy and direction) and the disturbance must be eliminated in order to apply the Bragg-Gray principle. As already pointed out, this can be done by the extrapolation chamber method. Actually this method takes care simultaneously of the requirement that the ionizing particles lose only a negligible proportion of their energy in crossing the cavity. When the gas is of high atomic number and/or the ranges of the ionizing particles in it are short, the accuracy of the extrapolation may be increased by reducing the gas pressure. It should be noted in this connection that in the case of electrons the true stopping power of a material per electron

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¶ In general if this requirement is fulfilled the interaction of the primary radiation with the gas in the cavity is negligible.

decreases slightly with increasing atomic number but scattering increases rapidly. Therefore, the flux of electrons originating in the solid medium may be altered considerably in orientation by a heavy gas in the cavity. Directional disturbances are more apt to occur, also, when the solid is of high and the gas of low atomic number.

Let us consider now the determination of an absorbed dose of gamma rays by means of an extrapolation chamber having tissue equivalent walls and air as the gas. By extrapolation we obtain the number N_a of ion pairs produced during the exposure per gram of air in an infinitesimally thin layer of air by the secondary electrons originating in the walls and traversing the air layer in exactly the same way as they would traverse a correspondingly thinner layer of wall material. If W_a is the appropriate value of the average energy lost by these electrons per ion pair produced in air, then the specific energy absorption in the air layer is $N_a\ W_a$. The specific energy absorption in a layer of tissue equivalent wall having the same mass per unit area as the air layer, resulting from the passage of the same secondary electrons through it, is not equal to $N_a\ W_a$ because an electron of a given energy does not lose the same amount of energy when it traverses the same mass of air or tissue. According to Gray the specific energy absorption E_s in the solid (tissue layer) may be derived from the expression

$$E_s = N_a\ W_a\ S_a^s \tag{1}$$

where S_a^s is the "relative mass stopping power of the solid and gas for electrons" (tissue and air in the present case). Heretofore it has been assumed that W_a is independent of electron energy in the range of interest and equal to 32.5 ev. Values of S_a^s are generally calculated according to the Bethe-Block theory making some simplifying assumptions. There is, however, some uncertainty as to the value of S_a^s appropriate to a given set of circumstances.

As the problem is presented here, S_a^s represents the ratio of the energy lost by the given electron flux (number per unit area, energy spectrum and directional distribution) in a vanishingly thin layer of tissue equivalent solid and in a corresponding layer of air having the same mass per unit area. Let these energies be ΔE_s for the solid layer and ΔE_a for the air layer. Then

$$S_a^s = \frac{\Delta E_s}{\Delta E_a} \tag{2}$$

The energy lost in the layer depends on the amount of matter per unit area, Δm_s or Δm_a, traversed by the electrons and on the rates at which the electrons lose energy per unit mass per unit area. The latter quantity is of the nature of mass stopping power. If we say that $\Delta E_s = [S_s]_{avg}\ \Delta m_s$, then $[S_s]_{avg}$ must represent the average true mass stopping power of the solid for all electrons in the flux in question. Similarly in the case of air if $\Delta E_a = [S_a]_{avg}\ \Delta m_a$ then $[S_a]_{avg}$ must represent the average true mass stopping power of air for the same electron flux. Since for the two layers under consideration $\Delta m_s = \Delta m_a$

$$S_a^s = \frac{\Delta E_s}{\Delta E_a} = \frac{[S_s]_{avg}}{[S_a]_{avg}} \tag{3}$$

Therefore, S_a^s is the ratio of the average true mass stopping powers of the solid and air for all the electrons in the flux in question.

EXPERIMENTAL DETERMINATION OF S_a^s

Referring back to the determination of tissue dose of gamma rays, let us consider the use of the extrapolation chamber with two different gases (one of which is air) under otherwise identical conditions. Since the specific energy absorption in the solid is the same irrespective of the gas used, we now have

$$E_s = N_a\ W_a\ S_a^s = N_g\ W_g\ S_g^s \tag{4}$$

where the subscript g refers to the other gas. Therefore,

$$\frac{N_g}{N_a} = \frac{W_a\ S_a^s}{W_g\ S_g^s} \tag{5}$$

Bearing in mind the significance of S_g^s explained in the preceding section

$$\frac{S_a^s}{S_g^s} = S_a^g \tag{6}$$

Also, W_g, the average energy lost by the electrons in the gap per ion pair produced in the gas, may be expressed in terms of the value, W_a, for air under the same condition, that is $W_a^g = W_g/W_a$. Therefore, $S_a^g = (N_g/N_a) \times W_a^g$. It is more convenient to express N_g/N_a in terms of the ionization currents per gram of gas, I_g and I_a, from which N_g and N_a are derived. Then if $I_a^g = I_g/I_a$

$$S_a^g = I_a^g\ W_a^g \tag{7}$$

This means that the average true mass stopping power of the gas relative to that of air may be determined experimentally in terms of relative ionization currents and relative values of W for any electron flux present in the extrapolation chamber under the conditions appropriate to the determination of absorbed dose. When the flux does not contain electrons of very high energy (that is, when the polarization effect on stopping power is negligible) the mass stopping power of a solid may be taken to be the same as that of a gas of the same atomic composition. Hence $S_a^g = S_a^s = I_a^g\ W_a^g$ and the value of S_a^s in the Bragg-Gray expression may be determined experimentally under the most appropriate conditions. It is hardly necessary to mention that the same procedure applies to any type of ionizing radiation.

For the determination of tissue dose the walls of the extrapolation chamber should be made of tissue equivalent material. A tissue equivalent gas may be obtained by mixing several simple gases in the necessary proportions. Tissue equivalence is most difficult to achieve in the solid walls of the chamber, rather than the gas, when the radiation consists of neutrons.[7]

It is of interest to note that by the method just described values of S_a^g may be determined with considerable accuracy. Ionization currents, of course, can be measured accurately and in general the extrapolation procedure does not offer any difficulties. While it is very difficult to determine absolute values of W for any gas, relative values may be obtained more readily. The problem is further simplified by the fact that W is essentially independent of electron energy, at least in the most useful range. In the writer's laboratory relative values of W in different gases have been determined for natural uranium alpha particles by H. H. Rossi[8] and for S^{35} beta particles by W. Gross,[9] using ionization chambers in which the ionizing particles were completely absorbed. The results are given in Table I and are in good agreement with those published by other experimenters.[10] Nevertheless, additional careful work on this problem is needed, especially as regards the presumed energy independence. It is obvious that the accuracy with which S_a^g can be obtained by the method just described depends almost entirely on the accuracy of W_a^g. (Of course, any gas other than air may be used as a reference point.)

TABLE I. Relative Values of *W* for Different Gases

Gas	S^{35} *beta particles*	*Natural uranium alpha particles*
Air	1.000	1.00
N_2	1.034	1.05
O_2	0.914	0.93
CO_2	0.968	0.97
C_2H_2	0.780	0.78
C_2H_4	0.778	0.80
CH_4	0.820	0.83

To investigate the dependence of S_a^g on electron energy (assuming W_a^g to be constant) it is simpler to incorporate a beta ray emitting isotope in the extrapolation chamber. The procedure then is to determine the extrapolated ionization currents produced by the same beta ray spectrum in different gases. The energy spectrum in the gap is not that of the beta particles as they are emitted, since absorption and scattering take place in the electrodes; but it is the same for all the gases, since the influence of the gas is eliminated by proper extrapolation. In Table II are given the results of such measurements made in the writer's laboratory by R. T. Hersh using S^{35}, Th^{204} and P^{32} as beta ray sources and more recently by Carlotta Paterna using Th^{204} and Ni^{63}. The values of S_a^g are given with more "significant" figures than are justified by the experimental precision and, perhaps, by the assumption that the value of W_a^g obtained with S^{35} beta rays applies to the other beta ray spectra. However, this helps to indicate some slight trends in the variation of S_a^g with respect to beta ray energy. It will be seen that for each gas the agreement is within the experimental error at the higher energies (Th^{204} and P^{32}). At the lower energies the values are less consistent and somewhat higher for the hydrocarbons rich in hydrogen. The work is still going on and will be extended to include beta rays of still lower energies. At this time we wish to emphasize the method rather than the accuracy of the results.

DETERMINATION OF THE ABSOLUTE VALUE OF W

Application of the Bragg-Gray principle to a homogeneous medium with a uniform concentration of beta ray emitting material results in the following relation:

$$3.7 \times 10^{10}\, C\bar{E} = N_a\, W_a\, S_a^s \qquad (8)$$

where C = concentration of radioactive material in curies per gram of the medium; $\bar{E}$ = average energy emitted per disintegration, in ev; N_a = number of ion pairs per gram of air per second; W_a = average energy lost by the beta particles per ion pair produced in air, in ev; and S_a^s = average true mass stopping power of the medium with respect to that of air for the beta particle flux in the medium. It is evident that knowing any four of the quantities involved in this equation the fifth one may be derived.

In his first attempt to make use of this relation in 1946, the writer constructed an extrapolation chamber with thick polystyrene plates containing "uniformly" distributed P^{32}. As was expected, if the radioactive isotope was only in one of the plates, the ionization current was exactly one-half of that obtained with both plates containing the same concentration of the isotope. This was so when both plates were made of the same material (polystyrene). When the material of the inactive plate was different it was

TABLE II. Average Mass Stopping Power of Different Gases Relative to That of Air

Ga	*Source and maximum energy of beta rays in Mev*			
	Ni^{63} *0.063*	S^{35} *0.166*	Th^{204} *0.783*	P^{32} *1.712*
	S_a^g = *Average true mass stopping power relative to air*			
Air	1.000	1.000	1.000	1.000
N_2..............	1.008	1.003	1.006	1.000
O_2	0.983	0.979	0.982	0.979
CO_2	0.968	0.986	0.986	0.994
C_2H_2		1.127	1.126	1.130
C_2H_4	1.251		1.238	1.237
CH_4	1.459	1.461	1.419	1.412

found that the ionization current in the air gap varied as log $(Z + 1)$, in which Z is the atomic number of the material. This made it unnecessary to use electrodes of the same material, which is important in connection with the next development.

The radioactive electrode was prepared by mixing the isotope with powdered polystyrene and then molding. Hence, there was considerable uncertainty as to the concentration of the isotope and its uniformity in the plate. For this reason it was decided to construct an extrapolation chamber in which one electrode was a solution of the desired radioactive isotope. By using upper electrodes of different materials it was found that when the solution contained S^{35} (practically all water) the current in the very narrow air gap of the chamber varied according to the following relation

$$I = 0.511 + 0.555 \log (Z + 1) \qquad (9)$$

in which Z = atomic number of the material of the inactive plate. It was found also that this relation holds for compounds, provided an effective Z is calculated according to the relation:

$$\log (Z_{eff} + 1) = f_1 \log (Z_1 + 1) + f_2 \log (Z_2 + 1) + \ldots . f_n \log (Z_n + 1) \qquad (10)$$

where f_1 is the fractional mass of the compound contributed by the element of atomic number Z_1, etc. For water $Z_{eff} = 6.61$ which substituted for Z in the above equation makes $I = 1$. Thus the correction factor for measurements made with an active electrode of any material may be readily calculated.

If the radioactive isotope can be measured accurately in curies, its concentration C in the liquid electrode of the extrapolation chamber can be adjusted to any desired and *known* value. If the average energy per disintegration $\bar{E}$ is also known, the energy emitted per second per gram of solution can be calculated readily (left side of first equation given in this section). N_a is known from the measurement of the ionization current (extrapolated value). S_a^s is now the average mass stopping power of the solution (water) with respect to that of air for the beta particle flux in the solution. If S_a^s is known then W_a can be calculated.

It was thought desirable to determine W_a by this method for the beta rays of S^{35}. Since this isotope cannot be measured accurately in curies it was thought best to determine its energy emission by a calorimetric method. This has the further advantage that it is not necessary to know $\bar{E}$ and the left side of the equation is then known essentially with the accuracy of the calorimetric measurement. Dr. W. B. Mann of the US Bureau of Standards has developed a very satisfactory calorimeter for this purpose[11] and he kindly consented to measure the rate of energy emission of about 450 mc of S^{35} in the form of sodium sulphate. This was then dissolved and kept in such a way that the energy emission rate per gram of solution is accurately known even if evaporation of water takes place. A preliminary determination of W_a by this method with this solution gives a value of 35 ev. Since we have not yet determined the value of S_a^s for water vapor and S^{35} beta rays by the method outlined above, an estimated value of 1.14 was used in the calculation. This work has been carried out by W. Gross and C. L. Wingate and will be continued with further refinements. Using other isotopes as well, it should be possible to obtain reliable values of W_a and at the same time investigate its possible dependence on electron energy, at least in widely separated energy regions.

The foregoing discussion is intended to clarify the application of the Bragg-Gray relation to a variety of radiation dosimetry problems. Using the extrapolation chamber method the necessary requirements may be fulfilled adequately even in cases in which the flux of ionizing particles is very non-uniform. The value of W to be used in the Bragg-Gray relation must represent the average energy lost by all the ionizing particles crossing the surface of interest in the solid per ion pair produced in the gas, with the proviso that the "energy lost" must also be locally available. S_g^s, no matter what it is called, must represent the ratio of the energy absorbed (that is, locally available) by a very thin layer of the solid from all the ionizing particles crossing it at the surface of interest and the energy absorbed by a corresponding layer of the gas having the same mass per unit area. If the value of S_g^s appropriate to a given set of circumstances is determined by calculation, this condition must be met. By methods described in this paper the required values of W and S_g^s may be obtained experimentally for the ionizing particle flux of interest or a close approximation thereto.

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Recent Developments in the Scintillation Counter Field

By G. A. Morton,* USA

Surveying the current literature in physics, one encounters a group of papers discussing the lifetime of positrons in various solids. It is found, for example, that the lifetime of the positron in a metal at room temperature is 1.5×10^{-10} second, while the corresponding lifetime for the same metal cooled to the point where it becomes superconducting is much longer being of the order of 6.5×10^{-10} second, a lifetime comparable to that of positrons in an insulator. Measurements of these fantastically small intervals of time and of the high time resolution implied by them has only been possible since the development of the scintillation counter. If this superior time resolution and high speed were the only attribute of the scintillation counter as a nuclear particle detector, it would still be a very important instrument in the field of nuclear research.

The scintillation counter, however, has a number of equally important characteristics which, taken together, make it one of the most important instruments for the detection of energetic particles in the nuclear field. Indeed, its importance is such that it is safe to say that nearly all laboratories engaged in research in this field employ one or more scintillation counters and a very large percentage of the articles published in the field describe experiments requiring these detectors.

The importance of the scintillation counter is not restricted to the realm of nuclear research alone. Instruments of this type are used in large numbers in prospecting for ores containing uranium or thorium and in prospecting for oil. They also find application in the fields of radio-biology, radio-chemistry, tracer technology, radiation protection and in the nuclear power field.

The characteristics of the scintillation counter which give it this importance are: (a) its high sensitivity to all forms of nuclear radiation; (b) its ability to discriminate and measure the energy of the incident nuclear particle; and (c) its high speed and excellent time resolution capabilities. Furthermore, scintillation counters can be made rugged and reliable. The class of instrument is flexible enough so that it can range in form from small compact units suitable for aerial work or lowering into oil well drillings up to instruments where the sensitive volume includes several cubic meters.

However, before discussing these characteristics, the general aspects of the counter and its principal components must be considered. The scintillation counter consists of a transparent phosphor scintillator, a photomultiplier and the presentation circuitry. When a nuclear particle or photon is absorbed by the phosphor, a flash of light is produced. This light is directed onto the photocathode of the multiplier where it causes the release of photoelectrons. The photoelectrons enter the multiplier structure where they are amplified by the cascaded secondary emission dynodes of the tube and appear at the output as a current pulse. This pulse is analyzed for size, duration, wave shape and time of occurrence by the succeeding circuitry to extract the required information. The two principal components of the scintillation counter, namely, the multiplier phototube and the scintillator must be examined in some detail.

THE MULTIPLIER PHOTOTUBE

The photomultiplier provides a means for amplifying the minute photocurrent involved in scintillation counting up to the point where it can be handled by existing amplifiers and circuitry. Any attempt to collect the original photocurrent pulse produced by a scintillation and to amplify this current by means of a thermionic amplifier would be unsuccessful because the size of the pulse is so small that it would be completely lost in amplifier noise. In a photomultiplier, the photoelectrons released from the cathode are directed onto a surface so sensitized that each electron which strikes it causes the release of several secondary electrons. The electrons emitted by this first surface or first dynode are focused onto a second similar surface where again each incident electron produces a number of secondary electrons. This process is repeated on successive dynodes until the current is built up to the desired level. It is evident that if there are k dynodes, each having a secondary emission ratio σ, a primary photocurrent i_p will produce an output current $i_o = i_p \sigma^k$.

As a result of the stimulus provided by the scintillation counter, a number of new photomultipliers have been developed. Table I gives a representative list of multipliers manufactured in the United States. Only three pre-date the scintillation counter. These tubes differ in cathode area, spectral response, method of collecting photoelectrons and dynode structure and activation. While they fulfill many of the requirements of the counter, there is still need

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* RCA Laboratories.

for further improvement, and research and development are in progress on these tubes. Table II lists a few of the new developmental tubes designed to give still better performance from scintillation counters. These will be treated in greater detail as the discussion proceeds.

If any one element of the photomultiplier were to be designated as vital, it would be the photocathode. The importance of the photocathode is self-evident, but will be put on a quantitative basis in the discussions of energy and time measurements with scintillation counters. The importance of the photocathode has prompted a great deal of work aimed toward a better basic understanding of the phenomenon of photo emission and toward the development of improved cathodes. Figure 1 shows the spectral response of a number of new cathodes under investigation at RCA Laboratories.[1] Curve III is included for comparison and is the response of a typical S-11 semitransparent cesium antimonide surface. Curve V is the response of a tri-alkali cathode which is cesium activated sodium potassium antimonide. The sensitivity of this cathode is very high, exceeding 200 μa/lumen response to a tungsten source heated to a brightness temperature of 2870°K. Its response extends well into the red portion of the spectrum, however, and therefore when used in a scintillation counter must be cooled to reduce thermionic emission. Cathode IV is of considerable interest for multiplier phototubes employed in scintillation counters. It has essentially the same spectral response as the conventional cesium antimonide, but employs no cesium. This is a considerable advantage in the high voltage tubes used for fast counting, in that wherever cesium is used its high vapor pressure and its tendency to form low work function surfaces make electrical leakage and cold discharge problems very severe. Research on photocathodes is actively continuing and preliminary results already indicate that new improved photoemitters can be expected from this work.

Two types of secondary emitting surfaces are used in the present commercial multipliers, namely, those activated with cesium antimonide and those activated with magnesium-oxide-silver. The former generally have a higher secondary emission ratio for a given incident electron velocity, while the latter have the advantage of withstanding a higher outgassing bake which increases tube stability.

The problem of directing photoelectrons from the cathode of a photomultiplier into the dynode structure is not negligible. The general philosophy here is to design an electron lens system in such a way that the first dynode is at the exit pupil of the system. The solution is complicated by the large spherical aberration found in most simple electrostatic electron lens systems. However, fairly adequate systems have been worked out.

The dynode system itself is an electron optical system for drawing secondary electrons away from one dynode and causing them to impinge upon the next succeeding dynode with a minimum loss of electrons.[2] A number of different electron optical systems have been devised for doing this. Two types of systems are used in the commercial tubes shown in Table I. Those produced by RCA use a system of electrostatically focused dynodes arranged in a circular array. Figure 2, which illustrates photographically and in diagram the RCA Type 6342, shows the dynode arrangement.[3] This design of dynode structure gives fairly strong fields drawing the secondary electrons away from each surface. As a consequence, the transit time and the transit time spread between each dynode is small and the tube is capable of resolving very small time intervals. Figure 3 illustrates a DuMont 6292 and shows the box type dynode structure used in all DuMont tubes.[4] With this sys-

TABLE I. Characteristics (Approximate) of Representative Commercial Photomultipliers

Type	Max. outside dimensions inches dia. length	Cathode			Dynode system				Over-all voltage	Light equivalent of noise (lumens)
		area cm	response	av. sensitivity (μa/lumen)	no. of dynodes	type	material	gain	(volts)	
RCA										
5819	$2\frac{1}{4}\times5\frac{13}{16}$	14.2	S-11	50	10	circular focused	Cs_3Sb	2.3×10^6	1250	7×10^{-11}
6342	$2\frac{1}{4}\times5\frac{13}{16}$	14.2	S-11	60	10		AgMg	0.55×10^6	1500	7×10^{-12}
6655	$2\frac{1}{4}\times5\frac{13}{16}$	14.2	S-11	50	10		Cs_3Sb	2.3×10^6	1250	7×10^{-12}
6199	$1\frac{9}{16}\times4\frac{9}{16}$	7.75	S-11	45	10		Cs_3Sb	2.8×10^6	1250	4×10^{-12}
2020	$2\frac{1}{4}\times5\frac{13}{16}$	14.2	S-11	60	10		Cs_3Sb	2.8×10^6	1250	7×10^{-12}
6217	$2\frac{1}{4}\times5\frac{13}{16}$	14.2	S-10	40	10		Cs_3Sb	2.8×10^6	1250	4×10^{-11}
6372	$2\frac{9}{16}\times7\frac{1}{4}$	80	S-11	33	10		Cs_3Sb	2.5×10^6	1200	1×10^{-11}
1P21	$1\frac{5}{16}\times3\frac{11}{16}$	1.9*	S-11	40	9		Cs_3Sb	8.3×10^6	1250	5×10^{-13}
1P28	$1\frac{5}{16}\times3\frac{11}{16}$	1.9*	S-5	40	9		Cs_3Sb	5.3×10^6	1250	7.5×10^{-13}
DuMont										
6291	$1\frac{1}{2}\times4\frac{1}{4}$	6.4	S-11	60	10	box	AgMg	2×10^6	2100	—
6292	$2\frac{1}{16}\times5\frac{5}{8}$	13.4	S-11	60	10		AgMg	2×10^6	2100	—
6363	$3\times6\frac{1}{8}$	31.4	S-11	60	10		AgMg	2×10^6	1800	—
6364	$5\frac{1}{4}\times7\frac{1}{2}$	88.8	S-11	60	10		AgMg	2×10^6	1800	—
6467	$1\frac{3}{4}\times4\frac{1}{2}$	5.1	S-11	60	10		AgMg	2×10^6	1800	—
6365	$\frac{3}{4}\times2\frac{3}{4}$	1.26	S-11	50	6		AgMg	3×10^3	1200	—

* Projected area.

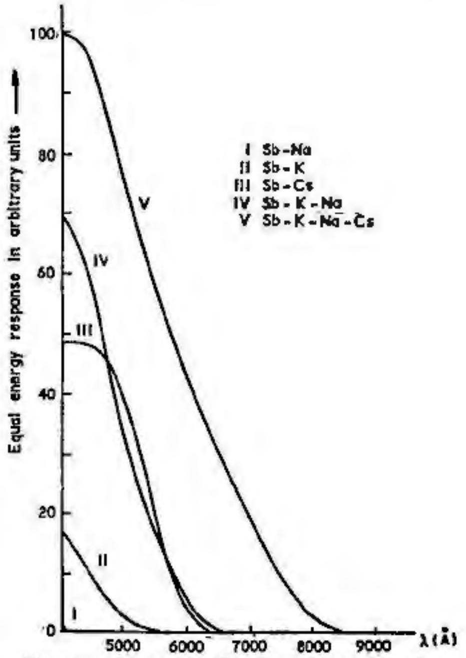


Figure 1. Response of various photocathodes

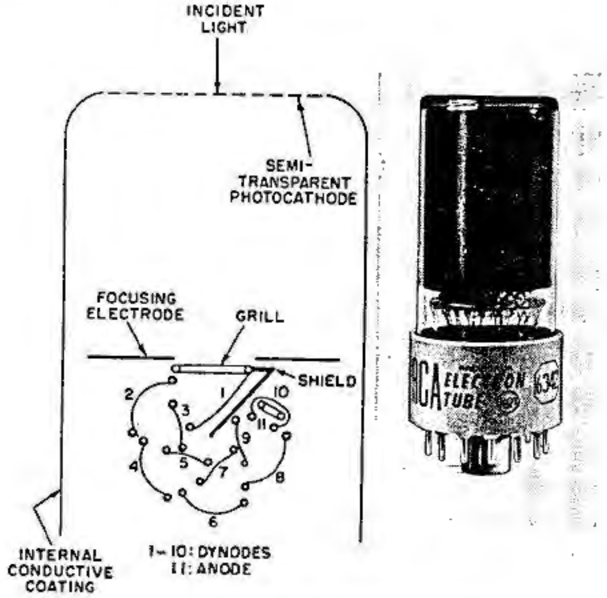


Figure 2. Multiplier phototube RCA 6342

tem, the field strengths obtainable at the secondary emitting surfaces are rather low so that the time resolution capabilities are not as good as for tubes using a focused structure. However, this type of dynode system is much less critical in interstage voltage adjustment than the focused system. This is a considerable advantage in many applications. Figure 4 is a photograph showing some representative tubes of the RCA listing, while Fig. 5 shows a corresponding group of DuMont multiplier phototubes. All of these tubes are giving good service in the scintillation counter field. Although many of the tubes listed in Table I have only become available in the last two years, they are, nevertheless, all in production and commercially obtainable.

The group of tubes listed in Table II are experimental and developmental tubes being investigated. Tube Type H5723 is illustrated in Fig. 6a. This tube has a diameter of ¾ inch and an over-all length of about 3 inches. The dynode configuration is noteworthy in that, although it has a simple circular cross section, it nevertheless satisfies the three principal requirements for the electron optics of a multiplier, namely, to (*a*) provide a strong field to accelerate electrons away from the dynode surface, (*b*) focus the emitted electrons into an area no larger than the area from which they leave, and (*c*) allow no free

TABLE II. Representative Developmental and Experimental Multipliers

Developmental Production

DuMont

Type	*Outside dimensions (inches)* *dia.* × *length*	*Area cm.*	*Response*	*Sensitivity*	*No. of dynodes*	*Type*	*Material*	*Gain*	*Over-all voltage*
K1209	5½×8½	88.8	S-11	60	12	box	AgMg	2×10^5	1330
K1213	3⅓×6⅜	31.4	S-11	60	12	box	AgMg	2×10^5	1330
K1295	2$\frac{1}{16}$×6¾	13.4	S-11	60	12	box	AgMg	2×10^5	1330
K1328	15⅞×15	995	S-11	40	12	box	AgMg	8×10^5	1460

RCA

Type	*Area cm*	*Response*	*Sensitivity*	*No. of dynodes*	*Type*	*Material*	*Gain*	*Max. current (instantaneous)*	*Remarks*
					Developmental (Lancaster)				
C7187A	14.2	S-11	60	14	linear (B)	AgMg	10^8–10^9	0.3	Prototype H5914
					Experimental (Princeton)				
H5723	1.6	S-11	40	9	linear (A)	AgMg	3×10^5	—	
H4646	3.2	S-11	30	16	linear (B)	AgMg	10^9	0.2	
H6687	14.2	S-11	40	14	linear	AgMg	10^8–10^9	1.1	Like H5914 with space charge electrodes
H5037	62	S-11	60	10	circular	AgMg	10^6	—	
H6699	410	S-11	40	10	linear (A)	AgMg	10^6	—	

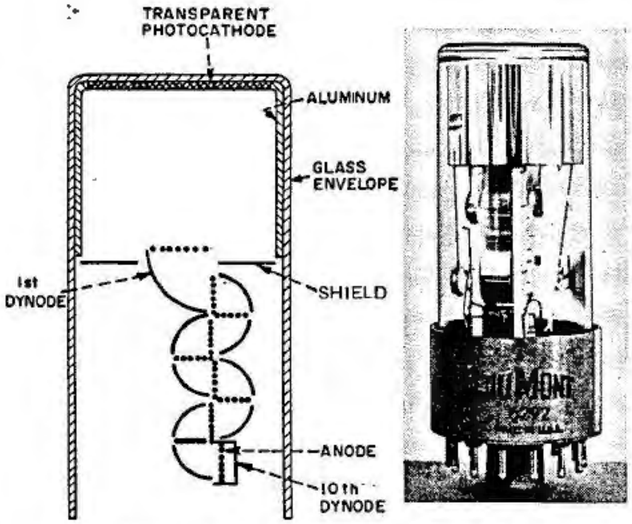


Figure 3. Multiplier phototube Dumont 6292

path from cathode to anode which would otherwise cause serious ion feedback. This simple geometric shape is easy to manufacture and stable under the heat treatment which the dynodes must receive during the processing of the tube. Small tubes of this type are useful in scintillation counter applications where space is at a premium.

The next tube is a high gain multiplier phototube. This is the H4646 whose structure is shown in Fig. 6b.[5] It has a small cathode arranged to have strong electrostatic fields at the photoemitting surface in order to minimize transit time spread of the photoelectrons. The structure is a linear array of dynodes having a rather complicated configuration. The linear structure makes it possible to use as many dynodes as desired, and these tubes have been built with 14 and 16 dynodes. With 16 dynodes, a single electron from the photocathode can be multiplied up to the point where it will produce a space charge limited current of about 0.2 ampere at the anode. The time characteristics of this tube have been measured by R. K. Swank, who found that a group of electrons leaving the cathode essentially simultaneously are spread into a pulse whose duration between the half maximum points is between 3 and 4 millimicroseconds.

Tube type H5914, shown in Fig. 6c, is a slightly slower tube than the preceding tube but has the advantage of a larger photocathode. This tube employs a 14-stage structure with dynodes of the type used in the H4646. An instantaneous space charge limited current of about 0.3 ampere can be obtained from this tube. It is the experimental prototype of the C7187A.

The H6687 is like the H5914 except that the four final stages have been modified as shown in Fig. 6d to include field forming electrodes operated at a more positive potential than the dynode with which they are associated in order to increase the space charge limited current. This modified structure permits a peak output current of more than an ampere. A high peak current output is very desirable for fast counting and measurements involving the resolution of small time intervals since the circuitry employed is necessarily low impedance. With output currents in the 0.1 to 1 ampere range fed into 100-ohm coaxial lines, it is possible to operate coincidence circuits, pulse height selectors, oscilloscopes, etc. without intervening distributed amplifiers.

The next two tubes provide large area photocathodes, needed where large scintillators are to be used, but are arranged to minimize the loss in time resolution which is associated in general with tubes having large cathodes. Figure 7a illustrates the H5037 which has a 3½ inch diameter emitting area.[5] The electron optical system for collecting photoelectrons is very similar to the electron optical system of the conventional image converter tube. The first dynode is located at the exit pupil of the lens system. If such a system could be made free from aberrations, the diameter of the electron bundle of the first dynode would be determined solely by the initial velocity of the photoelectrons. Furthermore, the point of arrival on the dynode would be completely independent of the point of origin of the photoelectrons. In a practical lens system there is considerable spherical aberration. If a small spot of light is focused on the cathode and moved along a diameter from one side to the other, the landing spot on the first dynode is found to make three transits across a spot on the dynode whose diameter is 0.1 or 0.2 inch. The multiplier structure for this tube is a conventional 10-stage circular dynode system.

The final tube, Type H6699, shown as Fig. 7b, has a 9-inch diameter cathode. The electron lens system for collecting photoelectrons is very similar to that of the H5037 in principle but is modified in detail of design in order to accommodate a more practical envelope shape. This tube uses a 10-stage linear structure which is an enlarged version of that in the H5723.

The above discussion of work on developmental and experimental tubes is not intended to be exhaustive but merely to give an indication of the direction, along which some of this work is proceeding. Advanced development work on photomultipliers[3, 4, 5, 6] is being carried on in a number of laboratories including those of the Radio Corporation of America, Allen B. DuMont, Inc., and others. Continued improvement in photomultipliers for scintillation counting is assured as this work proceeds.

THE SCINTILLATOR

A discussion of the processes involved in the production of scintillations when high energy particles are incident on a phosphor can be conveniently divided into two parts: the first considers the way in which the material is excited by the energy of the incident particle, while the second deals with the generation of fluorescence in the excited material.

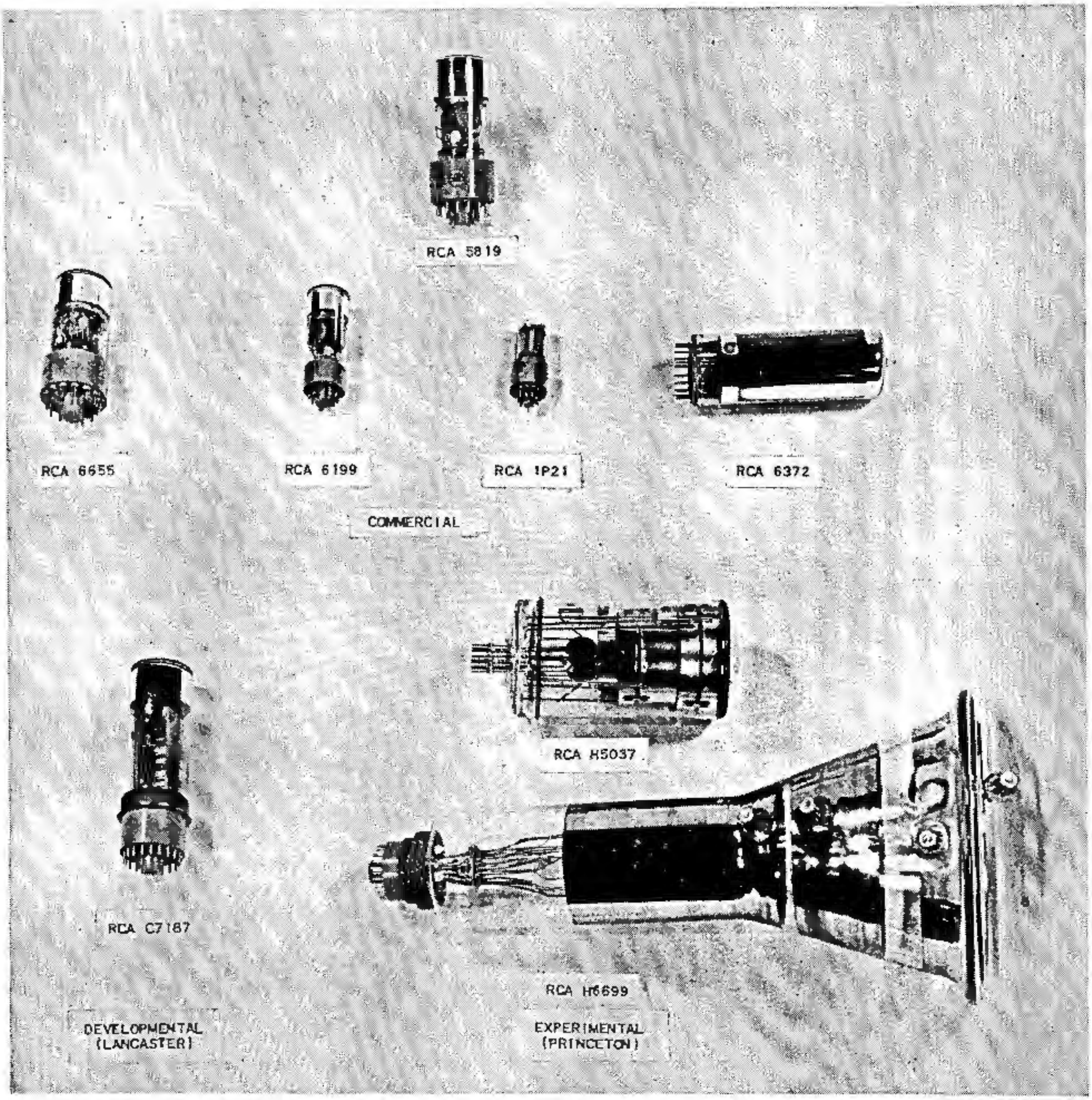


Figure 4. Representative RCA multiplier phototubes

It is probable that almost all, if not all, the conversion of energy of the incident nuclear particles into excitation energy is the result of the production of high energy electrons in the scintillator which, in turn, produce the excitation. For beta rays, passage of the fast electron through the material raises electrons in the scintillator into excited states from which they decay later to produce fluorescence. For gamma rays, the conversion is less direct. The gamma ray can give up its energy to the material by (*a*) a Compton collision; (*b*) photoelectric capture; (*c*) pair production. The Compton process is the predominant one for light materials (e.g., organic phosphors) and low energy gamma rays, i.e., up to 2 or 3 Mev. Here the gamma-ray photon interacts with a relatively free electron imparting energy to it. Photoelectric absorption occurs when the gamma-ray photon interacts with a tightly bound electron. The chance of photoelectric absorption increases rapidly with the atomic number of the atoms of the material, the probability being proportional to the fourth power of the atomic number. Pair production is impossible for gamma-ray energies below 1 Mev and is relatively unimportant for gamma-ray energies under 5 Mev. For very high energy gamma rays, however, it may be one of the major processes involved in absorption of energy.

The excitation of phosphors by heavier particles

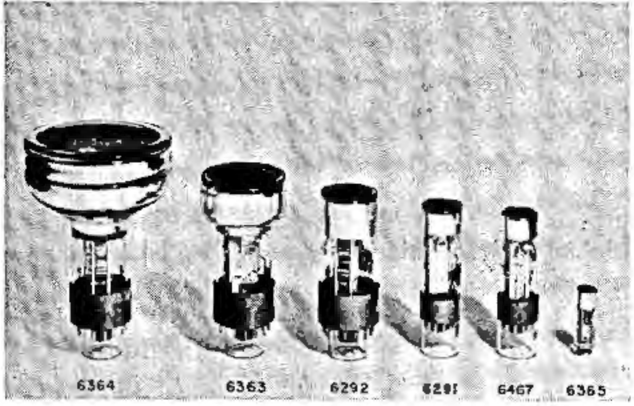


Figure 5. Commercial Dumont multipliers

such as protons, mesons and alpha particles is a good deal more complicated. In all these cases, probably high energy electrons serve as an intermediate between the excitation of the crystal and the giving up of energy to the material on the part of the incident particle.

Excitation of scintillators by neutrons is a rather special problem since the neutron has no electric field associated with it. For low energy neutrons, the process usually involves a nuclear reaction between the neutron and an atom of the phosphor with a subsequent production of beta particle, gamma ray or fission fragment.[7] For high energy neutrons, the transfer may be effected by using a scintillator having hydrogenous material as a constituent and using the knocked-on energy of the protons with which the neutrons collide as a means of the transfer of energy from the neutron to the lattice.[8, 9]

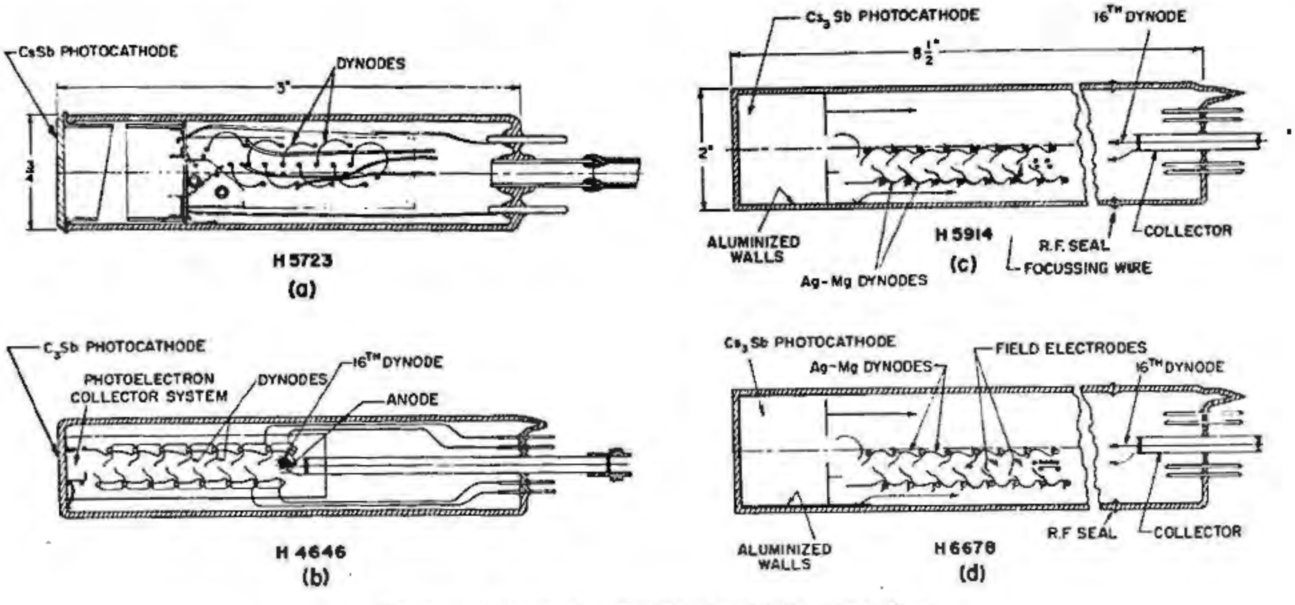
From the standpoint of fluorescence, scintillators used in scintillation counters can be conveniently divided into two broad classifications, namely, inorganic crystals and organic scintillators. This separation is made because of the very different mechanism of fluorescence involved in the two types of material. Organic scintillators, in turn, may be subdivided into single crystal phosphors and solution scintillators. Solution scintillators are comprised of a solvent in which the primary excitation occurs and an activator or fluorescent solute which emits the light actuating the photomultiplier. The solvent may be a crystal, plastic or liquid, all three having been used very successfully in scintillation counting.

A. Inorganic Scintillators

The excitation-fluorescence process in inorganic phosphors involves the crystal lattice as a whole rather than individual molecules. For photoconductive phosphors the excitation energy lifts electrons from the valence band to the conduction band. The electrons or holes then move through the crystal until they encounter fluorescent re-combination centers. Where a non-photoconductive phosphor is involved, the excitation probably produces excitons which again move through the crystal until they encounter an appropriate activator center. This process can be quite efficient but, in general, leads to fluorescence having a rather long time constant.

Thallium-activated sodium iodide is by far the most widely used of the inorganic scintillators. This material which was introduced by R. Hofstadter can be quite easily grown into large, clear single crystals.[10] These crystals are now in commercial production and can be obtained in a wide range of sizes and shapes.

Sodium iodide is one of the most efficient phosphors known for scintillation counter work. A good crystal will yield one useful photon per each 30 to 50 electron volt energy of the incident particle; in other words, a 1 Mev beta particle will generate 20,000 to 30,000 photons. The spectral response out-



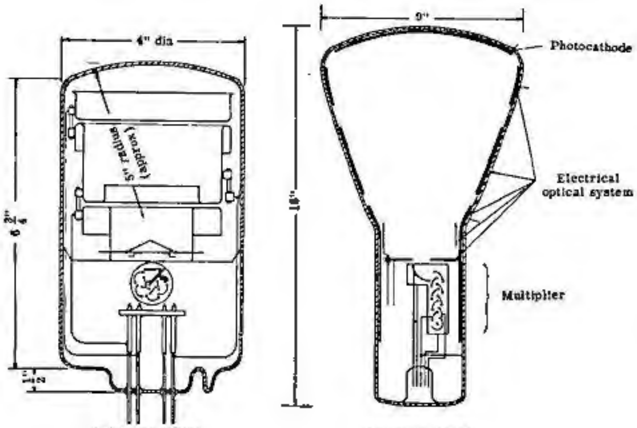
H 5723
(a)

H 4646
(b)

H 5914
(c)

H 6678
(d)

Figure 6. Diagram of experimental multiplier phototubes



(a) TUBE H5037 (b) TUBE H6699

Figure 7. Large area cathode multipliers

put of this phosphor is a band some 800 Ångstroms wide centering around 4100 Ångstroms. This corresponds very favorably to the spectral peak of a cesium antimony photocathode. The decay time of this material, though small compared to a good many inorganic phosphors such as zinc sulfide, zinc oxide, etc., is very much longer than that of organic materials. The measured decay time is of the order of 200 or 300 millimicro-seconds.

One of the important attributes of this material for gamma-ray detection is its very high density. A relatively small amount of the sodium iodide will completely absorb gamma rays in the ½ to 1 Mev range. Furthermore, the high atomic number of iodine means that a fairly large percentage of the energy given up to the crystal by a gamma ray will be in the form of photoelectric excitation. This, in turn, leads to light flashes which are strictly proportional to the energy of the incident photon. As will be discussed later, sodium iodide is for this reason very frequently used in gamma-ray spectrometers.

Sodium iodide, however, does have two disadvantages. The first is that its optical index of refraction is very high, being of the order of 1.8. The problem of making efficient optical coupling between the photocathode and the scintillator is therefore quite difficult. It is a common practice to cement the crystal to a glass or lucite "light-piper" which, in turn, is cemented to the multiplier cathode. Even here the reflection loss at the crystal light-piper interface is severe. In order to conserve some of the light lost by the first reflection, the sides and end of the crystal are coated with a highly reflecting matte finish. The second difficulty is that the material is chemically unstable in the presence of water vapor. Even when the humidity is quite low, the material cannot be exposed to air for more than a very short time without risk of damage. The moisture causes the dissociation of iodine which colors the crystal slightly yellow. Since the emission is entirely short wavelength blue light, even a small amount of yellow discoloration makes the material quite opaque to its fluorescent radiation. Techniques, however, have been developed for mounting these crystals in air-tight containers sealed with a glass or plastic disk. A properly mounted commercial crystal will retain its efficiency satisfactorily for a long time.

A large number of other inorganic crystals have been studied as scintillators. Cadmium and calcium tungstates may offer interesting possibilities. Both of these have a high density and contain a high atomic number constituent.[11] Measurements to date indicate that while their scintillation efficiency is high, the time constant of the materials is very long which has made them rather unsatisfactory for scintillation counter use. The difficulty of growing these crystals is rather formidable because they are very refractory. More experimental work on the preparation and purification of the materials must be carried out before the question can be answered as to whether the long time constant is fundamental to the materials. Cesium iodide has shown some interesting properties as a scintillator.[12] It is a good deal faster than sodium iodide. In fact, its speed is such as to indicate that a different mechanism may be involved. The high atomic number of its two components enhances its photoelectric excitation and gives it a high absorption coefficient for gamma rays.

The lithium halides have been investigated as possible scintillators for thermal neutrons. Here the excitation depends upon a nuclear interaction between lithium and slow neutrons.

Inorganic phosphors have been used in scintillators for the detection of alpha particles.[13] Here the penetration is very small and the specific ionization is large. Where high speed is not necessary, screens formed of a rather thin layer of silver-activated zinc sulfide have given very good performance.

B. Organic Scintillators

The fluorescent processes in organic scintillators are quite different than those in the inorganic materials discussed above. Most of the organic phosphors contain in their molecules carbon rings having conjugated double bonds involving π electrons. The molecules in organic crystals are rather loosely held together so that the bonding has little effect on the emission spectra of the material.

As with inorganic materials, the excitation may occur at one point in the crystal and be propagated to another molecule at some distance away where the fluorescence is actually emitted. The excitation transfer in organic materials appears to be due to photons of considerably shorter wavelength than the fluorescent photons or by some form of intermolecular coupling. In either case, the velocity of propagation is essentially that of light and, as a consequence, the fluorescent decay times are very short. The efficiency of some of the organic phosphors is fairly high but is lower than that of sodium iodide. Their reasonably high efficiency coupled with the extremely short decay time makes them very valuable scintillators wherever time measurements are involved. Because of their importance, these phosphors have been investigated extensively by such workers as Kallmann,[14] Birks,[15] Hayes,[16] Liebson,[17] Reynolds,[18] Swank[19] and others. This work, which is continuing, has given a fairly good fundamental and practical understanding of this type of scintillator.

Organic scintillators may be in the form of single crystals or may be in the form of an activator solute in a liquid or plastic solvent. Both play important roles in scintillation counting, the former being the more efficient but with a longer decay time. Crystals are used for small and moderate sized, fairly fast, efficient counters, while solutions are used for the ultimate in speed and for very large counters. Before proceeding with an account of practical organic scintillators, it would be well to review some of the hypotheses concerning the physical mechanisms in solution scintillators.

One of the characteristics of these scintillators is that they show a relatively high efficiency when only a minute amount of the fluorescent solute is present. An estimate shows immediately that, if the fluorescence is the result of interaction of the nuclear particle with the solute molecules, their quantum efficiency must be far in excess of 100%.[19] This is, of course, contrary to all expectation and is not consistent with their behavior otherwise. Consequently, it is necessary to assume that the excitation is the result of the interaction between the nuclear particle and the solvent molecules and that this excitation is transferred to the solute molecule where it is converted into visible light. The mechanism of the transfer of energy from the solvent molecules to the solute has been studied extensively by Kallmann, Swank, Birks and others. There seems to be little doubt that photon exchange is in some measure involved. There is some question as to whether the entire energy transfer can be accounted for on the basis of photon transport and some workers feel that at least part of the energy is transferred by some form of intermolecular coupling. However, evidence is accumulating indicating that the whole effect can be accounted for on the basis of photon exchange.

J. B. Birks has discussed this matter in some detail and shows that all of the effects which have been observed in solution phosphors can be accounted for on the basis of photon exchange if it is assumed that at least a fraction of the photons are of an extremely short wavelength (i.e., in the neighborhood of 1000 Angstroms) where the penetration through the solvent is extremely small.[20] The mechanism postulated is very similar to that used by J. Q. Umburger[21] and others to explain the fluorescent behavior of fluoresceine solutions. In this model, short wavelength ultraviolet photons are emitted by the solvent as a result of the excitation from the nuclear particle. Each photon is absorbed by the solvent after traveling a short distance through it and then after a short time delay is re-emitted. This process is repeated until eventually the photon interacts with an activator solute molecule which it excites. The return of the activator molecule to ground state gives rise to the longer wavelength photon which is one of the photons making up the scintillation.

The quantum efficiency of excitation and re-emission by the solvent is less than unity, and also each re-emission has a finite decay time. Consequently, when the cascade process is repeated many times, the efficiency of the scintillator decreases and its time constant increases. The relationship between scintillator efficiency and decay time is given by:[22]

$$\begin{aligned} \log S &= \log A + Bt_I \\ B &= \log q_0/t_0 \end{aligned} \qquad (1)$$

where S is the pulse height, q_0 re-emissinn quantum efficiency, t_0 re-emission time, and t_I decay time. Birks has used this relationship to explain the variation of scintillation decay time with temperature for certain organic scintillators as determined by Liebson.[23]

The efficiency of a scintillator increases as the concentration of activator solute is increased up to a certain point. During this time, the time constant of the material decreases. Beyond this point, the efficiency of the scintillator again decreases as the concentration is increased. In this range of concentration, the time constant is essentially constant. Figures 8a and 8b illustrate this behavior for tetraphenylbutadiene in polystyrene.[24] The falling off of the efficiency with high concentration of the solute is probably the result of absorbtion of the visible photons by the solute with little or no re-emission.

Birks' assumption of photons of two different energies involved in the radiative transfer is adequate to explain the results obtained by Swank and Buck[24] in their experiment comparing the variation of scintillation efficiency with concentration for tetraphenylbutadiene in polystyrene which showed an abrupt saturation when the solution phosphor was covered with a layer of polystyrene impervious to the exciting alpha particles, whereas when the alpha particles were allowed to strike the solution scintillator directly, the saturation was much more gradual and at a higher concentration of the solute. In this instance, even the thin layer of pure polystyrene was sufficient to absorb the short wave component of the transfer photons.

It is very probable that this same photon transfer mechanism occurs in single crystal organic phosphors. Evidence for this is the variation of fluorescent decay time with temperature and also the fact that the decay time for ultraviolet excitation has been shown by Liebson and co-workers[17] to be invariably shorter than the decay time for gamma stimulated fluorescence.

C. Single-Crystal Scintillators

Single crystal anthracene is the most widely used organic crystal scintillator. Anthracene consists of three benzene rings linked together in a straight line. The material forms a monoclinic crystal having a

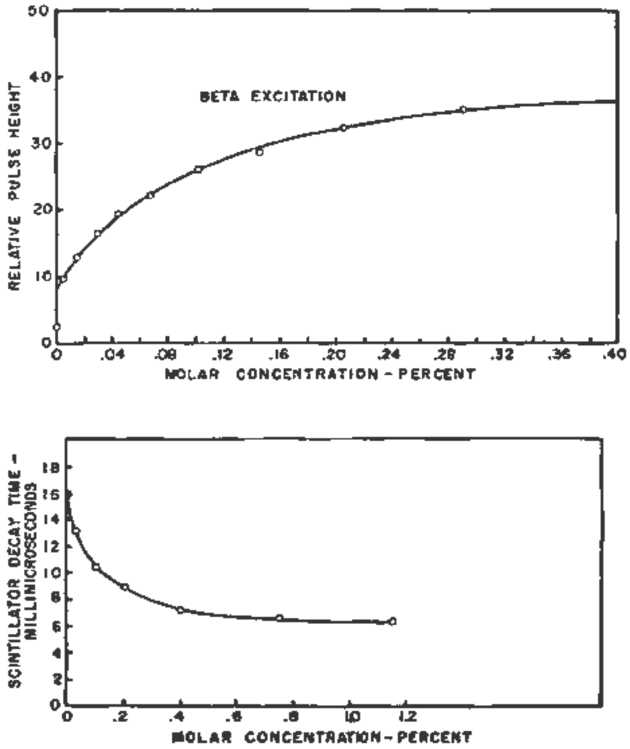


Figure 8. Response and time constant of tetraphenylbutadiene in polystyrene

specific gravity of 1.25 and an index of refraction of 1.5948. The melting point of the material is high enough to be practical for most scintillation counter applications but low enough so that the material can be easily melted for crystal growing. The growing procedure most commonly used is the Stockbarger method, the material being sealed in a vacuum crucible. If care is taken with the purification and cleanliness of the materials, large water-clear crystals can be readily grown. The efficiency of this material to beta ray excitation is about 40 to 50% that of sodium iodide. Its decay time is about 36 millimicroseconds. The decay is temperature dependent, and Fig. 9 shows a variation of decay time with temperature as measured by Liebson. The emission spectrum is a series of bands centering around 4400 Ångstroms. It is, therefore, well suited to the spectral response of a cesium antimony activated multiplier phototube.

A second organic crystal which finds wide application in scintillation counting is trans-stilbene. Again, it is a material which can be easily crystallized into large water-clear masses. The scintillation efficiency of stilbene is considerably lower than that of anthracene, but for many applications this is compensated for by its very short decay time. The efficiency of trans-stilbene relative to anthracene is about 30%, while its decay time is of the order of 6 millimicroseconds at room temperature. As with anthracene, the decay time decreases as its temperature is lowered. Because of its high speed, it is widely used for coincidence work.

D. Solution Scintillators

High-energy particle physics, both for cosmic ray work and with high energy machines, frequently requires scintillators occupying many cubic feet of volume. Similarly, there are a number of problems in biophysics where it is desirable to have very large scintillators. It is both expensive and impractical to grow crystals large enough for such applications. However, solution scintillators appear to be a very practical alternative and are coming into wide use. H. Kallmann[25] at New York University was among the first to describe a number of types of this interesting scintillator. He and other workers[26, 27] have found many organic liquid solvents which will serve as the scintillator base and an almost equal number of phosphor activators. Toluene, xylene and phenylcyclohexane have all been used very effectively as the solvent, while terphenyl, diphenyloxazole and tetraphenylbutadiene are all effective as solutes. The efficiency of a solution scintillator can frequently be improved by the addition of a so-called wavelength shifter. For example, a liquid scintillator with phenylcyclohexane as the solvent and 3 grams per liter terphenyl as the solute plus 0.01 gram per liter diphenylhexatriene yields about 30% greater pulse height than does the material without the wavelength shifter. However, the use of the wavelength shifter decreases the speed of the scintillator in almost every case.

As has already been mentioned, the solvent may be a plastic with a solute dissolved in it instead of a liquid. Polyvinyltoluene and polystyrene are very effective plastic solvents. Figure 10 shows the pulse height as a function of concentration for a number of solutes in polystyrene.[27] In polyvinyltoluene, the response is in general similar but slightly higher. These plastic phosphors have a very great practical value because they can be made into rigid masses of very clear material of any size or shape desired.

A summary of the more important scintillators together with the wave length of their peak emission, relative efficiency to beta-ray excitation and fluorescent decay time is given in Table III.

SCINTILLATION COUNTER PERFORMANCE

A. Energy Spectrometry

As was pointed out earlier, one of the very impor-

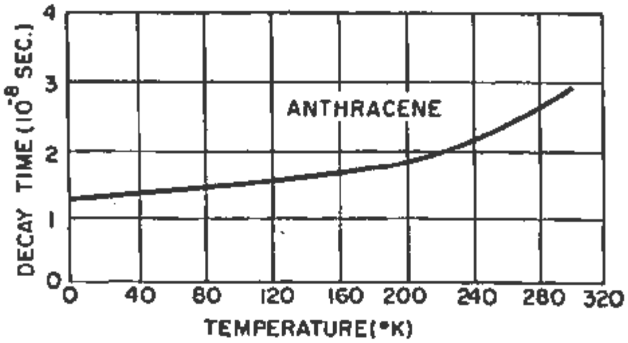


Figure 9. Temperature variation of decay time

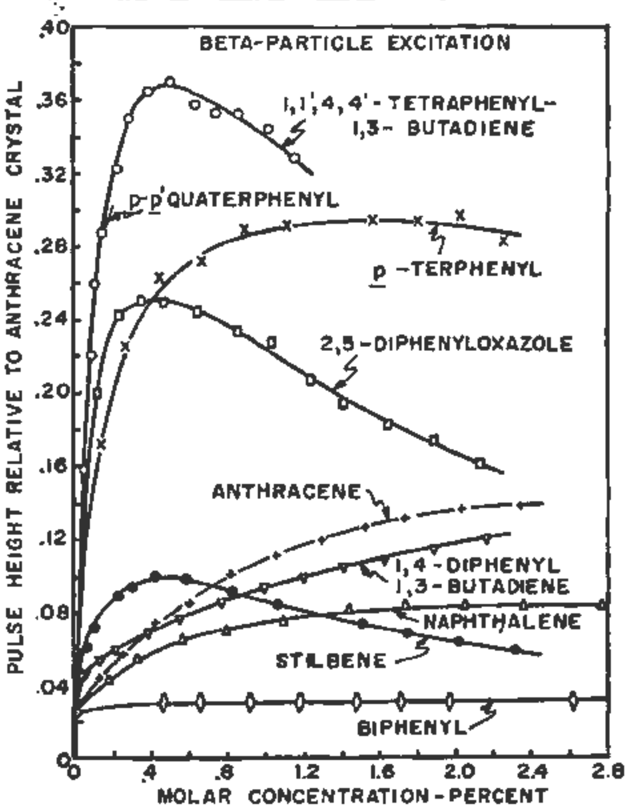


Figure 10. Response of various solutes in polystyrene

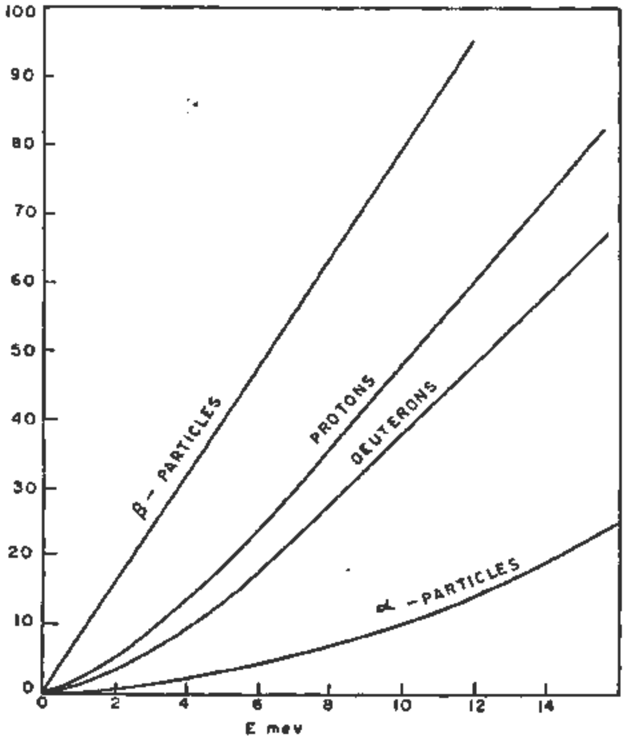


Figure 11. Response of anthracene for various particles

tant properties of the scintillation counter is that it can give an indication of the energy of the incident particle. As discussed in the previous section, the number of photons emitted by the phosphor is, for beta and gamma rays (complete absorption), proportional to the energy of the particle. For heavier particles, protons, deuterons and alpha particles, the amount of light is a monotonic function of the energy. Figure 11 shows the behavior of a scintillator for a number of heavy particles.[28] Since the number of photoelectrons emitted from the cathode of the photomultiplier is proportional to the incident light, the pulse height is a measure of the energy of the incident particle.

Both the emission of photons and the emission of photoelectrons are purely random phenomena. Consequently, if the expected number of photoelectrons from the cathode is n, the root-mean-square deviation in number from the expectation will be equal to $\sqrt{n}$. The photoelectrons from the cathode are multiplied by a secondary emission process which is in itself statistical. This tends to increase further the root-mean-square deviation in charge or pulse height at the output. A statistical analysis of the process involved shows that the percentage half-width of the pulse height distribution of the pulses generated when the expected number of photons generated by the scintillator is N_0 is given by the relation:

$$W_{1/2} = 236\left[\frac{1}{N_0 \epsilon \gamma \eta_p}\left(1 + \frac{\delta^2}{\sigma}\left(\frac{1}{\sigma - 1}\right)\right)\right]^{1/2} \quad (2)$$

where ϵ is the optical coupling between the scintillator and photocathode, γ the quantum efficiency of the cathode, η_p the fraction of photoelectrons collected by the multiplier structure, σ the secondary emission ratio per dynode, and δ the root-mean-square deviation in number of secondary electrons when the expectation is σ.

δ can be closely approximated by $\sqrt{\epsilon\sigma}$. If secondary emission followed the Poisson Law for distribution, ϵ would have the value unity. Experimentally, from measurements of the pulse height distribution

TABLE III. Scintillator Characteristics

Scintillator	*Density*	*Index of refraction*	*Spectral maximum, Å*	*Time const, mμsec*	*Efficiency*
Sodium iodide (NaI:Tl)	3.67	1.7745	4100	250	100
Cadmium tungstate	7.90	2.2–2.3	5200	10^3	100
Anthracene	1.25	1.59	4400	36	48
Trans-stilbene	1.16	1.622	4100	6	28
Xylene + terphenyl + diphenylhexatriene	0.86	1.500	4500	5	23
Xylene + terphenyl	0.86	1.500	4000	3	16
Polyvinyltoluene + terphenyl + diphenylstilbene	—	—	4400	3	23
Polystyrene + tetraphenylbutadiene	1.06	1.595	4000	5	17

of single electrons from the photocathode of a variety of multipliers, ϵ has been found to have an approximate value of 1.5. Substituting in Equation 2, the expression for the percentage half-width becomes:

$$W_{1/2} = 236\left[\frac{1}{N_0\,\epsilon\,\gamma\,\eta_p}\left(1 + \frac{\epsilon}{\sigma - 1}\right)\right]^{1/2} \qquad (3)$$

Within the accuracy of the knowledge of the various constants involved, this expression can be verified experimentally.[29, 30, 31] There are also a number of common non-fundamental factors which can decrease the pulse height resolution capabilities of a scintillation counter. Among these are optical imperfection of the crystal, non-uniformity of the photocathode, instability in the photomultiplier, excessive noise in the photomultiplier, etc.

Spectrometric measurements where large scintillations are involved, giving an energy resolution of a per cent or less, place the severest demands upon the system. Up to the present not very many measurements of this type have been made. To obtain 1% resolution, the scintillation must be constituted of at least 10^4 photoelectrons, which, in turn, means that the particle energy must be of the order of 10 Mev. The extreme penetration of gamma and beta rays of this energy would require scintillators which are impractically large from the standpoint of coupling them to photomultipliers. Some measurements in this range have, however, been made with heavy particles at Brookhaven and Princeton and reveal some interesting instabilities in the photomultipliers. Certain multipliers employing silver magnesium dynodes have shown a tendency to have a higher gain as more charge passes through them. In other words, the pulse height depends upon the counting rate. This is the reverse of the effect which has been observed for multipliers working in a much higher current range where a decrease in gain is found. The recovery of the multiplier from the first named effect is almost immediate as the pulse counting rate is reduced. The fatigue effect, on the other hand, persists for a long time. This effect of increasing gain has only been observed in multipliers with silver magnesium dynodes and not in all multipliers of this type. It has not been observed where the dynodes are activated with cesium antimonide. Where the effect has been observed in a given multiplier, it is quite reproducible, and pulse height measurements can be corrected for it.

In order to test the performance of photomultipliers under conditions of large scintillations and extremely narrow pulse height distributions, K. W. Robinson[32] carried out a series of measurements with an artificial scintillator proving that fractional deviations of 0.1 or 0.2% could be obtained if care was taken to eliminate all spurious effects. This established unambiguously that fundamentally a multiplier phototube is a high resolution device.

Probably the largest number of spectrometric measurements have been made in the energy range from 0.1 Mev to 1½ Mev. Here pulse height resolutions from 5 to 20% are obtained. A great deal of very accurate experimental work has been done in this spectral range by R. Hofstadter at Princeton and Stanford, P. R. Bell at Oak Ridge, H. W. Koch at the National Bureau of Standards and many others. Where the scintillator is sodium iodide, quite accurate measurements can be made of gamma-ray line spectra using the pulse height distribution peak obtained from the photoelectric conversion. Here it is only necessary to have a crystal large enough to absorb the energy of the photoelectrons. However, where it is desired to obtain the energy of the total gamma-ray yield, it is necessary to use a total absorption spectrometer, namely, a spectrometer with a scintillator large enough to absorb all the energy of the gamma rays. A sodium iodide crystal, 4 inches in diameter and 4 inches long with a well in its center, gives essentially total absorption of gamma rays in this energy range. Figure 12 illustrates the arrangement of crystal and phototube of such a spectrometer as used by P. R. Bell.[33] While the majority of the work in scintillation spectrometry has been concerned with line spectra, the method is applicable to continuous gamma-ray distribution.[34]

A good deal of the instrumentation for spectrometry in this range has concerned itself with circuitry and presentation problems, i.e., amplifiers which will respond without blocking or distortion to the wide range of pulse heights involved, multi-channel pulse height selectors and selectors where efforts have been made to obtain maximum channel width and position stability. Improvement in photomultiplier cathodes both in respect to sensitivity and spectral response has been important in improving resolution in this range. Multipliers with large cathodes which avoid the necessity of paralleling a large number of small multipliers in total absorption spectrometers have contributed to the reliability and accuracy of measurements in this range.

Energy measurements in the extremely low energy range are primarily used as a means of separating the gamma-ray photon count from spurious counts due to background thermionic emission, etc. Here the percentage half-width of the pulse height distribution curve is very large so that the device is not suited for accurate energy measurements. How-

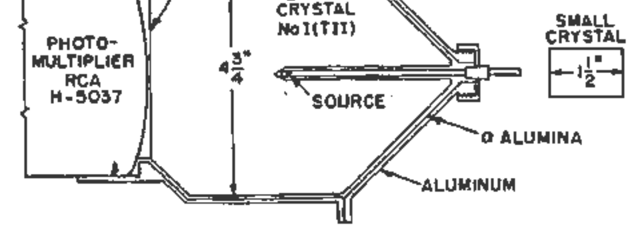


Figure 12. Total-absorption spectrometer

ever, the ability to discriminate to the extent possible in this range has been very valuable when the instrument is used for carbon counting such as is involved in carbon dating, organic radio-chemistry, radiobiology and similar research problems.[10] It is also useful for tritium counting.

The application of the scintillation counter to the detection of X-rays represents the lowest energy region where the device is used. Here the energy discriminating capabilities are of not much value since in the main only one or two photoelectrons are released by each X-ray photon.

B. Time Resolution

The speed and time resolution capabilities of the scintillation counter are important in many ways. First, they make possible the precise measurement of very short time intervals required in nuclear and high energy particle physics. Second, they permit measurements at the very high counting rates that are frequently encountered in the bursts produced by high energy accelerators. Third, they provide means for accurately triggering other equipment and otherwise discriminating between wanted and unwanted events with the aid of gating circuits, high speed coincidence circuitry and other selection means.

The time resolution of a scintillation counter is determined by the fluorescent decay time of the scintillator and the time spread in the passage of electrons through the multiplier.[35, 36] The time constant of a number of phosphors was given in the preceding section where it was pointed out that the organic scintillators were most suited for time measurements because of their short decay times. Three principal factors must be considered in connection with multiplier time spread, namely, secondary emission time spread, differences in trajectory lengths for electrons between two successive dynodes and the effect of electron initial velocities. It has been shown by Greenblatt at RCA Laboratories and Ernst at the University of Illinois that the time spread in secondary emission is entirely negligible in comparison with the time spread in present multiplier tubes. The time spread due to the difference in path lengths between two adjacent dynodes (100 volts per stage) has a root-mean-square value of the order of 0.5 mμ second and is only slightly increased by the effect of initial velocities.

Assuming an exponential decay constant α for the scintillation, and an interstage time spread β for the multiplier, the root-mean-square deviation in time measurement with a scintillation counter is found to be:

$$\langle t\rangle_{rms} = \sqrt{\frac{\alpha\beta}{N_e} A} \qquad (4)$$

when the timing circuit triggers on the output pulse rise at a level corresponding to n_{opt} electrons from the cathode where

$$n_{opt} = N_e B\ \beta/\alpha \qquad (5)$$

N_e is the total number of photoelectrons released by the scintillation, and A and B are constants related to the statistics of the multiplier and have a value of the order of unity.

The estimated rms time errors for scintillation counters using three different scintillators are given below. β is assumed to be 0.5 mμsec.

	N_e	α (mμsec)	n_{opt}	$\langle t\rangle_{rms}$ (sec)
Liquid	100	3	17	2.2×10^{-10}
Trans-stilbene	300	6	25	1.7×10^{-10}
Anthracene	500	36	7	3.3×10^{-10}

It is interesting to note that, in spite of the greater time constant for anthracene, the actual time error does not differ greatly from that of the faster stilbene and liquid scintillators. This is because the initial rate of emission of photons, which is the dominant factor in time resolution, is nearly the same for the three scintillators.

The time resolution capabilities have been used extensively in measuring the decay constants of short-lived radioactive isomers, for the determination of the lifetime of mesons and other high energy particles, and, as was mentioned above, the annihilation time of positrons. This is not by any means an exhaustive list of this class of application. The majority of these time measurements are based upon the delayed coincidence of pulses occurring at the start and finish of the interval being measured. A schematic diagram of a typical delayed coincidence circuit is shown in Fig. 13. The initiating particle is received in detector 1 and the pulse from it traverses a fixed delay line to one arm of the coincidence element. The terminating pulse from detector 2 passes a shorter variable delay line to the second arm of the coincidence element. The difference in these two time intervals when coincidence is obtained is the wanted time interval. Where the problem requirements make it necessary, more complicated timing circuits have been worked out. The chronotron, proposed by Neddermeyer in 1949, and improved later by Keuffel and others for cosmic ray studies, is an example.[37] The chronotron, illustrated in Fig. 14, uses a tapped transmission line as the timing element. A crystal diode at each tap serves as a coincidence detector. Delay lines of increasing length connecting each tap with an oscilloscope make it possible to display the coincidence information in its relative sequence.

It is interesting to consider the possible improvement in time resolution of the scintillation counter which can be expected.

A major decrease in the time constant of conventional scintillators is difficult to foresee. However, it is certain that improvements will be made in the efficiency of scintillators, in optical coupling and in spectral match which will increase the number of photoelectrons released.

Work is in progress at present on gaseous scintillators. These are of interest from the standpoint of speed because of the very rapid rise of pulses which

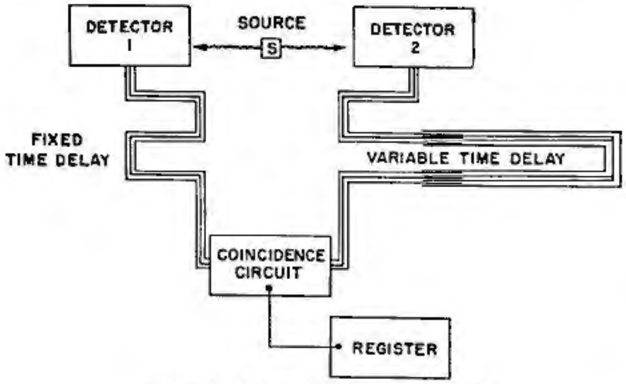


Figure 13. Delayed coincidence circuit

can be achieved. It is too soon, however, to be able to determine the improvement possible in this direction.

Cerenkov scintillators may give a major decrease in scintillation time. The constancy of angle of emitted light from this type of scintillator should make it possible to design an optical system with essentially isochronous light paths which would result in an extremely short duration flash.

There is considerable promise of improving the time resolution of the multiplier phototube and work is in progress toward this end. Multipliers with improved structures where the transit time spread is reduced because of better dynode design and electrode arrangements which give greater field strengths without increasing the inter-dynode voltages are in the research stage. An order of magnitude improvement should be possible through these means.

C. Scintillation Counter Sensitivity

The high sensitivity of the scintillation counter makes it valuable for a great many applications both in research and technology. The factors which must be considered in connection with this aspect are: (1) the high useful absorption of energy from nuclear particles which can be obtained with scintillators; (2) the efficient production of photoelectrons by the scintillator-photocathode combination (i.e., approximately one electron per 500 electron volts of energy of particle); and (3) the low spurious signal in the system. Recent work toward improving sensitivity has concerned itself with all of these factors.

Considerable effort has gone into the problem of developing techniques for growing very large sodium iodide crystals and for producing very large volume organic solution scintillators.[38] Since no scintillator is completely transparent to its own fluorescent radiation, it is not possible even with an ideal optical integrating sphere, to concentrate all of the light from a large scintillator onto a small cathode. Consequently, the development of large scintillators has been paralleled by the development of multiplier phototubes with very large cathodes. Mention has already been made of tubes with 5, 9 and 16-inch cathode diameters. The possibility of even larger multipliers is being explored.

The efficiency of the phosphor-photocathode combination is of equal importance to absorbing capabilities of the scintillator. Some of the developmental work along these lines has been discussed in preceding sections. Improved cathode processing, better optical coupling and more efficient organic phosphors have led to considerable improvement.

Where the high sensitivity of the device is used to measure the low energy emission from C^{14} and H^3 labeled compounds, interesting developments have been made in the direction of dissolving the samples under investigation in the scintillation solution. For example, counting efficiencies of 70% for cholesterol (C^{14}) in a xylene-terphenyl-diphenylhexatriene scintillator have been obtained, and better than 30% for stearic acid (H^3) in the same scintillator.[39]

High sensitivity measurements are only possible when the background counting rate is low. The problem of reducing radiation background by suitable shielding and geometry is outside the scope of the present discussion. However, the spurious signal generated by the multiplier is of equal importance. This signal is the result of several causes. Nonfundamental effects due to residual gas, cold discharge and fluctuating leakage in the phototube have been largely overcome within the last two or three years as a result of better design, construction, and activation procedures. Cesium ions contribute noise when tubes are operated at high temperature. Also, since the Cs_3Sb photocathode has a relatively low work function, it is a good thermionic emitter even at room temperature. The spurious count from these last mentioned sources can be greatly reduced by cooling the multiplier. Much of the very high sensitivity work has been done with tubes operated at a low temperature. The new cathodes mentioned earlier which have a shorter wave length cutoff and contain

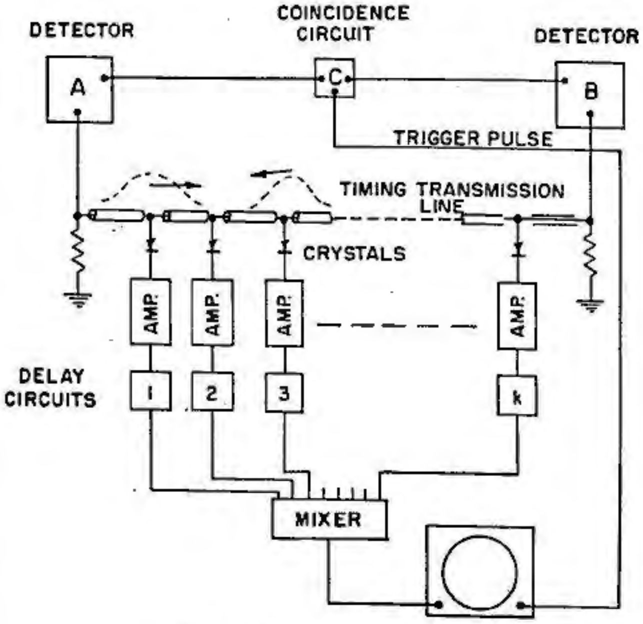


Figure 14. Chronotron timing circuit

no cesium may be very valuable when maximum sensitivity is desired.

The developments of the past two or three years in the scintillation counter field have materially advanced the usefulness of the device. It can be said with certainty that the rate of improvement will continue at the present level so that a prediction of continually increasing usefulness of the device in the future is not unduly optimistic.

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Methods of Pulse Analysis

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The primary objective of this paper is to describe the equipment most widely used for pulse analysis at the present time and to indicate present trends in this field. Background information is included in an attempt to make the paper intelligible to those who have had no previous contact with the problems involved, but much in the way of historical development has been omitted. Some equipment still in use is not mentioned, because it represents techniques which have been abandoned. An excellent survey of the amplitude analyzer field prior to mid-1952 is given by Van Rennes.[1]

NATURE OF THE DATA

Energy proportional response is an outstanding characteristic of most radiation detectors in use today. An ion chamber, proportional counter or scintillation counter gives for each incident particle an electrical charge at its output whose magnitude is a measure of the energy lost by the particle in the detector. This magnitude is subject at best to statistical fluctuation of the Poisson type. Line widths for mono-energetic radiation are proportional in scintillation counters to the square root of energy, being about 5.5 per cent (full width at half maximum counting rate) at 1 Mev for the best sodium iodide thallium crystals. Ionization chambers and proportional counters are relatively better in their regions of applicability. The charge is collected at the output of these detectors over a period of time after the arrival of an emanation. This time may be as short as several millimicroseconds in a fast phosphor scintillation detector or as long as tens of microseconds in an ionization chamber.

Certain features of this data may be of special interest depending on the nature of a given experiment. Usually the relative frequency of pulses per unit energy interval as a function of energy is required. In some instances it is desired merely to know the number of peaks in a spectrum and roughly their relative magnitudes, but far more often it is required that the measurement be carried to the limit set by counting statistics and the state of the art. The detailed shape of a spectrum over a pulse frequency range of as great as a thousand to one yields valuable information leading for example, with other information, to a knowledge of the decay scheme of an isotope, with branching ratios and conversion coefficients, to the nature and amount of a contaminant, or to the type of a beta interaction.

The time relationship between radiations from a given source or the time of arrival of a radiation at a detector measured from some reference time often is of significance. Periods dealt with lie anywhere in the range of from tenths of one millimicrosecond to tens of seconds. This information leads to such answers as the lifetime of a meta-stable nuclear state, the decay scheme of an isotope, or the speed of a particle.

PULSE AMPLITUDE ANALYSIS

A practical limit is set on the precision with which a spectrum can be measured by the speed of available pulse analyzing equipment, the resolving power of available detectors and the achievable over-all stability of a detector and analyzer. Most methods of amplitude analysis divide a voltage spectrum from an amplifier into equal increments and record the number of pulses arriving in each increment or channel during some fixed time. The analysis may be done one channel at a time, or a number of channels may be counted concurrently. Different types of experiments require different degrees of fineness of the increments. Seldom, however, is less than 60 channels sufficient or more than 200 to 300 channels required. Where counting speed is not limited by the nature of an experiment, a basic limitation is the capability of present analyzing techniques for deciding into which channel the peak of a pulse has fallen. Cost and complexity rise sharply when the time allowed per pulse is decreased below about two microseconds. Where, on the other hand, the pulses to be analyzed are produced at such a low rate that the speed capability of a fast analyzer is not needed, schemes of analysis may be used which are considerably cheaper.

The present state of the art seldom warrants statistical accuracy much better than one per cent, corresponding to the accumulation of 10^4 counts in a channel. Because of the wide range of counting rates of interest in some individual spectra, it is highly desirable that the storage capacity of each channel be of the order of at least 10^6 counts. Percentage-wise fluctuations of the counting rate in low counting rate regions often are of as much importance as in high counting rate regions. Therefore, digital storage usually is employed. Analog methods either are of logarithmic response or require an extremely good

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per cent-of-full-scale accuracy. Individual channel acquisition speed becomes a limitation in the fastest analyzers at a rate depending on the shape of the voltage spectrum. With scintillation counters and energies below several Mev a capability of about 1000 counts per second is enough to insure that the channels will not limit the total usable counting rate.

To satisfy the experimentalist the stability of channel width and position should be of the order of one per cent over periods of several days. Also adjacent channels should neither overlap nor fail to join. If all pulses are counted but counted only once, channel width errors cancel when the counts in a number of channels are added to find the counts in a peak.

One way in which the spectrum is divided into increments is by the use of individual voltage discriminators, one for each channel boundary. Analyzers developed at Chalk River,[2] Oak Ridge[3, 4] and Los Alamos[5] are of this type. A large number of such units are in use. One Oak Ridge type, the MC-3, is available commercially from Atomic Instrument Company in Boston or Detectolab in Chicago. Figure 1 is a view of this instrument. It divides a spectrum nominally into 120 channels and records 20 concurrently.

Analyzers of this type require that the discriminators give no output to the storage element until after the peak of a pulse. Then only the highest boundary discriminator to be fired is caused to store a signal. This result is achieved in various ways. The MC-3 holds the pulse at its peak value by means of a pulse lengthener and generates an interrogation pulse a fixed time after the beginning of a signal. Output from all but the uppermost discriminator fired is suppressed by a simple anticoincidence arrangement. Interrogation and lengthening of the pulse is permitted only if the pulse in the analyzer is acceptable from the point of view of adequate separation from other pulses and the condition of the gate circuit used in coincidence work. The Los Alamos analyzer's discriminators return to their "off" condition a fixed time after they are triggered "on." This return produces an output to storage if the discriminator just above has not recently been in the "on" state. The latest type Oak Ridge analyzers, the MC-4 and MC-5 interrogate a fixed time after the end of a signal provided that the interval surrounding the signal is free of other pulses, and that the pulse is not longer than an adjustable maximum value, which would indicate pile-up. An adjustable but otherwise constant dead time is provided to permit accurate counting loss correction.

The Oak Ridge MC-3 requires one microsecond signal pulses and requires 2.4 microseconds for analysis. It may be adapted for longer signals but in any event the time uncertainty between the start of a signal and its peak must not be more than 1.6 microseconds. The Los Alamos analyzer will analyze properly any pulse whose rise and fall times lie within certain limits, but it has memory effects which tend to bias the result at high counting rates. The MC-4 and MC-5 are not critical as to pulse duration and do not have a counting rate limitation in the sorting process.

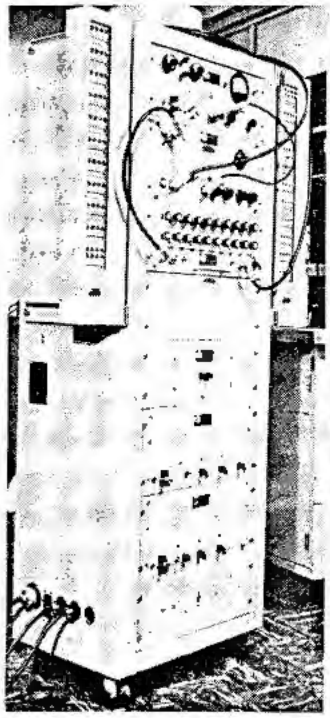

Figure 1. Multichannel analyzer type MC-3. One-hundred-twenty channels

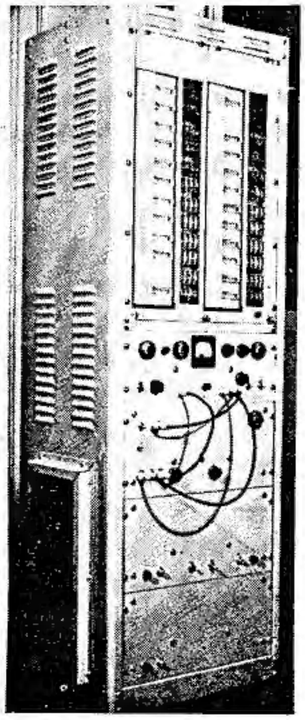

Figure 2. Multichannel analyzer type MC-4. Low cost 120 × 20 channel analyzer, 20 at a time

Figure 2 shows the MC-4. It records 20 channels at a time from a 120 or 240 channel spectrum. The MC-5 uses the same basic circuitry but covers 120 channels simultaneously. Minimum dead time is 1.6 microseconds plus the width of the signal pulses.

Slower analyzers may use a sorting scheme which involves only one discrimination level. D. H. Wilkinson[6] developed apparently the first analyzer to convert pulse amplitude to time. The interval required for a linearly rising discrimination level to reach the lengthened signal's amplitude is measured by counting pulses from an electronic clock. The answer is used to produce an output pulse to the proper register by means of a matrix. More recently Hutchinson and Scarrott[7] combined Wilkinson's principle with a circulating delay line storage. A linearly changing discrimination level is synchronized with the period of the line. Channel storage is in coded form and is regenerated serially. A discriminator pulse adds "one" to the next channel count to be regenerated. This type analyzer has been reproduced with modifications at Oak Ridge[8] and Los Alamos.[9] It seems to be the best answer at present to the sorting problem where only low counting rates need be handled.

Another class of analyzer using only one discriminator has been introduced by Porter and Borkowski.[10] They use a cathode ray tube which has as its target a metal ladder with equal bars and spaces.

The signal sweeps the electron beam across the ladder a distance depending on signal amplitude, and the number of pulses produced adds a count to the proper channel through the use of two Burroughs beam switching tubes and a coincident arrangement.

Bell has proposed that Porter's and Borkowski's tube can be replaced by an ordinary cathode ray tube using an optical projection system and a ladder which is alternately reflecting and transmitting. Two photomultipliers are connected in opposition. One is illuminated when the beam image is on a space while the other receives the reflected light when the beam image is on a bar.

Photographic techniques have been applied by many to the problem of pulse amplitude analysis. One method is to photograph pulses as they appear on a cathode ray tube using a moving film to produce a spacing between pulses. No horizontal sweep need be used on the oscilloscope and the crests of the pulses, preferably lengthened, may be intensified. Automatic readers have been devised.[11, 12] Another method, used by Campbell,[13] involves the use of a fixed film camera viewing pulses on an oscilloscope with triggered sweep. Lens aperture and trace intensity usually are adjusted so that a number of traces of a given height are required to produce appreciable exposure. A densitometer can be arranged to give a plot of the logarithm of counting rate density. This method is plagued by the fact that the degree of exposure is a function of pulse rate as well as pulse integral and the difficulty of maintaining a constant relationship between degree of exposure and film density.

The gray-wedge analyzer introduced by D. Maeder[14] and carried to a considerable state of refinement by Bernstein, Chase and Schardt[15] is a modification of the above method. Lengthened pulses are displayed, say horizontally with a vertical sweep of such a magnitude that the display is a number of vertical straight lines. A gray wedge is placed between tube and camera with the darkest region at the top. The film is developed and printed with the greatest possible contrast, with the result that a sharp boundary of constant exposure traces the spectrum. Since the wedge's density is a logarithmic function of vertical position, the answer is in the form of a semi-log plot. This method gives multichannel information at low cost but its relative accuracy is limited to about 5% and answers of even this accuracy must await darkroom processing. Absolute answers, often important, cannot be obtained. Polaroid cameras are used for qualitative work.

Pulse lengtheners are an important part of most of the analyzers described. Unlengthened pulses from most detectors do not remain essentially at their crest value long enough to be sensed by inexpensive discriminators, and too, most schemes require that the crest value be remembered until storage is completed. Successful pulse lengtheners have been developed by Chase[15] for the gray-wedge analyzer and at Oak Ridge for the MC-3[3] and a considerably improved type for the MC-4 and MC-5.[4] This latter lengthener is of a quality that window amplification, used in most analyzers to increase volts per channel for stability, may follow it. This arrangement provides the window amplifier with a slow signal, considerably easing the problem of its design. The window amplifiers in the MC-5 have a gain of 6 and an output capability of 160 volts. Three of them drive 120 channels each 4 volts wide.

The greatest problem from the point of view of cost and complexity in the process of pulse height analysis is that of data storage and readout. Individual scalers and registers in spite of their obvious disadvantage have proven to be the only satisfactory answer to date for the fast analyzers. In these machines effort has been concentrated on reducing cost and increasing reliability. Some workers in the field advocate the use of cold cathode ring counters. Stoddart has developed a simple but reliable circuit for the use of such tubes.[16] Borkowski's group is using them at Oak Ridge. Bell's group, principally P. R. Bell and C. C. Harris, has developed a very compact scaler using subminiature filamentary tubes each stage of which is boxed in a molded plastic case along with its interpolation light. A scale of 16 of this type as used for each channel on the MC-4 and MC-5 occupies 1⅝ inches × 2½ inches of panel area, is 2 inches deep and consumes about one watt. Due to the extremely long life of this class of tube the reliability is excellent. An improved register, silenced and with a speed capability of about 60 counts per second, has been made available by Veeder Root under the direction of P. R. Bell and the author. A scale of 16 plus register is a particularly good combination because with the fast register, channel count rate is not a limitation, yet the statistical significance of the counts left in the scaler, when rounded off by clearing, is negligible except in cases where of the order of 20 registers or less have been accumulated.

It is in the method of storage that the slower analyzers gain their advantage. They spend time and non-multiplicate equipment to buy storage capability. The Hutchinson-Scarrott type with delay line storage requires moderately complex equipment for adding and regenerating, but there is only one set of such equipment per analyzer. The storage is approximately one thousand pulses per millisecond of line which may be divided among as many channels as desired. Readout is accomplished through the use of a cathode ray tube with horizontal sweep synchronized with channel boundary. The coded information in the line is applied to the cathode ray tube's control grid producing a vertical column of dots and spaces corresponding to scaler interpolation lights for each channel. This display may be photographed. Hutchinson and Scarrott used a mercury delay line. Both of the more recent versions, at Oak Ridge and Los Alamos, use fused quartz lines.

The Hutchinson-Scarrott scheme suffers from the disadvantage that since access to any one channel occurs only once per line period, on the average the processing time per pulse is half the line period. Other methods have appeared recently using computer techniques in which pulse height information is translated into an address which may extract immediately the count in the proper channel. A "one" then may be added and the count restored. This process in principle may be quite rapid. A 100 channel analyzer of this type using magnetic core storage has been developed at Los Alamos,[17] and is available from Pacific Electro-Nuclear Company, Hollywood, California. Amplitude to time conversion is used. One hundred clock pulses, one every 4 microseconds, correspond to maximum pulse amplitude. They are applied to a scale of 100 decimal scaler. The 10's unit and the 1's unit each select a plane on the 10 × 10 face of a 10 × 10 × 20 matrix of cores. The 20 cores along each intersection store channel count in decimal form. When a channel is selected, it is read out into a scaler, one is added and the information restored. Dead time is dependent on pulse amplitude, being 4 microseconds per channel plus 15 microseconds for readout and restorage. An automatic reader is provided which prints on a paper tape. Readout of all channels requires about 2 minutes.

Electrostatic storage, another computer technique, may be used. It is basically less expensive than magnetic cores both intrinsically and from the point of view of driving equipment. Storage is in the form of a charge pattern on the face of a cathode ray tube. A horizontal stripe may correspond to each channel. When a beam is swept across the region pulses are produced corresponding to the channel count in coded form. A beam suitably intensity modulated restores the new information. With this scheme speed is limited by the time required to read a count out serially into a scaler, approximately 7 microseconds per bit. If separate tubes are used for each bit, readout may be in parallel in which case readout-restore speed is comparable to magnetic core storage. Over-all speed can be greater because less time is required to obtain an address.

The need for extreme speed in an analyzer is not immediately obvious. It is a convenience but not a necessity to be able to measure the spectrum of some relatively long-lived isotope at the rate of 2×10^4 integral counts per second. It becomes a necessity, however, for example in fast rabbit experiments where a source which was being irradiated a moment before is studied for short-lived nuclear states. It is necessary also in many coincidence experiments where very high singles rates are required to produce a usable coincidence rate. There does exist for this type of work the possibility where window amplifiers are used of gating out the unwanted signals before they enter an analyzer. Since the window amplifier normally is biased "off," it may be held "off" except when a signal is to be passed, without producing a pedestal. Workers using particle accelerators often require a high speed analyzer since instability of the machine often makes it necessary to get a full set of data for a given experiment as quickly as possible. Pulsed accelerators present an even greater need for speed. Instantaneous counting rate must be very high during a pulse to result in even a moderate average rate. Cunningham[18] uses electrostatic storage to permit analysis of pulses relatively at leisure during the machine's "off" time. Pulses are displayed vertically on an oscilloscope with no horizontal sweep. The peak of each pulse leaves a charge discontinuity on the phosphor. When a readout beam is swept up linearly along the same path, a signal is produced in a capacitively coupled pickoff electrode every time the beam passes the peak of a stored pulse.

PULSE TIME ANALYSIS

Many of the analyzers just described convert pulse amplitude to time. The reverse of this process permits existing analyzers to be used to make a spectrum of the frequency of occurrence of pulses as a function of their delay from some event. One method was developed by Braid and Detenbeck.[19] It consists of a balanced amplifier whose two inputs are linear sweeps, one initiated by the initial event, the other by the delayed pulse. During a conveniently long succeeding interval the sweeps continue, but the amplifier, sensitive only to this difference, gives a constant output proportional to this difference, and therefore the delay. Another arrangement used at Oak Ridge involves supplying a condenser with a constant current beginning with the first pulse and ending with the second. The signal produced is essentially identical to that from a scintillation counter, and is applied to a conventional linear amplifier. Both of these methods may be used to catalogue delays in the tenth millimicrosecond region.

Any of the amplitude-to-time analyzers may be adapted directly to the time analysis problem. In addition to the schemes they employ, many others have been used. For best results all must satisfy certain conditions with regard to the manner in which the time of an event is determined. In most detectors and especially in scintillation counters, the uncertainty of the time of an event as measured by the time of arrival of the first measurable charge at the detector output is less than by any other characteristic; as for example the time of the peak of the signal obtained by differentiation of the charge waveform. Also since the accumulation of charge is a statistical process the tail of a signal is quite lumpy if it is differentiated with a time constant short compared to the mean time of charge accumulation. These lumps may be mistaken for other signals by a coincidence circuit. Therefore it is essential that the signal be allowed to decay with a time constant somewhat larger than the mean accumulation time, and that it be limited severely so that only the initial rise is used to produce a pulse of proper width for the resolving time desired. Many coincidence circuits have been built which work well when tested with a signal generator but not with actual signals because they do not fulfill the requirements just stated.

An effective method of coincidence measurement is by adding pulses of the type described (with suitable delay) and using a pick-off diode biased to be insensitive to single amplitude pulses. For very high speed work a UHF mixer type crystal must be used because the more common diodes have memory effects and slow response. Type 1N26's are used at Oak Ridge. Resolving time stability is greatest when the singles pulses are rectangular and the pick-off circuit indicates a coincidence with the smallest possible pulse overlap. McGowan[21] at Oak Ridge uses two Hewlett Packard amplifiers following each detector, driving negatively a pair of 404A amplifiers whose plates are paralleled through delay lines and a pulse forming stub. He and R. E. Bell[22] at Chalk River (using a similar circuit) have been able to study isomeric states with half-lives in the region of 2×10^{-10} seconds.

Multichannel time delay analyzers have been used principally with time of flight neutron spectrometers. One at Oak Ridge developed for the fast chopper uses 100 sequentially closed gates driving separate scaler-register circuits. Johnstone, Kiepin, Koontz and Gallagher[20] at Los Alamos recently described one developed there which is suitable for use in the millisecond and second region. Clock pulses control the operation of a scaler and readout mechanism. Each pulse transfers the scaler reading to a magnetic drum or paper tape and clears the scaler. Rankowitz and Graham[23] use the Wilkinson scheme for the Brookhaven neutron chopper. Neiler at Oak Ridge uses the time to amplitude converter described above in conjunction with an MC-3 analyzer for neutron energy measurements in the region from about 8 kev to 17 Mev.

Often it is required that a coincidence be formed with the output of a single channel analyzer. Because this device necessarily cannot give an answer until after the crest of a pulse and because this time is considerably more uncertain than the beginning of the pulse, a rather large resolving time is required to insure the overlap of truly coincident events. Here the fast-slow technique is used to reduce the background of random coincident pulses. A fast coincidence circuit operating on the leading edges of the signals detects every true coincidence regardless of energy. This information suitably delayed is put in slow coincidence with the analyzer output. Most commonly the slow coincidence is used to gate "on" a multichannel analyzer. The effective resolving time is essentially that of the fast coincidence except in very rare cases, if the slow coincidence is made no longer than necessary.

Recently interest has been increasing in methods of performing a multichannel-multichannel coincidence. Campbell proposed a photographic method a number of years ago which gave qualitative information.[24] He deflected a cathode ray beam horizontally with one signal and vertically with another, intensifying during the lengthened crest of each signal. For very slow work Shoemaker at Princeton photographs the registers of two analyzers, gated from a fast coincidence circuit, every time a pulse is recorded. Faster readout methods will permit this idea to be applied at higher speeds.

CONCLUSION

It is apparent that no one type of equipment is most suited to all applications in the field of amplitude or time analysis. All the types described are expected to continue in existence since each is well suited to certain jobs.

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High Resolution Radiation Detectors in Research

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I. INTRODUCTION

The increased availability of radioactive isotopes has led to a more widespread application of the tracer technique in fundamental scientific research. The value of these tracers, however, depends greatly on our ability to detect them. In tracer experiments, one seeks two types of information: the amount of radioactive material present and its location. Instruments and procedures for determining the quantity of radioactive tracers have been developed to a high degree, and the sensitivity of present instruments leaves little to be desired. The techniques for determining location, however, are relatively poor when we consider the inherent resolvable unit in tracer experiments is the emitting atom with a diameter of about one Ångstrom, or one ten-thousandth of a micron. But present localization techniques are able to provide, under ideal conditions, resolution of the order of only one micron; indicating the amount of improvement possible.

The best available autoradiographic systems depend on the light microscope as the means for examining the film and specimen being studied. With one micron resolution now possible, improvements in present techniques will soon exhaust the resolution range of the light microscope (on the order of 0.2 microns), and new procedures will be required to permit examination of tracer distribution at magnifications attainable with the electron microscope.

Not only resolution, but the physical and chemical properties of the detecting medium can be substantially improved. Present localization techniques utilize photographic film, which responds not only to light and ionizing radiation, but is also quite sensitive to chemical constituents prevalent in many materials studied. For reliable results, it is usually necessary to provide an impermeable protective layer between the film and the specimen. To overcome these basic limitations of photographic film, development of new detecting media which are stable to light and chemical action while remaining sensitive to the high energy radiation of radioactive materials has been undertaken.

Where speed and convenience justify reduction of resolution to about 75 microns, a new direct reading radiation microscope can be used.

2. CRITERIA FOR HIGH RESOLUTION RADIATION DETECTION

The ideal high resolution autoradiographic system would possess the following properties: (*a*) a vanishingly thin film; (*b*) high sensitivity to high energy particles; (*c*) low sensitivity to light protons; (*d*) high chemical stability in the unexposed state; and (*e*) a developable or directly observable image produced by high energy radiation.

The film or detection method which meets all these requirements simultaneously has not yet been perfected. However, existing materials do fulfill some of these criteria and others, as discussed in this article, are being developed which are designed to possess all the listed properties. The approach has not been limited to the use of silver halide emulsions, but includes also monomer-polymer systems which can be made highly sensitive to ionizing radiation and yet remain stable in the presence of ordinary light.

3. RESOLUTION DEFINED

The term "resolution" for an autoradiographic system describes the ability to distinguish two radiation sources which are close together. The distinction is made by measuring the variation in the film density or blackening. As long as two separate blackened areas are distinguishable, the radiation sources are resolved; when the blackening merges into one area, no such distinction can be made and resolution has been lost.

Other workers[1, 2] have chosen to define resolution on a one-point rather than a two-point basis. The criterion used in this case is the distance from the point of maximum grain density in the film to the point where density is half the maximum. The resolution is measured by the diameter of the half maximum density circle surrounding the maximum density point.

It is felt that this definition fails in two important aspects. First, it does not take cognizance of fundamental concept of "resolution." If only one radiation source is present in a field being studied, there is seldom difficulty in correlating it with film blackening. The real problems arise when several adjacent structures must be examined for the absence or

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presence of radioactivity; or the amount present measured individually in each element of a microstructure system. Second, when compared to the "two-point" criterion, the "one-point" system is much too optimistic in its prediction of performance for a given film and geometry.

Calculations of expected resolution, for both criteria, have been made using the following assumptions:

1. The radiation sources are vanishingly small points;
2. The emulsion is essentially continuous; its grain structure can be neglected; and fog may be neglected;
3. For this calculation only, the radiation sources are 0.1 microns from the emulsion;
4. The radiation sources are isotropic and the radiation passes all the way through the emulsion with constant energy loss (and film blackening) per micron of particle travel;
5. Two points are resolved when the film density between the points is half that over the points proper, or
6. The resolution distance for a single point is the diameter of the half maximum density circle, as noted previously.

The resolution obtainable by these standards for progressively decreasing film thickness is shown in Fig. 1. Two characteristics are noteworthy. The minimum resolvable distance is, in general, about 2.5 times greater by the two-point criterion. Also, the resolution improves substantially as the emulsion thickness is reduced to that of the separation layer. When film thickness is less than that of the separation layer, there is diminishing improvement ending with the theoretical resolution for zero film thickness. Improvement by the two-point standard is greater proportionally than that predicted by the one-point analysis. Thus, 0.9 micron resolution predicted for thick emulsions by the two-point system can be improved to 0.45 micron resolution by reducing film thickness to 0.1 microns.

As will be described later, studies are now proceeding on the making of ultra-thin radiation-sensitive films. It should be noted that the improvement predicted here is in contradiction to the concept previously developed[2] that, for a thin separating layer, film thickness is of minor significance in achieving high resolution. This conclusion was based on calculations in which the emulsion thickness was always much greater than that of the separation layer. From Fig. 1, it may be seen that for a 0.1 micron separation layer, reduction of film thickness well below 5 microns is necessary to achieve optimum resolution.

In general, the requirements for high resolution on the geometry of the autoradiographic system are:

1. Intimate contact between the film and the radiation source. This is the single most important factor; although to prevent chemical reactions, protective layers must often be used.

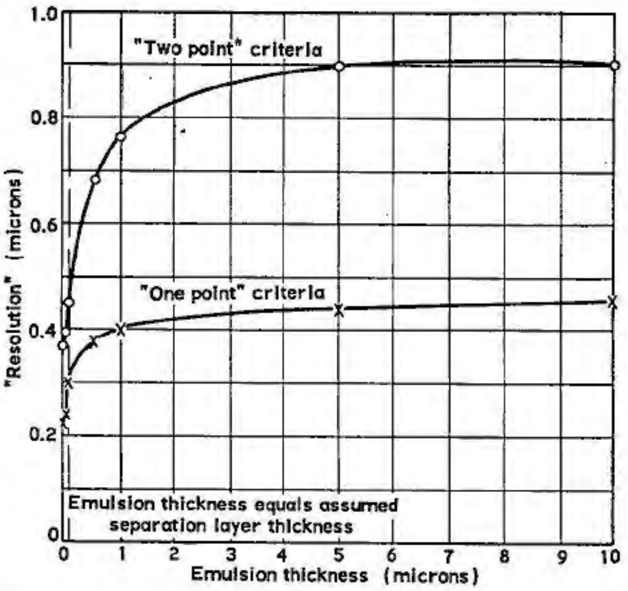

Figure 1. Resolution by the one and two point criteria vs emulsion thickness. Thickness of separation layer equals 0.1 microns. Note rapid improvement in resolution as emulsion thickness is reduced to that of separation layer

2. The specimen should be as thin as possible. This can often be achieved by sectioning, particularly soft materials such as tissue. However, source thickness can be a serious problem in metallurgical and other solid state work.
3. The film should, as shown, be as thin as possible. This point is often neglected or misunderstood.

4. HIGH RESOLUTION METHODS NOW IN USE

A. Stripping Film and Liquid Emulsion

The most satisfactory methods for high resolution autoradiography currently in practice utilize various forms of a stripping film or a liquid emulsion. The stripping film consists of a photographic emulsion about 5 microns thick mounted on a gelatin layer also 5 microns thick and supported on a plastic sheet. The film is stripped from its plastic support, floated on water, and picked up on the specimen.[3]

Liquid emulsions are applied by carefully "painting" a layer of warm emulsion, supplied in bulk form, over the specimen.[4] After suitable exposure periods, the specimens are processed with routine developer and fixative.

In the liquid process, the emulsion thickness can be varied, and, if desired, the specimen can even be imbedded in emulsion. The resulting beta-ray tracks can be correlated with tissue morphology, whereas single background grains can be ignored. Fortunately, background tracks are usually sparse. However, high fog level and high concentration of activity will obscure the specimen detail, and resolution is severely limited by beta particle energy. Furthermore, the irregular path of the electron does not permit easy identification of the point of origin.

Stripping films usually indicate radioactivity by a general increase in grain count over the source, but now individual fog grains must be dealt with. Both techniques, with sufficient practice, are easily handled in a routine manner. We have found the stripping film to be the easier of the two with less likelihood of high fog. Some experimenters have claimed resolution with stripping film of less than 1 micron when using tritium as the label and sections 1 micron thick.[5]

The sensitivity of these commercial emulsions to radioactive particles is quite high, and loading of the silver halide is sufficiently dense to be considered continuous (actually it is only 50% by volume). Grain size varies with the particular emulsion but is of the order of 0.2 to 0.3 microns in the stripping film. However, stripping films are sometimes subject to shifting on the specimen or peeling during development. Since the developed film area must maintain registry with specimen morphology, shifting after exposure could lead to erroneous results. Adhesive agents to prevent shifting have been recommended[3] but, from our experience, are not reliable.

In addition, the high susceptibility of photographic emulsions to chemical artifacts[6] requires the presence of a protective layer between the specimen and the emulsion. An adequate protective coat must be able to prevent diffusion of chemical agents from the specimen over long periods of exposure, maintain tissue integrity, and enable the film to adhere to the slide.

A number of materials suitable as a protective layer have been studied for their ability to meet the criteria noted above. Our results indicate that the best protection is provided by the 90:10 copolymer of vinyl chloride and vinyl acetate (Vinylite VYNS manufactured by the Bakelite Corporation). Two to four per cent solutions of this material in methyl ethyl ketone are used in coating samples. Two per cent solutions of vinylidene chloride (Saran F120, Dow Chemical Corporation) also yield a good protective layer. While these materials are excellent for work with metallographic mounts, difficulty has been encountered in their use with biological tissues. The Saran or VYNS material, on drying, destroys the tissue integrity, yielding a highly distorted specimen. For tissue work, we recommend first applying a layer of polyvinyl alcohol (two per cent in water) followed by a two per cent parlodion in alcohol coating. The parlodion layer has been found to give much better film adhesion than a Saran or VYNS coat, although its "protective" ability is not as good as these latter materials.

B. Wet Collodion Process—Silver Bromide

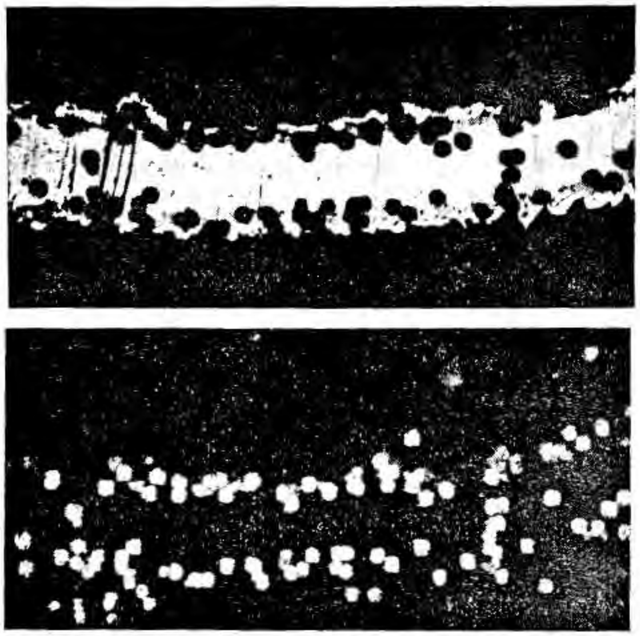

Figure 2. Wet process autoradiograph of nickel-63 specimen using five-micron thick collodion emulsion. Bright illumination, 600×. Polarized illumination, 600×

One of the first high resolution autoradiographic methods developed was the wet collodion process. Details of this technique and results of some of its applications have been published,[7] but a short description of the process and some of its characteristics seem appropriate here. Essentially, the technique utilizes a modification of the wet collodion method of photography. Adequately protected specimens are coated with a metallic bromide solution of collodion and exposed in solutions of silver nitrate. After a suitable exposure period, the specimens are processed using physical development. Figure 2 shows bright and dark field views of a wet process autoradiograph on the edge of a 15-micron platinum foil plated with Nickel-63 and overplated with silver. A dark field view, showing only the autoradiograph silver grains facilitates an interpretation of the pattern unbiased by surface detail in the specimen.

The wet process can produce better than 10-micron resolution.[8] The loading of the emulsion as well as the grain size and uniformity are comparable to that of stripping film emulsions. However, the film is formed on the surface of the specimen in a collodion matrix, and shifting or slipping of the film from the specimen is impossible. Thus the registry here is never in doubt. Furthermore, the background of the wet process film can be controlled to 1000 grains/mm^2 as compared to a measured value of 10,000 grains/mm^2 for commercial emulsions.[9]

The significance of this factor may be realized in considering the quantitative aspect of the autoradiographic process. The most important question in an analysis of an autoradiograph is whether an observed silver grain in a given area is an indication of the passage through the film of an ionizing particle. The probability that two adjacent silver grains is due to radioactivity is much greater in a film which has 10 background grains per 100 μ^2 area than one that has 100 such grains per 100 μ^2. Despite these advantages, routinely reproducible results are difficult to attain with the wet process. Another serious limitation is the short permissible exposure of the specimen in a silver nitrate solution. The maximum exposure time for the process has been found to be on the order of 48 hours when exposed at 2°C. Under these conditions, specific activity of specimens must be relatively high as compared to the stripping film process in which exposures of three months are possible.

C. Wet Collodion Process—Silver Phosphate

A method somewhat similar to that of the wet process has recently been perfected in the laboratories of Dr. Manfried Siess at Tübingen, Germany.[10] In Dr. Siess' procedure, a silver phosphate film is formed in a collodion matrix over a specimen carrying a radioactive tracer. Exposure is in a silver nitrate solution, and processing is carried out by physical development. Under the experimental condition outlined for this technique, the silver phosphate film was shown to be more sensitive to light than the wet process but less sensitive than commercial film. Siess presents examples of autoradiographs of I^{131} labelled thyroid tissues and other radioactive specimens. There is some question in this work as to the possible presence of chemical artifacts in these reactions. Not enough material is available regarding this technique to evaluate it further.

5. NEW TECHNIQUES NOW UNDER DEVELOPMENT

A. Thin Film for Electron Microscope Autoradiography

Recently, much effort has been expended in attempts to prepare an ultra-thin, densely loaded silver halide film which would be suitable for electron microscopy studies. This film would be similar to the collodion screen now used for electron microscope specimens but sensitized to detect radioactive tracer materials. The tagged specimen would be placed on the sensitized screen and exposed for a suitable period of time. It would then be processed and examined in the electron microscope. The scattering of the electron beam by the deposited silver would thus serve to represent an autoradiograph of the specimen. With a film sufficiently loaded with very small, sensitive and uniform grains, resolution approaching that of the electron microscope itself could be obtained.

During the past year, various plastic-solvent systems have been studied for preparing a film with the necessary properties. By carrying the silver nitrate in the plastic-solvent material and the bromide in a water solution, it has been possible to form thin monograin-layer films of silver bromide in a manner quite analogous to forming thin collodion specimen screens for electron microscope use. The most satisfactory films to date are obtained with a solution of silver nitrate in benzyl alcohol mixed with a parlodion-methyl ethyl ketone solution. A drop of this mixture is placed on a cold water solution of potassium bromide. Benzyl alcohol appears quite unique in that silver nitrate can be dissolved to the extent of 15% while remaining completely stable if kept in the dark. Most organic solvents prove to be either unstable to silver nitrate or quite low in their ability to dissolve it. Potassium bromide is only slightly soluble in benzyl alcohol and reversing the materials (i.e. silver nitrate in water and bromide in the solvent) does not produce a densely loaded film.

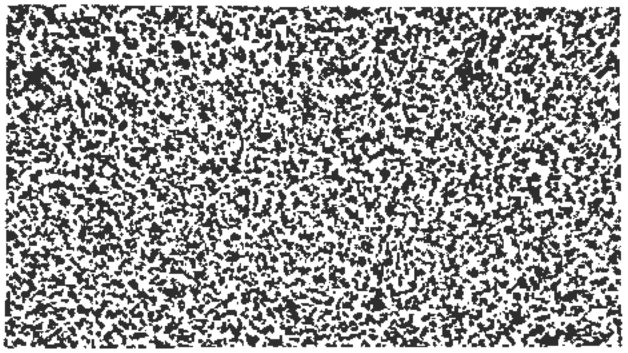

Figure 3. Electron-micrograph of collodion-silver halide film formed over water bath. Film is a monograin layer 0.2 microns thick. (×2000)

In Figs. 3 and 4, electron photomicrographs of the thin film and of autoradiographic stripping film are compared. The thin film has better uniformity of grain distribution than the stripping film. This thin film had 0.1% potassium iodide plus the bromide in the water and 10% ammonium hydroxide in the silver nitrate mixture. These films can be processed by routine chemical developers and fixative. Response of the bromide film to light, X-rays, alpha particles from polonium, and C^{14} and Tl^{204} beta particles has been obtained. A measure of the film sensitivity has not yet been undertaken although our studies have indicated that the presence of iodide drastically reduces the sensitivity. Electron-micrographs indicate the need for increased loading and more uniform grain distribution. Experiments designed to perfect this thin film are in progress.

B. Radiation Sensitive Monomer-Polymer Systems

In the search for new procedures of radiation detection, systems have been examined that are stable to light yet respond to the higher energies of radioactive particles. A likely mechanism containing

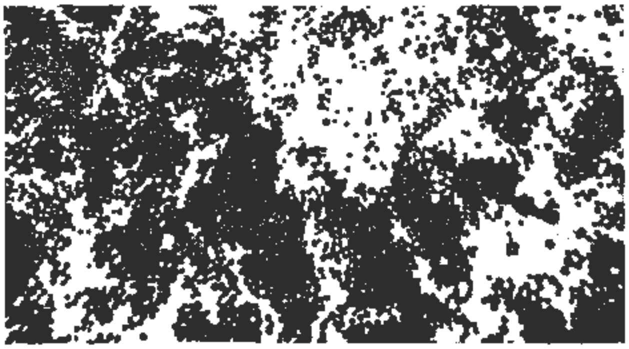

Figure 4. Electron-micrograph of Eastman permeable base stripping film swelled and stretched by water flotation. (×2000)

an inherent amplification factor is that of a polymerization reaction. Several experiments[11, 12, 13, 14] have indicated that certain monomers under special conditions are radiation sensitive while otherwise remaining stable to heat and light. The possibility of using such a system to infuse into a "tagged" specimen and polymerize at the site of radioactivity or to polymerize locally on an irregular solid at the site of radiation suggested itself, and some of these systems have been investigated with this purpose in mind.

On the basis of experimental work on polymerization reactions, it has been demonstrated that the sensitivity of many of these systems to heat, light, and high energy radiation is reduced by the presence of oxygen.[15] It was necessary, therefore, to work under conditions in which oxygen was excluded. Furthermore, a system had to be found that would be sensitive to high energy radiation under conditions in which it was stable to heat and light.

Work by Schmitz and Lawton[16] indicated that the difunctional monomer tetra-ethyleneglycol dimethacrylate (TEGMA) was much more sensitive to radiation-polymerization than monomers such as acrylates, methacrylates, styrene, and acrylonitrile which possess only one functional group. In work with TEGMA, it was found that polymerization of the bulk material under nitrogen proceeded at doses of 7000 r of X-rays. Ethylene dimethacrylate, under the same conditions, polymerized at 30,000 r. However, the stability of both materials was quite poor under these conditions.

The solid monomer materials, methacrylamide and *N-N′* methylene-bis-acrylamide (*N-N′*) have also been examined. An alcohol solution of methacrylamide proved to be quite stable to heat and light under nitrogen and was found to polymerize to an insoluble white gel at an X-ray dose of 4000 r. The *N-N′* monomer in aqueous solution of 0.1M concentration was noted to react at doses of the order of 250 r of X-rays when under deaerated conditions. This material appeared to be quite stable under these conditions.

A more detailed study of the *N-N′* system was undertaken and data was obtained with respect to the extent of polymerization as a function of dose and dose rate. The effect of concentration, temperature, pH, and the presence of various chemical species on the reaction was also investigated. The material was prepared in 15 ml quantities of a 0.1*M* (1.5%) solution under high vacuum conditions. Analyses were made by filtering the polymer through fine-porosity gooch-type crucibles and weighing them. Accuracy is considered good to 15%. Figure 5 indicates the response of this system to total dose. The effect of increased temperature may also be noted (the monomer has been found stable at 65°C over a four-week period). The straight line curve for the room temperature reaction is seen to extrapolate to the zero point indicating no threshold dose exists.

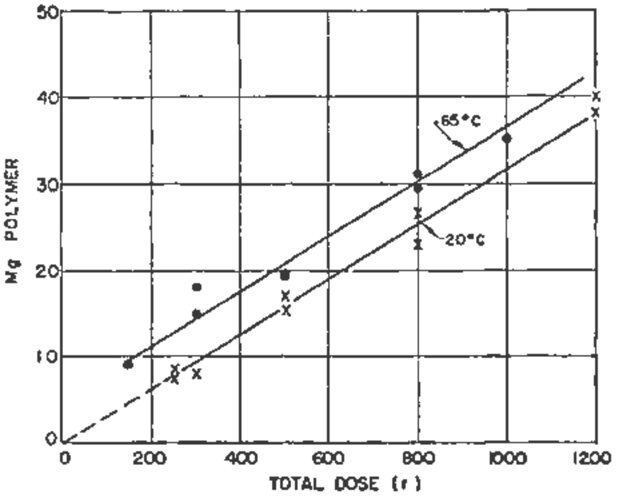

Figure 5. Polymerization response of N-N′ methylene-bis-acrylamide to 200 kvp X-irradiation (dose rate of 100 r/min)

Because of limitations of radiation facilities, data on the dose rate response has not been substantiated, but indications are that the efficiency of the reaction increases with a decrease in dose rate. For a total dose of 500 r, at a rate of 900 r/minute, about 12 mg of material react as compared to some 23 mg at 15 r/minute. Using cobalt-60 irradiation at a dose rate calculated to be 0.5 r/min, an average value of 20 mg was obtained for a total dose of 500 r. Increasing the concentration (the monomer is soluble to 3 gm/100 cm^3 at 20°C) does not substantially increase the yield while tending to make the solution unstable at high temperatures. Attempts to adjust the pH with phosphate-citric acid buffers led to unstable solutions. Placing different metallic ions and potential sensitizing agents in the monomer did not appear to be helpful, and the most stable and sensitive system thus far has consisted of the purified monomer alone in distilled water.

Preliminary studies have been carried out, placing radioactive specimens in the evacuated solutions of *N–N′*. Under conditions where the material is adequately "protected," stability is maintained and response to P^{32} and Tl^{204} has been noted. Calculations of the dose from the activity of the isotopes used indicates that the visible reaction in these cases occurs at dosages comparable to those noted in the X-ray induced response—around 200 r.

Experiments at Brookhaven National Laboratories with radiation-induced polymerization of acrylamide in the solid state indicated that this material might prove more reactive than the *N–N′*.[17] Acrylamide is considerably more water soluble (78 gm/100 cm^3) than the *N–N′*, but it was noted that a 20% solution of acrylamide was quite unstable under high vacuum conditions. However, when nitrogen gas was bubbled through a 20% aqueous solution of acrylamide the X-irradiation induced polymerization proceeded visibly at doses of the order of 100 r. Furthermore, the material appears to be quite stable under these conditions, although further experimentation is needed to confirm these results.

A re-examination of all the systems investigated was carried out under the condition of nitrogen bubbling, and the following table indicates the results:

Monomer	*Response to 200 kvp X-irradiation (no filler, 100 r/min)*
Tetra-ethyleneglycol dimethacrylate (bulk)	No reaction at 500 r
Tetra-ethyleneglycol dimethacrylate (aqueous solution)	No reaction at 500 r
Ethylene dimethacrylate (bulk)	No reaction at 500 r
Ethylene dimethacrylate (aqueous solution)	No reaction at 500 r
Acrylonitrile (bulk)*	No reaction at 500 r
Acrylonitrile (1.0 *M* aqueous soln.)	No reaction at 500 r
Methacrylamide	
(20% soln. water)	No reaction at 500 r
(20% soln. methyl alcohol)	No reaction at 500 r
(20% soln. 0.1 *M* *N-N′* in water)	No reaction at 300 r
N-N′ Methylene-bis-acrylamide (0.1 *M* soln. water)	Turbidity at 300 r
Acrylamide	
(30% soln. water)	71 mg at 150 r
(50% soln. water)	100 mg at 150 r
(30% soln. 0.1 *M* *N-N′* in water)	700 mg at 150 r (dose rate of 50 /r minute)

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* A Tracerlab report[18] indicated that bulk acrylonitrile, under high-vacuum air evacuation, is sensitive to radiation doses as low as 50 r with visible turbidity. Their data show 1.2% polymer formed at 1600 r of cobalt-60 at a dose rate of 80 r/minute.

The stability of the acrylamide systems has been checked for twenty hours of nitrogen bubbling with no polymer formation noted.

From data thus far obtained with these monomer-polymer reactions, some estimates of the amplification factor associated with these materials have been made. A Q factor (defined here as the number of molecular units reacting per initial ionization event) of 2×10^3 has been calculated for the N–N' system. The acrylamide reaction (30% solution 0.1 M N–N') gives a Q value of 2×10^7. These values may be compared to that of photographic film in which it has been estimated that 10^9 to 10^{11} atoms of silver react in the development process per latent image grain.

On the basis of these very recent developments, the likelihood of radiation localization techniques utilizing the polymerization reaction appears quite encouraging. Further experimentation is being carried out in this area.

C. Beta-Ray Microscope

Still another approach to the problem of radioactivity localization has been undertaken. This has included the development of an instrument which makes possible the direct measurement of radioactivity within the field of the optical microscope. Details of this Beta-Ray Microscope—as the instrument is termed—have been reported elsewhere,[19] and only a brief description will be included here. This method utilizes a thin solid scintillator placed immediately over a radioactive specimen as the primary radiation detector. An optical system, which includes a standard microscope objective, collects light from the scintillator. It also enables one to select a particular area of scintillator and source for study. The arrangement of source, scintillator, and optical system is shown in Fig. 6. Anthracene crystals as thin as 75 microns have been used as the scintillator. Satisfactory detection of beta particles from nickel-63, carbon-14, thallium-204 and cobalt-60, with a resolution of 75 microns has been achieved.

Since the amount of light available from the passage of a beta particle through a crystal this thin is small, detection of the light is accomplished with the electron multiplier phototube. The dark noise of the electron multiplier phototube becomes an acute problem when detecting light pulses of this level, and two phototubes have been used in a coincidence arrangement to decrease this background noise. Light gathered by the optical system is split approxi-

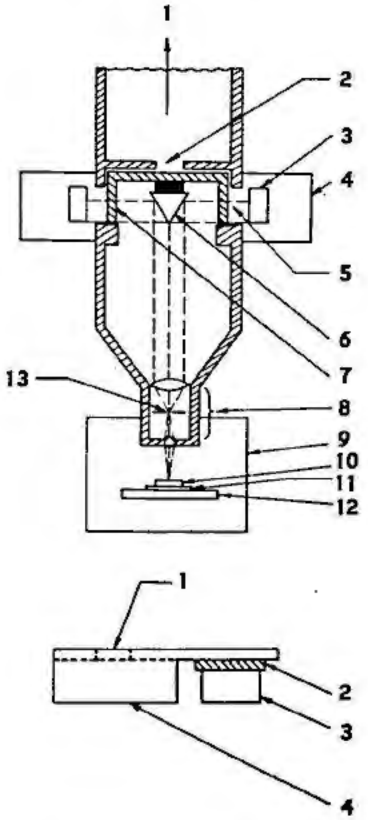


Figure 6. Cutaway view of proposed optical system for Radiation Microscope: (1) to visual viewing optics; (2) aperture for visual viewing—closed when prism is in place; (3) phototube; (4) phototube mount and shield; (5) aperture for phototube—open when prism is in place; (6) prism; (7) sliding light shield and prism mount; (8) compound objective-special; (9) light tight housing for crystal and source; (10) crystal-scintillator; (11) source; (12) microscope stage; and (13) diaphragm to control field size. (Below) Side view of prism mount; (1) visual viewing aperture in slide; (2) prism holder; (3) prism; and (4) sliding shield for phototube

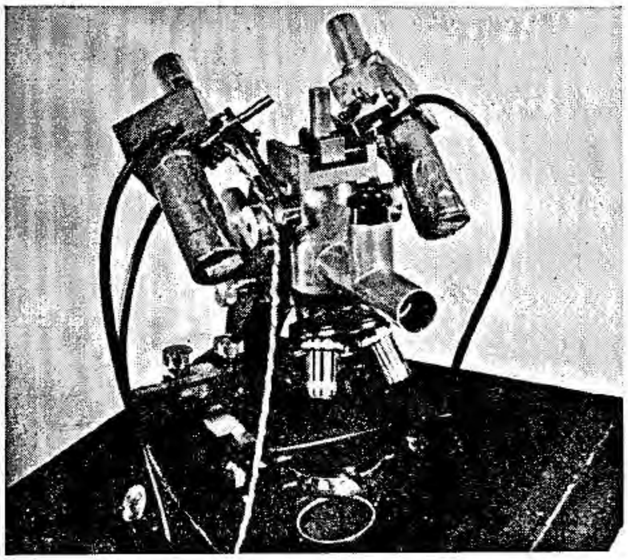


Figure 7. Optical system of beta-ray microscope

mately in half and fed simultaneously to the two phototubes. A front silvered prism properly placed in the optical system serves as a light splitter.

Figure 7 is a picture of a refined model of the optical system. The phototube detectors are mounted as an integral part of this system. Electronic preamplifiers immediately adjacent to the phototubes feed the electrical signal to remotely located electronic counting circuits.

The modified microscope optical system has been so arranged that, with the electronic light detector turned off, the specimen surface may be visually inspected through the transparent crystal with the aid of an appropriate microscope light source. By this means it is possible to correlate visually observed surface characteristics with measured activity.

Development of this instrument is continuing with efforts to improve the electronic circuitry and to further perfect the scintillator materials.

6. APPLICATION OF AUTORADIOGRAPHY TO SPECIFIC PROBLEMS

The application of the autoradiographic technique to a variety of problems has been noteworthy recently, and a description of two studies in which this laboratory has participated seems appropriate here.

One study by Dr. Sumio Yukawa and Professor M. J. Sinnott[20] is concerned, in part, with a study of the diffusion of nickel along bicrystal grain boundaries in copper. The relative rate of diffusion through the matrix of the crystal as opposed to that along the boundary and the influence of the angle between the planes forming the boundary on the diffusion rate has been measured.

The first problem solved was that of growing copper bicrystals with the desired angle between the 100 faces of the crystals. This was done by using single crystals as seeds for nucleating solidification and a furnace arrangement where the position of the liquid-solid interface is controlled. The charge metal is melted and fused onto the seed crystals, and then by progressive solidification, crystals having the same orientations as the seeds are grown from the melt. Very clean and satisfactory bicrystals with accurately controlled interface boundaries were grown in this way.

To measure diffusion of nickel in the copper crystals parallel to the bicrystal face and along the bicrystal face itself, radioactive nickel was used as a tracer. Specimens cut perpendicular to the bicrystal boundary were annealed and cleaned and then radioactive nickel plated onto one of the cut and cleaned faces. A special plating cell and techniques were developed for the plating operation.

The plated specimens were sealed under vacuum in Vycor glass capsules and placed in tubular muffle furnaces maintained at the desired temperature.

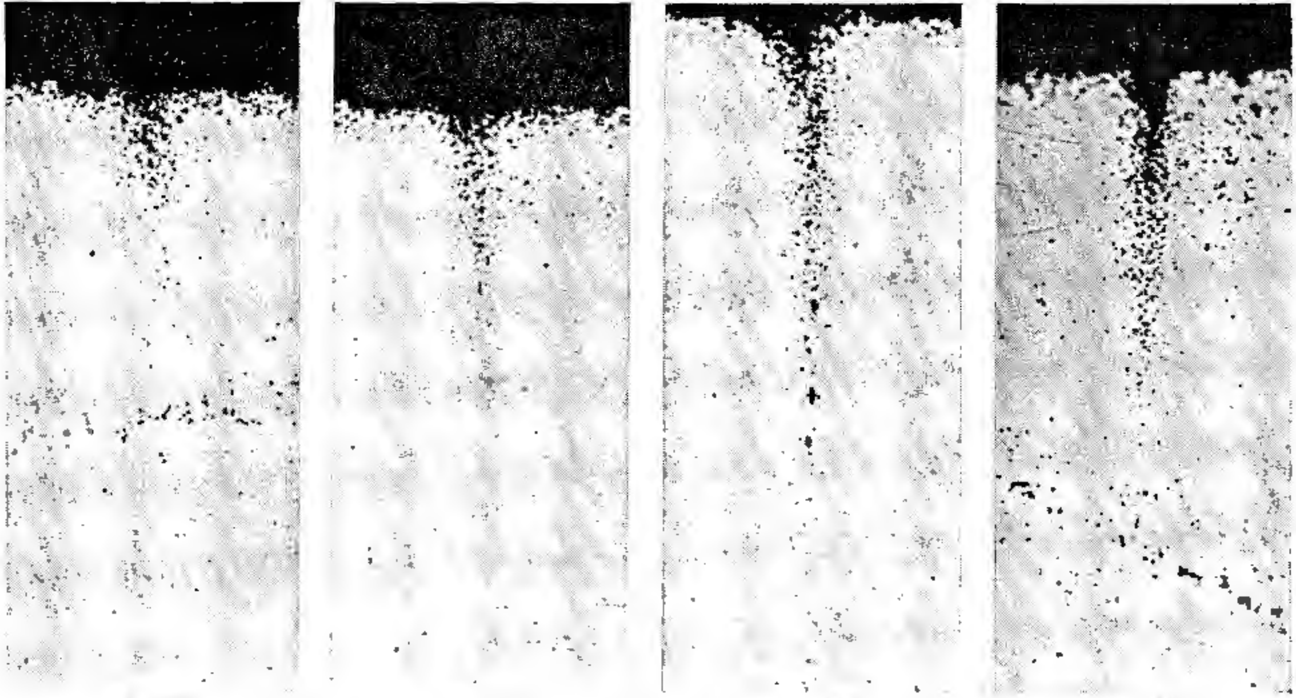
After diffusion, the samples were cut at right angles to the diffusion interface and the plane of the grain boundary. The specimens were mounted in Bakelite with the cut surface exposed and then polished and etched.

When the metallurgical preparation was completed, the specimen was given a coating of 2 per cent Vinylite VYNS in methyl-ethyl ketone as a protective layer. Stripping film was then applied and, after a suitable exposure, developed and fixed.

A typical set of results may be seen in Fig. 8. The influence of bicrystal angle on diffusion rate of the nickel into copper along the grain boundary and the very limited diffusion into the crystal lattice proper may be observed directly for the first time. The amount of diffusing metal involved here is well below the limit detectable by etching methods. From autoradiographs such as these, quantitative evaluation of activation energy for grain boundary diffusion as a function of angle has been made.

Work is now proceeding on the study of nickel-nickel diffusion, a problem which can be solved only with the aid of these high resolution autoradiographic methods.

The other study utilizing autoradiography is being carried out by Professor C. L. Markert of the Zoology Department, University of Michigan.[21] Professor Markert has been studying the precursors of melanin formation in the hope that by identifying particular substrates used in the synthesis of melanin pigments by cells of different genetic makeup, some correlation can be drawn between gene structure and enzyme specificity. The autoradiographic procedure has provided a unique method for carrying out this investigation. Briefly, the experiments have been conducted as follows: tissues are incubated with radioactively labelled materials thought to be precursors of melanin, and sections are prepared and exposed to stripping film (Eastman Permeable Base). The appearance of radioactivity in the melanin granules as revealed by autoradiographs indicates the extent to which the labelled substrate acts as a precursor in melanin synthesis.



$\theta = 15°$ $\theta = 31°$ $\theta = 45°$ $\theta = 66°$

Figure 8. Diffusion of nickel into copper bicrystals at 650°C (1200°F). Autoradiographs ×500; diffusion time, 144 hr; autoradiograph exposure time, 5 days

Several interesting results have been obtained from this study. When tyrosine or DOPA (dihydroxy phenylalanine) labelled in the side chain carbon were injected into the tissue cultures of chick epidermis and skin, in tadpoles, and in mouse skin (*in vivo* and *in vitro*), no radioactivity was detected in the melanin granules. Figure 9 shows the autoradiographic response of the carbon-14 DOPA polymerized in the gut of the tadpole while no activity is seen over the tissue melanin strip. However, when uniformly labelled tyrosine was incubated with tissue cultures of embryonic chick skin, radioactivity was localized to the melanin particles as noted in Fig. 10. This figure also serves to illustrate the problem of the film shifting during processing, since the emulsion containing the silver grains may be noted as having shifted to the right and up from the specimen. In this case, dense areas of silver may be correlated with the dark melanin granules. However, in less well defined situations, such shifting would prevent correlation of autoradiograph with specimen detail.

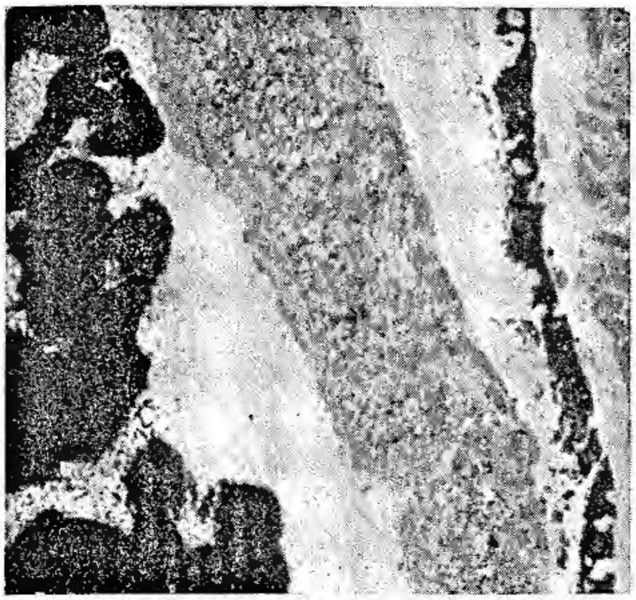


Figure 9. Stripping film autoradiograph of C^{14}-labelled DOPA localization in tadpole gut. (×500)

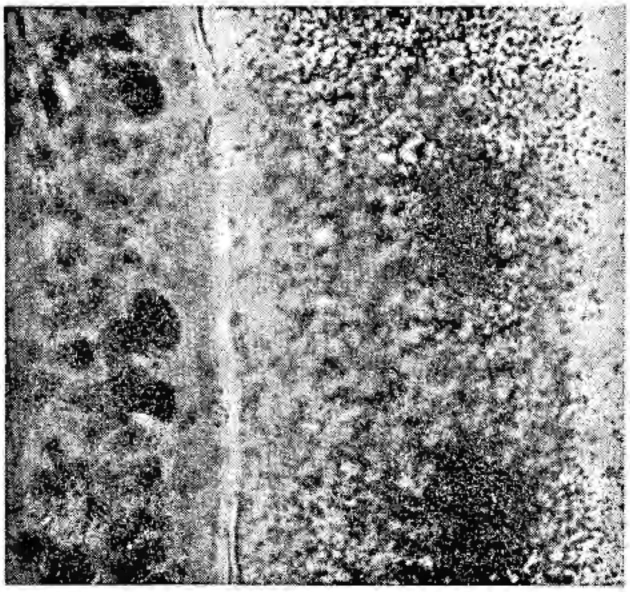


Figure 10. Stripping film autoradiograph of melanin granules in embryonic chick skin. Film has shifted to the right, uncovering granules in specimen below. (×1000)

The implication from these experiments is that the side chain of tyrosine is not involved in melanin formation. An even more recent study of this problem has indicated that yellow and black melanin granules which appear in strains of mice believed to possess only one gene not in common, are derived from the same substrate—the uniformly labelled carbon-14 tyrosine. Somewhat less activity appeared in the yellow granules than in the black under the same experimental conditions. Thus, it would appear from these studies that the site of gene action is not at the stage of initial substrate utilization—but effective further along the chain to melanin formation.

SUMMARY

The autoradiographic technique is finding increased applications in a variety of problems—revealing information not otherwise gained from other procedures. However, serious limitations exist on the resolution obtainable and on the chemical and light stability of the materials and techniques presently in use. Investigations in this laboratory have been directed toward creating new high resolution autoradiographic techniques. A thin photographic film suitable for autoradiographic work with the electron microscope is currently being developed. New systems utilizing polymerization reactions are aimed at yielding more stable high resolution detectors. A beta ray microscope enabling direct and immediate determination of the presence of radioactivity in a 75-micron field has also been developed and is presently undergoing refinement. With perfection of these developments, the autoradiographic technique should prove more valuable as a method for investigating fundamental scientific problems.

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New Geiger Tube Designs, Hollow Anode and Parallel Plate Counters

By J. Hermsen, A. M. J. Jaspers, P. Kraayeveld, and K. van Duuren,* The Netherlands

The geometric form of the Geiger counter tube has remained substantially the same since its invention in 1908. The anode is generally a wire of 0.1–1 mm diameter, coaxially fitted in the hollow cathode. With the conventional types of gas filling it is necessary to keep the anode diameter small to achieve adequate quenching of the discharge. Increasing of the anode diameter inevitably results in higher working voltage, greater slope of the plateau and shorter plateau length.

Recent experiments in this laboratory have shown that the use of the low-voltage halogen mixtures makes it possible to construct counters with thick anodes, that have very good counting characteristics. This alteration yields the advantage that the geometry of the design can be much better adapted to the pertinent application of the Geiger tube. An additional advantage is that the dead time can be made smaller. We made various experimental counter tubes with large anode diameter. The details of some examples that can be useful for radiochemical and tracer work will be given.

Increasing of the anode diameter offers the possibility of making the anode hollow. The radioactive source may then be placed within the counter and by this procedure and solid angle increases considerably. In Fig. 1 a cross-sectional view of a γ counter constructed according to this principle is shown. This counter is primarily meant for counting the radioactivity of a solution. In the hollow anode of 23 mm diameter is inserted a glass tube in which the liquid can be poured. The volume available for the liquid amounts to 20 cm^3. In the case of a counter filled with an iodine solution with a specific activity of 0.001 μc/ml, a counting rate of 62 counts/minute was observed, whilst the background count amounted to 100 per minute. The slope of the plateau is 2%/100 volts; the other counting properties are shown in Fig. 4.

A thin wall β counter suited for measuring the activity of a flowing liquid, for example with an ion exchange column, is shown in Fig. 2. The solution flows through the thin glass tube that has a wall thickness of 30 mg/cm^2. The sensitive volume amounts to 0.4 cm^3. The counting rate curve is given in Fig. 5; the slope of the plateau is 5%/100 volts.

As one could expect from the above mentioned results it was also possible to drop the cylindrical geometry. Flat parallel plate counters showed the

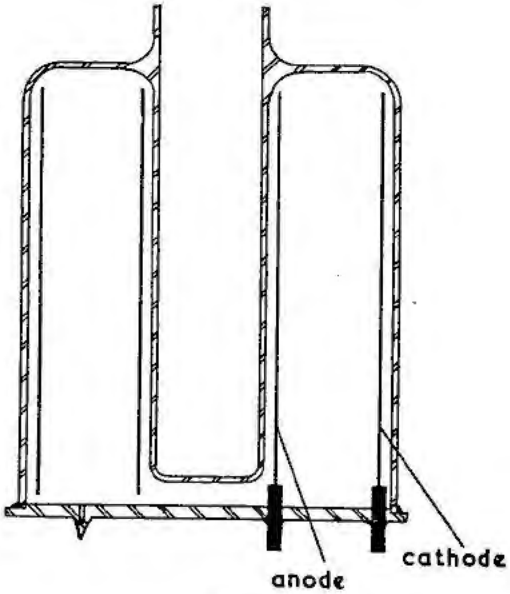

Figure 1

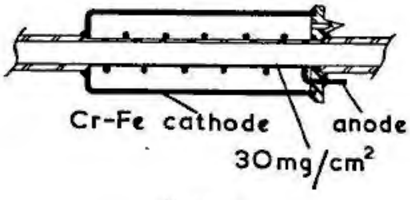

Figure 2

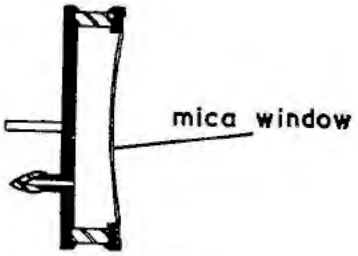

Figure 3

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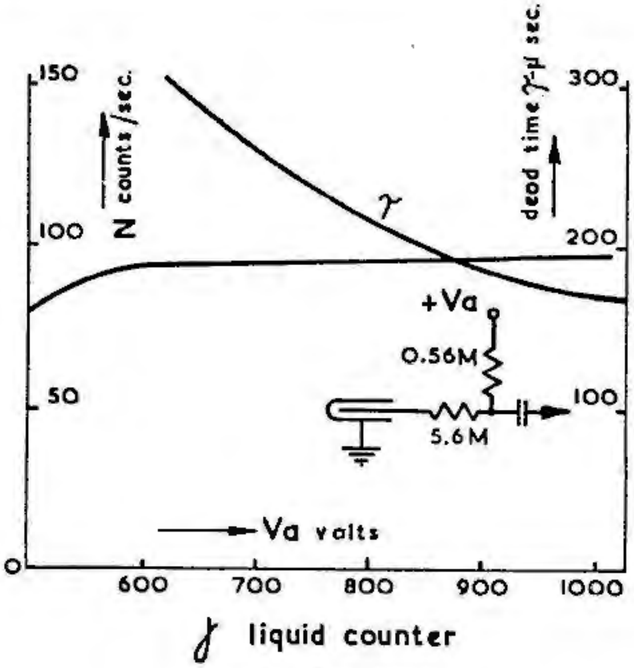

Figure 4. γ liquid counter

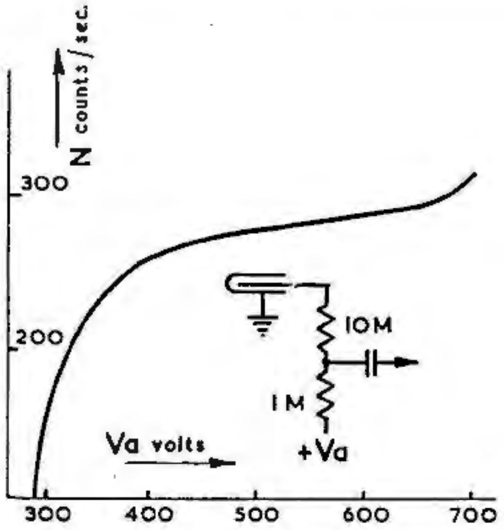

Figure 5. Thin wall flow counter

same favourable properties. An example from this is shown in Fig. 3; this is a mica window β counter with a small dead time. The window can be internally covered with a thin conducting layer or supported by a metal grid. The counting characteristics are given in Figs. 6 and 7. The plateau slope amounts to 5%/100 volts for a series resistance of 10 megohm.

The graphs of Figs. 6 and 7 show a common property of all these counters. The plateau and the dead time are strongly dependent on the value of the series resistance R. High values of R, for instance 10 megohm, give excellent plateaus. The dead time, however, is optimum with smaller resistance values. Therefore, the value of this resistance can be determined in accordance with the requirements of the experiment.

The authors are indebted very much to N. Warmoltz of the Philips Research Laboratories, Eindhoven, who suggested the idea of the halogen filled parallel plate counter and made the first trial with it.

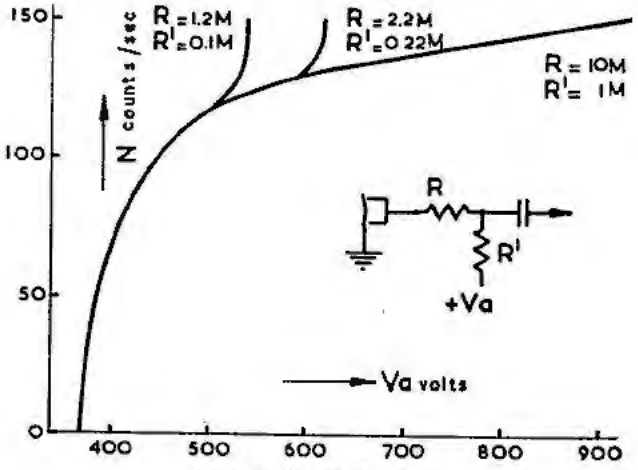

Figure 6. Flat plate counter

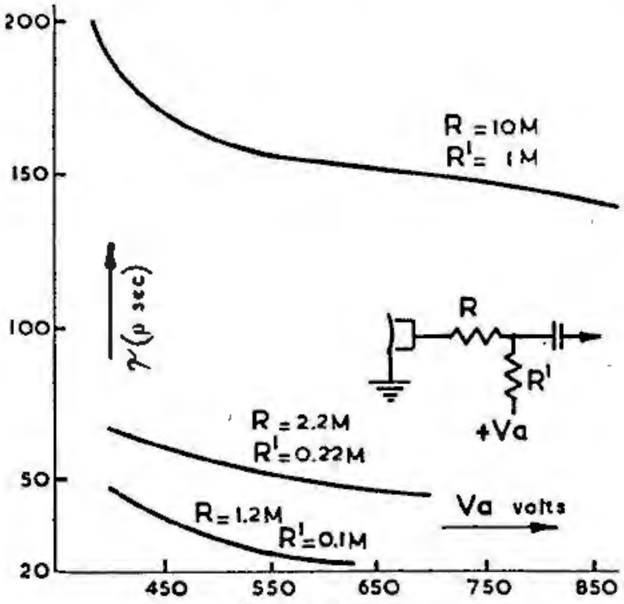

Figure 7. Flat plate counter

A Brief Review of the Radiation Instruments in Japan

By Fumio Yamasaki,* Japan

1. INTRODUCTION

Before World War II, the radiation instruments used in the field of the application of radioisotopes to research problems in Japan were limited to those such as Lauritsen electroscopes, and simple radiation counters with thin-walled Geiger-Müller counter tubes made of aluminium. Since the first import of radioisotopes from the United States in 1950, Geiger-Müller counting equipments with mica-window counter tubes have come into the market in addition to the Lauritsen electroscope of new design. In Japan, electronic industries are relatively well developed; this fact has resulted in a rapid progress also in the field of radiation instruments. At present, most of the instruments for ordinary purposes are supplied by several manufacturers. Moreover, the experience of the Bikini incident has served to accelerate the progress in this field.

The present status of manufacture and investigations of the radiation instruments in Japan will be summarized below.

2. ION CHAMBERS AND ELECTROSCOPES

The electroscope of the Lauritsen type was already made at the Scientific Research Institute more than fifteen years ago; the present model of which has the sensitivity of 1/200 mr per division, contains the electrostatic charging device in itself, and is widely distributed in this country. The pocket dosimeter with a full scale range of 200 mr and its charger of small size are also produced at the Scientific Research Institute, while the pocket ionization chamber and its charger-reader, as well as the precision r-meter and the minometer at Tokyo Shibaura Electric Co. Radiation survey meters of the ionization chamber type are manufactured at Kobe Kogyo Corp. and Tokyo Shibaura Electric Co., but miniaturization of electrometer tubes and high resistances are not yet completed.

One of the recent investigations in this field is that of the vibrating foil electrometer developed by G. Shinoda *et al.* at Osaka University. It is based on the modification of the principle originally established by Y. L. Yousef and R. Kamel,[1] under which an ac voltage is superposed to the dc signal of an electrometer, and the vibration of the foil electrode thus provoked is picked up electrically. Its frequency is equal to the first harmonics of the superposed ac voltage when the potential difference between the signal electrode and the vibrating foil electrode is zero. However, if any potential difference does exist, there appears the fundamental oscillation, the amplitude of which is proportional to the potential difference. This principle, when directly applied to the determination of the input dc signal, was found to be impracticable owing to the instability of operation due to microphonic and vibrational disturbances. Shinoda *et al.* have eliminated these defects by inserting two auxiliary electrodes for damping, and one more electrode has been added to suppress the unnecessary first harmonics. The displacement of the vibrating foil has been detected with a high sensitive capacity micrometer. A negative feedback incorporated in the electronic circuit improved the stability and reduced the apparent input capacitance. At present, an electrometer having the sensitivity of 10^{-16} coulomb and the apparent input capacitance of 10^{-3} pF is obtained.

3. GEIGER-MÜLLER COUNTERS

Mica-window Geiger-Müller counter tubes (window diameter 15–25 mm, window thickness 1.5–3 mg/cm^2) are manufactured at Kobe Kogyo Corp., Tokyo Shibaura Electric Co. and the Scientific Research Institute. All of them are filled with organic quenching gas, and the halogen-quenched ones are still at the research stage. Needle-type counters, stomach counters, as well as the Geiger-Müller counters for X-rays, gamma-rays, and cosmic rays are also produced.

Various types of scalers such as scale of 32, 100, and 1000, including those which use dekatron tubes, are made by several manufacturers. Since the Bikini incident, portable radiation detectors and rate meters have been widely distributed for hygienic purposes.

4. SCINTILLATION COUNTERS

Scintillation counters are extensively investigated and used at the physics department of several research laboratories, but they are not yet manufactured commercially. Production of inorganic scintillators such as thallium-activated sodium iodide and tin-activated lithium iodide is attempted at the Scientific Research Institute and the University of Tokyo. As for organic ones, terphenyl-polystyrene plastic scintillator has been studied by S. Schimizu *et al.* at Kyoto University. They have found an interesting relation between light output and degree of polymerization

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* The Scientific Research Institute, Tokyo.

of the scintillators. According to their data, it was confirmed that the luminescence in the plastic solution was mostly due to a non-radiative energy transfer from the solvent to the solute. This work will serve to produce new plastic scintillators of high efficiency.

As to photomultiplier tubes, those similar to RCA-5819 are being produced and tested at Tokyo Shibaura Electric Co.

Besides the instruments for the purpose of physical research, scintillation recording gamma-ray spectrometers of high resolution, scintillation scanners for scintigram, sensitive airborne as well as carborne scintillation geological survey meters are constructed and used at the University of Tokyo, Kyoto University, the Scientific Research Institute, etc.

5. PHOTOGRAPHIC PLATES AND EMULSIONS

Most of the photographic materials used in the several fields of application of radioisotopes, both in industry and research works, are supplied from the two large firms in Japan, namely the Fuji Foto Film Co. and the Konishiroku Photo Industry Co. Several kinds of industrial X-ray films with different degrees of sensitivity and with various resolving powers are now available for the radiographical purpose.

Nuclear emulsions are also produced, but they are not, as yet, so sensitive as to register an electron track at a velocity of minimum ionization power. Radioautographic plates of both stripping and contact types with the coating of these nuclear emulsions to 15 micron thickness have been manufactured and are being used by research workers.

6. RADIATION DOSIMETRY

In 1937, a standard ionization chamber was constructed at the Electrotechnical Laboratory of the Agency of Industrial Science and Technology (ETL), with which X-ray doses could be measured up to 300 kvp with an error of less than 0.5%. Recently the radioactivity of the Co^{60} gamma-ray standard solution prepared at the US National Bureau of Standards was measured at ETL using a 4π beta ray counter, and the observed value well agreed with a value within 3%. Investigations are made at the Scientific Research Institute on assaying Co^{60} and Na^{24} by the beta-gamma and gamma-gamma coincidence method, while Tl^{204}, P^{32}, and RaE are assayed by using an end-window G-M counting equipment having a definite solid angle. An equilibrium mixture of RaD + E + F is also assayed with a parallel plate alpha chamber. RaD + E beta standards similar to that of the US National Bureau of Standards are made at the Government Industrial Research Institute of Nagoya and the Scientific Research Institute by electrodeposition of them on palladium-clad or platinum-plated silver disc as a form of lead peroxide from alkaline media.

7. MISCELLANEOUS

Among neutron detectors, BF_3-filled proportional counters have been made successfully. The production of BF_3-filled and boron carbide-coated ionization chambers is now at the research stage. The construction of an electromagnetic isotope separator (calutron) which will enable the production of enriched boron compounds will be completed before long. The tissue-equivalent ionization chamber is investigated at St. Paul's University.

Electronic techniques of various kinds developed in a number of research laboratories have allowed them to supply instruments for various kinds of special purposes. Among them, a pulse-height analyzer has been devised by Shimizu *et al.* at Kyoto University, and the use of a 6BN6 gated-beam tube in the circuit insures the stability of operation and the simplicity of construction.

A new direct-reading counting rate ratio meter has been constructed by H. Miwa at Kobe Kogyo Corp. It is based on a principle that a voltage inversely proportional to one of the referred incoming pulse rates is obtained from a saw-tooth oscillator controlled by these pulses and smoothed by a low-pass filter, and then it is compared with the voltage proportional to the other referred pulse rate at the tank circuit in an ordinary rate meter. Fluctuation of both voltages inherently obeys the same statistics. The actual model of the counting rate ratio meter has shown an operating range of the ratio between 1/100 and 100 at the pulse rate of zero to 3000 cps with the accuracy of ±2%.

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The Development and Design of a Series of Precision Radioactive Airborne Particle Detectors for Health Monitoring and Control Systems

By Nicholas Anton,* USA

I. THE NEED FOR AIRBORNE PARTICLE DETECTION

The extensive investigations by health physicists and radiological safety specialists in recent years have clearly shown that the hazard due to inhalation of radioactive airborne particles is far greater than that due to generalized exposure to radiations from external sources. Furthermore, it has been established that inhalation, rather than ingestion, is the principal mechanism by which radioactive toxic substances are absorbed into the body. These factors have led Eisenbud, Blatz and Barry to urge that "time-consuming surface monitoring procedures should be de-emphasized in relation to air sampling and bio-assay criteria of absorption." It follows that reliable, accurate, and sensitive monitors for airborne radioactivity are of the greatest importance in installations where work with radioactive substances is in progress.

The presence of airborne particulate radioactivity may be unavoidable, as in uranium, radium, or other mining operations, or may be the result of an accident arising from the inexorable processes of aging and corrosion of the structural materials utilized in reactor installations. A number of such accidents have recently been reported in the handling of radium sources and in the operation of nuclear reactors. The importance of measurements of airborne radioactivity in establishing safe ventilation rates in mining operations has been emphasized by Tsivoglou and Ayer. It is obvious that the detection of unusual amounts of airborne radioactivity in the vicinity of a nuclear reactor can lead to the early discovery and remedy of incipient cooling system leaks, corrosion pitting, etc.

II. THE PROPERTIES OF AIRBORNE PARTICULATE RADIOACTIVITY

The particulate contamination may be roughly classified into three categories: (a) particles greater than about 10 microns in diameter which settle rapidly in air and become temporarily airborne only as a consequence of breezes and drafts; (b) those particles which have sizes in the range between 1 to 10 microns and which remain airborne for various periods of time depending upon particle size and concentration as shown in Fig. 1; and (c) particles smaller than about 1 micron which remain suspended in air due to Brownian motion and form so-called "stable" aerosols. These stable aerosols settle eventually for due to their random motion the aerosol particles undergo collisions with each other and coalesce into larger particles which sediment under the influence of gravity.

It can be shown that a stable aerosol coagulates more rapidly the smaller the particle size and the greater the number of particles per cubic centimeter of volume. This is illustrated by Fig. 1, based upon data of LaMer, Inn, and Wilson.

It can be seen that for an aerosol to remain stable for an appreciable length of time, it must be more dilute the smaller the particle size. Figure 2 shows

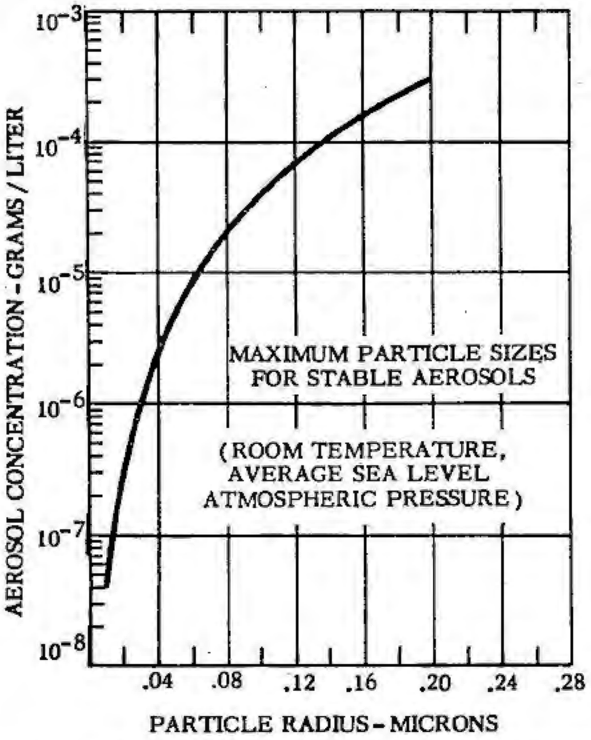

Figure 1. Maximum particle sizes for stable aerosols (room temperature, average sea level atmospheric pressure)

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the maximum aerosol concentrations, in grams per liter of air, that can exist in a stable condition for a considerable period of time for various radii of the particles comprising the aerosol.

It has been shown that aerosols having particle sizes of about 1 micron radius and concentrations of about 10^8 particles per cm^3 may be invisible in direct light; and aerosols of about 0.05 micron particle size containing up to 5×10^6 particles per cm^3 are so invisible as to fail to show a Tyndall effect of scattered light when illuminated with even the brightest light beams.

Wilkening has shown that about 90% of the natural radioactivity found on particulate matter in the atmosphere is attached to particles that have effective diameters less than 0.5 microns. Most of this fraction resides on particles in the size range 0.001–0.04 microns. The concentration of particles is quite small being less than 100 per cm^3. This, therefore, represents a very stable aerosol. The size and concentration of the aerosol particles produced by radon decay products is approximately the same as this, and it is clear that this radioactive particulate matter will form a stable, invisible aerosol under any conditions where radon (or natural uranium, thorium, or radium) is exposed to the air. This is the situation that prevails in uranium and thorium mining and processing, and in the radium dial-painting industry.

In the case of nuclear reactor technology, the particulate matter from plutonium processing is somewhat larger than the radon decay products, as are the aerosol particles to be expected from exposure to the atmosphere of sodium or potassium metal coolant, or from the escape of fission products from a homogeneous reactor solution. Furthermore, under all normal conditions the particle concentrations per unit volume of air that would be formed in this way would be very small. Hence, it may be concluded that such aerosols would also be stable and invisible.

III. SOURCES AND PERMISSIBLE LEVELS OF AIRBORNE ACTIVITY

Reference has already been made to the origin of airborne particulate matter arising from the exposure of natural uranium, thorium, or radium to the atmosphere. In uranium mining an additional source of particulates is the attrition of the rock and ore materials that inevitably accompanies all operations.

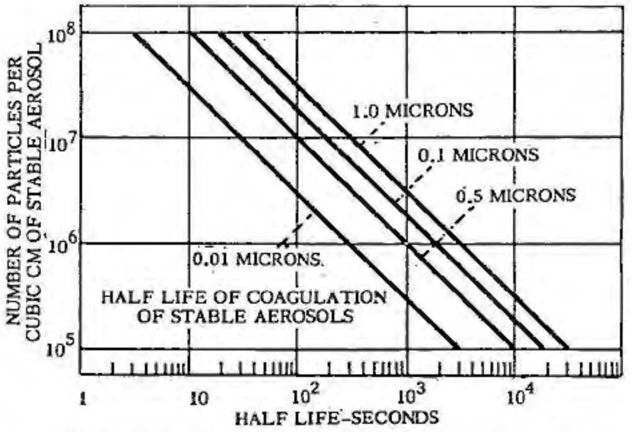


Figure 2. Half-life of coagulation of stable aerosols

Power and experimental reactors can give rise to a large variety of radioactive aerosols. Nonmoderated reactors require the most efficient heat transfer media and hence employ NaK eutectic. Although great strides have been made in liquid metal technology, the lifetime of such a heat transfer system is at present measurable in thousands of hours, and some leakage of the coolant may be expected eventually to occur with consequent release to the atmosphere of the 15 and 12 hour high-energy gamma-emitting isotopes of Na and K. The Los Alamos homogeneous reactor is gas-cooled, and the off-gas from the reactor is highly active, being rich in the activities from the iodine and bromine decay chains that yield xenon and krypton, and, finally, solids. Leaks from the cooling or recombiner system can generate aerosols containing these activities in the atmosphere. Breeder reactors have been proposed based upon a blanket of very finely divided ThF_4, with PaF_6 or UF_6 diffusing through it. This system could release fine particles of ThF_4 as a dilute aerosol under adverse circumstances. Other reactor types that might release particulate activities into the atmosphere include those based upon fluidized UO_2 particles circulated at 600–700°C, and aqueous suspensions of UO_2, U_3O_8 and UO_3 powders circulated at 300–370°C and 35–80 atmospheres, and suspensions of uranium in bismuth or of $U\text{-}Sn_3$ in bismuth-tin-lead alloy.

It is evident that air particle monitors are needed that can detect and monitor all of the types of activity referred to above. In addition, the monitors must be sufficiently sensitive to detect these activities at levels corresponding to the biologically maximum permissible amounts in air that may be regularly taken into the body without deleterious effects during the course of a lifetime. Table I summarizes the sources of airborne particulates, the nature and estimmated particle sizes of the aerosols, and the maximum permissible levels of radioactivity of these aerosols as recommended by the National Committee on Radiation Protection.

IV. EFFICIENCY OF AEROSOL FILTRATION

When an aerosol is drawn through a filter the solid or liquid particles suspended in the air may be captured by the filter as a result of one of the following mechanisms: (a) *Direct interception*—because of their initial positions some of the aerosol particles will approach the filter fibers along a path such that they are in a direct line with the fiber, and hence will strike it. (b) *Diffusion*—Brownian motion causes an aerosol particle to deviate from its original flow line. If this deviation causes a particle to touch a fiber, the particle will be captured. (c) *Inertia*—

TABLE I. Sources and Permissible Levels of Airborne Radioactivity

Operation	*Aerosol produced*	*Particle size, microns*	*Maximum permissible level, μc/liter*
Uranium mining	UO_2, U_3O_8, UO_3	0.5–10	1.7×10^{-8}
	Radon and active deposit	0.001-0.04	1.7×10^{-8}
Thorium processing	RaO (Ra^{228})	0.01–0.5	4×10^{-9}
Radium dial painting	RaO and active deposit	0.001–0.5	4×10^{-9}
Power and experimental reactors	Na_2O (Na^{24})	0.1–10	2×10^{-8}
	K_2O (K^{42})	0.1–10	2×10^{-8}
	I_2, NaI (I^{131} to I^{136})	0.1–10	3×10^{-6} 2×10^{-9}
	NaBr (Br^{82} to Br^{85})	0.1–10	2×10^{-9}
	ThF_4 (Th^{234})	0.1–10	6×10^{-4}
	UO_2 (U^{233})	0.1–10	1.6×10^{-8}
	Pu Oxides (Pu^{239})	0.01–0.5	2×10^{-9}

some of the flow lines of the gaseous medium past a filter fiber must necessarily exhibit marked curvature. Some aerosol particles will deviate from their flow lines because of inertia. If this deviation causes a particle to touch a fiber, the particle will be captured.

The efficiency of a given sample of filter paper in removing the particles from an air stream depends upon a number of factors, including the physical properties of the aerosol employed (i.e., the shape, particle size, density, concentration, and physical state of the particulates), the velocity of the aerosol through the filter, the moisture content of the aerosol, and the chemical nature of the particles.

Thus, it has been shown that large particles, of the order of 5 microns in diameter and greater, are readily deposited in a filter either by centrifugal force, due to the eddy currents set up in the filter, or by the small mesh. The velocity of diffusion and violent Brownian movement of the particles having diameters less than 0.1 microns assist their filtration by deposition on the surface of the filter fibers. Particles of an intermediate size are relatively highly penetrative. For example, in the filtration of a cloud of diphenylchlorarsine through a paper filter, the efficiency reaches a minimum value when the radius of the particles is between 0.1 and 0.2 microns.

Generally speaking, the efficiency of a filter decreases with increase of the rate of air flow, but at very high flow rates, turbulence is created at the paper surface, producing impingement effects that lead to increased efficiencies. Particles collect on the surface of an obstruction around which dust-laden air flows. This is a form of centrifugal precipitation resulting from the inertial resistance of the particle to change in its direction of travel in conformance with the streamlines of the flowing air. A number of applications of this principle have been made in air particle samplers in which the air is discharged through a nozzle at high velocity against a closely positioned collecting plate. The sharp, 90 degree bend in an air stream creates a strong centrifugal field, resulting in the deposition of particles with good efficiency. It has been claimed that a smoke-recording instrument based upon impingement of the dust on moving paper is about 35 times more sensitive than a similar device based upon filtration through paper. The efficiency of an air impinger decreases if the velocity of air flow is decreased.

Tachibana has found that the permeability of filter paper to an aerosol of ammonium chloride is greater when the aerosol is wet than when the aerosol is kept dry. Rowley and Jordan have demonstrated that the chemical nature of the dust particles has an important effect on the measured efficiencies of test filter papers.

From the foregoing, it is evident that data relating to the efficiencies of filter papers may show wide variations when the results of different investigators are compared. However, there is general agreement in the literature that the special asbestos-cellulose composition papers first introduced by the Drager Company function essentially as "absolute" filters—i.e., these papers have effectively 100% filtering efficiencies for aerosols of all types. By suitable treatment, asbestos is dispersed into extremely fine fibers, which are then mixed with coarser cellulose fibers to provide a support for the matte.

The reported efficiencies of a number of different commercial filter papers under roughly comparable conditions have been collected in Table II. The Hollingworth and Vose papers are asbestos cellulose; the others are high-grade rag papers consisting of almost pure alpha-cellulose.

V. PREVIOUS AIR PARTICLE MONITOR INSTRUMENTATION

A number of devices have been described previously for the collection and measurement of airborne particulate radioactivity. Instruments based upon a continuously moving strip of filter paper which is drawn past a sucking aperture where the airborne activity is deposited on it, and then under a counting tube, have been described by Kuper, *et al.*, and by Hogg. Reich describes a portable device in which the sampling port deposits the activity on a disc of filter paper until an arbitrary amount has been collected, and Hosey, *et al.*, have added provision for

TABLE II. Efficiencies of Several Commercial Filter Papers

Commercial paper	*Aerosol tested*	*Efficiency, %*
Whatman No. 41	Iron dust, 1–5μ	67
Whatman No. 41H	Iron dust, 1–5μ	46
Whatman No. 44	Normal air, 0.2–1.0μ	95
MSA Red All-Dust Filter	Iron dust, 1–5μ	91.5
Chemical Corps Type 5	Iron dust, 1–5μ	76
Hollingworth and Vose, Acc-6 and ACC-7	Iron dust, 1–5μ	100
Acc-6 and ACC-7	Methylene blue, 0.7μ	99.99
Acc-6 and ACC-7	Normal air, 0.2–1.0μ	99.98
Eaton and Dikeman 623-026	Chromic oxide	99.0–99.9

mounting 24 such discs in a large turntable in the instrument. Mansfield and Hogg have described systems in which air is drawn through two sampling ports simultaneously, and the activity is deposited as two side-by-side spots on a filter paper strip. The beta activity of one spot is measured, while the alpha activity of the other spot is determined. Instead of deposition on filter paper, use has been made of electrostatic precipitation of the particulates on an aluminum sheet in devices described by Wilkening and Hogg.

VI. LATEST DEVELOPMENTS IN AIRBORNE PARTICLE DETECTORS

Anton Laboratories recently completed the task of developing a series of precision radioactive airborne particle detectors whose purpose is to detect any sudden and dangerous use of beta particle radiation in the atmosphere of the upper reactor compartment and engine room of one of the proposed "nuclear" propelled submarines. This equipment serves as a typical example of equipment which has been designed to detect airborne particles produced by the first source listed in the introductory paragraphs of this paper, namely radiation caused by a loss or contamination of shield water, reactor malfunction, or other abnormal conditions.

A review of existing air particle detectors revealed that no one of them could fulfil the unusually stringent requirements which were needed for this application with respect to accuracy, ability to operate under adverse environmental conditions as well as reliability especially as would be required for continuous use for long periods under the varied severe conditions of marine service without overhaul and with very little maintenance.

The air contamination present was known to contain sodium-24, and it was accordingly requested that the detectors be calibrated in microcuries per cubic centimeter of Na^{24}. Other prime requirements which were set up can be stated briefly as follows:

a. Range: tolerance was defined as a concentration of Na^{24} particles in air in the amount of 2×10^{-7} microcuries per cubic centimeter. The equipment was to detect over a range of 2 × tolerance (0.4×10^{-7} μc/cm^3) to 25 × tolerance (50×10^{-7} μc/cm^3) in the presence of a gamma background of 100 mr/hr.

b. Environmental conditions: (1) continuous and satisfactory operation over the temperature range of −0°C to +65°C; storage up to +80°C; (2) satisfactory operation after being subjected to shocks of up to 1200 ft-lb; continuous operation while vibrated over the range of 10–33 cycles/second; satisfactory operation while inclined 30 degrees with the normal and while subjected to a sudden roll of 60 degrees each side of vertical; and (3) satisfactory operation while subjected to the standard military conditions of humidity and fog as well as transient fluctuations in pressure.

c. Alarms: adjustability of the alarm over the entire range of detection. Provision for remote indicators (250 ft).

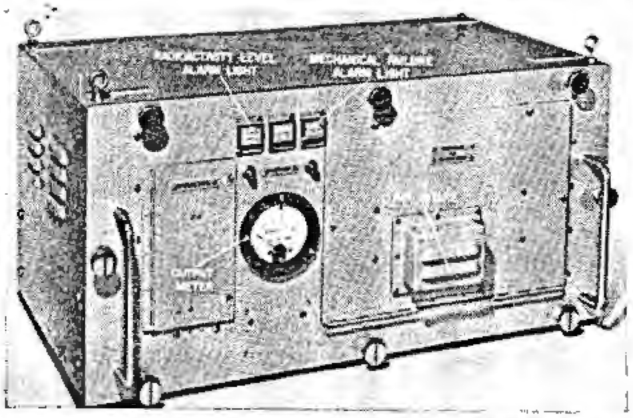
d. Input: ability to operate within required accuracy for a variation in input amounting to ±10% of line voltage and ±5% of line frequency.

e. Size and weight: reduced to as compact a size as practically possible consistent with ruggedness and reliable operation.

f. Reliability: stable operation with reduced requirements for adjustments and alignment.

While it is true that most laboratory installations do not need all of the requirements set forth by the military, there can be no argument at all that an equipment which meets these requirements is certainly one whose dependability cannot be questioned. In fact, most laboratories are now requiring that their equipments be able to pass military specification, because it has been recognized that the resulting equipment is superior. They are discovering that their equipments even though located in a laboratory do quite often receive very severe treatment, and that a "solderized" equipment ultimately delivers the best service.

A description of this particular unit serves to illustrate the many unusual and desirable features which can be designed into any given equipment. This air particle detector consists basically of an air flow system which draws air continuously through a filter paper strip; a paper drive system for the filter strip; a shielded integrator tube which serves as the detector of both gamma and beta rays; and an electronic system for amplifying the detector signal and indicating on the associated meters and actuating the alarms. A brief description of each of these systems follows.



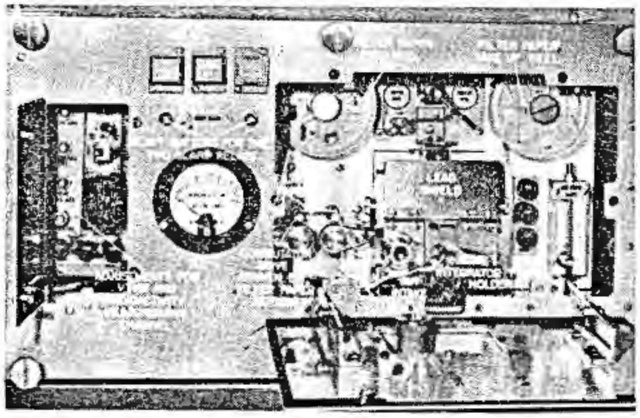
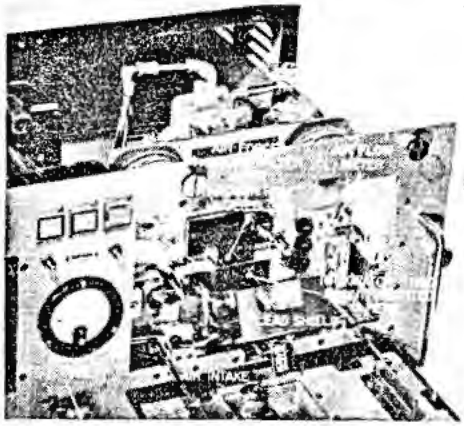


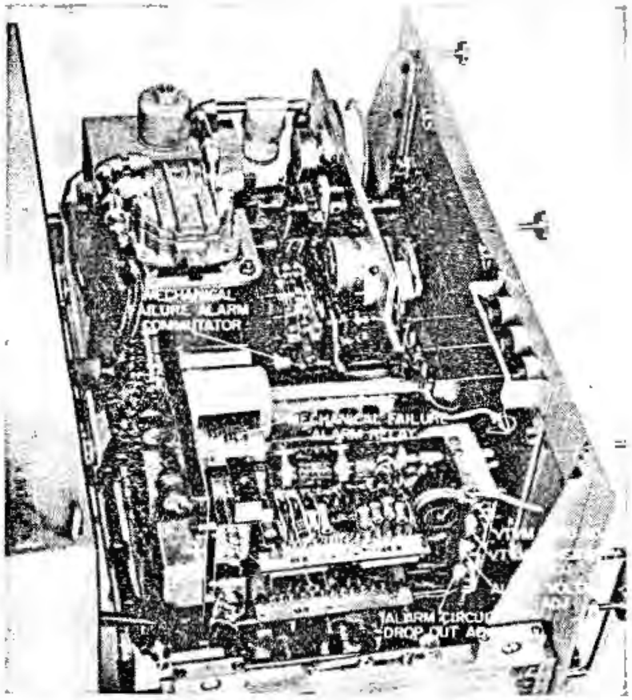
Figure 3 (above left). Front view of air particle detector showing location of indicating meter, alarm lights and controls

Figure 4 (above right). Front view of air particle detector with access doors open to show details of paper drive, tube shield and air systems

Figure 5 (below). View of air particle detector with chassis drawn out on its slides. Lead shield is also open and detector tube has been removed

Figure 6 (right). Top view of chassis of air particle detector showing details of electronic system





Figures 3 through 9 delineate this equipment in detail and should be referred to in connection with the following description. The rugged stainless steel enclosure measures 24½ in. wide × 12 in. high × 13 in. deep and weighs 175 lb. The meter face shown in the photographs represents still another range which is available with this equipment: (0.5 to 500) $\mu c \times 10^7/cm^3$. It can be readily seen that maintenance and accessibility have been prerequisites of the design.

Air Flow System

The air flow system consists of a 60-cycle split-phase motor directly coupled to a positive displacement, oilless, vacuum pump. It is this pump which draws the air into the system and through the filter paper. The air also passes through an air flow meter which measures the volume of air flowing through the system. The flow meter is located inside the large front panel door and is mounted on the right side of the paper drive plate. The adjustment valve on top of the flow meter is pre-set in the laboratory and is then locked in place. The air flow is readjusted whenever necessary by means of the bleeder valve mounted just behind the air flow meter. The normal air flow for this equipment is 20 cubic feet per hour.

A differential pressure switch monitors the pressure drop across the filter paper. This switch is connected with one end open to the atmosphere and the other end tapped into the air line between the output of the lead shield and the input to the flow meter. The shaft moves back and forth along its axis as the difference in pressure between the two ends changes. Two round discs mounted on the shaft move with the shaft. Two normally closed microswitches are rigidly mounted between the discs in such a way so that as the discs move back and forth one or the other of the switches will be activated by one of the discs. The discs are movable along the shaft and can be adjusted to determine the amount of differential pressure change that is required to activate one of the switches. Because the pressure drop measured is the drop across the paper it is a measure of the actual air flow through the paper. The microswitches are connected in the mechanical alarm system in such a way that an alarm is given if either of the switches is activated. The differential pressure switch therefore indicates when the air flow falls too low or rises too high; if the air input orifice is clogged; or if the paper breaks and is thus removed from in front of the air orifice.

Paper Drive System

The paper drive system (illustrated in Fig. 10) consists of a 60-cycle drive motor which operates at one revolution per hour, and two additional hysteresis motors, for the supply reel and for the take-up reel. The drive motor turns a driving shaft which accurately controls the speed of the paper. The take-up reel motor operates at 1 rpm and the supply reel motor operates at 10 rpm; the take-up and supply motors turn in a clockwise direction looking at the motor from the shaft end and the drive motor turns in the counter clockwise direction. Therefore, the supply reel pulls back on the paper opposing the motion of the driving shaft; and the take-up reel pulls forward on the paper. However, these motors have been designed so that the torques which they exert are sufficient only to maintain the paper taut and will not cause the paper to tear.

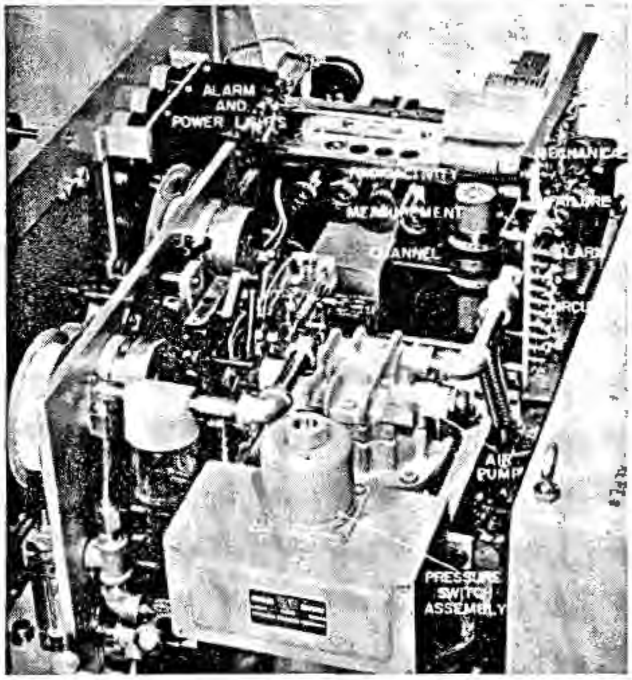


Figure 7. Top view of chassis of air particle detector showing details of air flow system and paper drive mechanism

The diameter of the paper drive capstan is 0.31 in. and it is geared to the drive motor by a 18/28 gear ratio. The speed of the paper is therefore $18/28 \times 0.31\pi = 0.625$ in./hr.

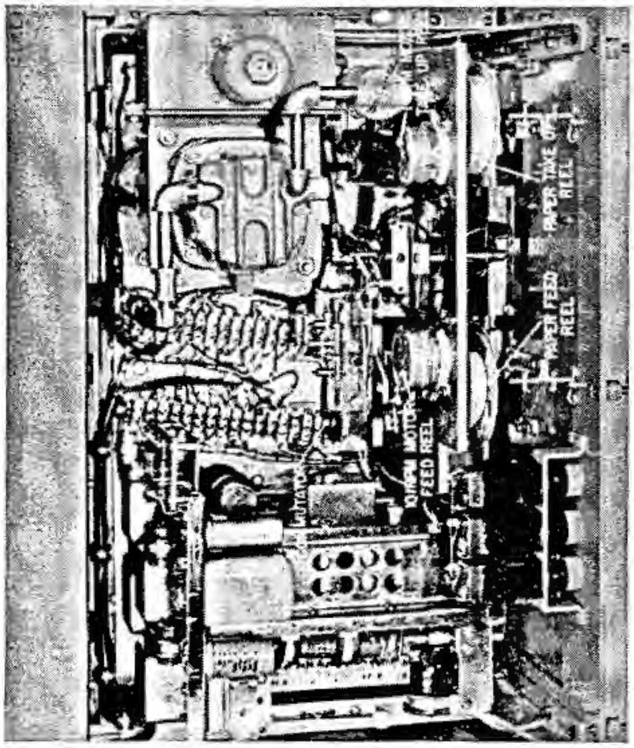


Figure 8. Head on view of top of air particle detector

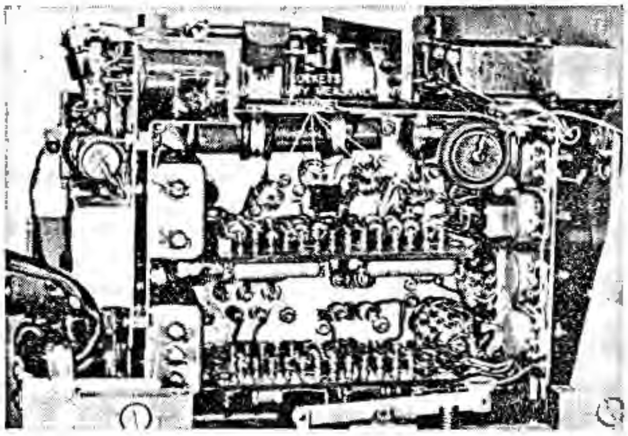


Figure 9. Head on view of electronic chassis of air particle detector

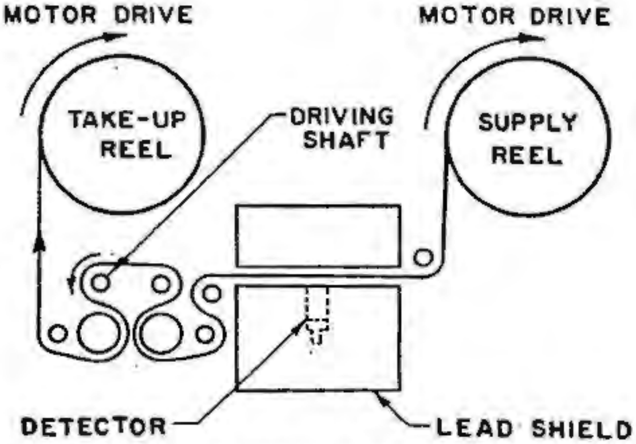


Figure 10. Paper drive system: showing how filter paper tape is maintained taut

In addition to the driving capstan there is another similar capstan which is driven by the paper and it in turn drives a rotating commutator which is a novel and integral part of the mechanical failure alarm system. This rotating commutator is described in greater detail later in this paper.

In traveling through the lead shield which houses the radiation detector element, the paper passes over two openings. The first opening is an air orifice through which passes air drawn in from the outside and then through the filter paper. In passing over the second opening, the paper which is now loaded with the radioactive particles which have been filtered from the air system is now scanned by the detector. The Anton Type 214 integrator tube which serves as the detector element in conjunction with the electronic circuitry measures quantitatively the amount of contamination deposited by the continuous air stream on the filter paper.

Each reel of paper which is supplied with the unit contains about 40 feet of filter paper and will last for approximately 30 days of continuous operation.

The filter utilized is Hollingworth and Vose No. H-70 asbestos-cellulose tape which is ⅝ in. wide and gauze backed for added strength which may be necessary in damp or very humid atmospheres.

Detector and Shield

The detector tube utilized in this equipment is an Anton Type 214 beta-gamma integrator shown schematically in Fig. 11. While mechanically the integrator tube appears to be very similar to a Geiger counter tube, there is a considerable difference in its performance. The output of a conventional Geiger counter tube consists of a series of pulses having a rate of occurrence that is related to the rate of intercepted ionizing events. The Geiger tube is therefore essentially a pulse generating tube to be used in a circuit that counts the number of ionizing events occurring within the tube. The associated counting circuit is triggered by pulses that have an amplitude above a certain height and generally consist of a scaler or ratemeter. Because the charge per pulse is usually small, the ion density within the tube is low and the ionic bombardment of the cathode does not become a serious problem.

An integrator tube such as that utilized in this equipment, while very similar to a conventional Geiger tube in both construction and basic operation, produces a pulsating *average current* which is proportional to the radiation intensity and hence requires processing of the cathode and anode to enable these electrodes to withstand the higher current necessitated by the type of circuitry which is employed with the integrator tube. In such a circuit the integrator tube usually delivers a very sizable current (of the order of 30–50 μa) which is read directly on a series microammeter. If the electrodes were not specially processed to withstand the bombardment they receive from the high ion densities, they would disintegrate rapidly and tube failure would result.

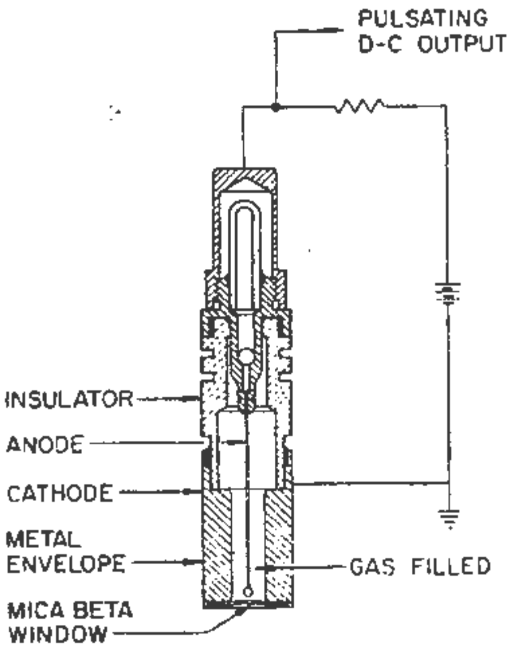


Figure 11. Anton type 214—details of integrator tube

These tubes cannot obviously be produced using an organic quenching material for such a quenching agent would immediately be destroyed. Non-destructible materials—in this case halogens (combinations of chlorine and bromine) are utilized for quenching. The quenching process depends on the ability of the halogen molecule to dissociate while performing its quenching duty and to then recombine in time to quench again These tubes are the sole development of the Anton Laboratories and have found very wide application because they cannot be destroyed by overvoltage; operate over a range of −50°C to +175°C; are stable and readily interchangeable.

The integrator tube does not necessarily have a Geiger-Mueller plateau, but is characterized by a response curve of average current versus applied voltage. "Cleanliness" of pulse, dead time, etc., are of very little importance. In utilizing these tubes the stability of the power supply voltage is important and should be held within close limits since the tube output is a function of the voltage supplied to the anode.

The detector tube is shielded with approximately 2 in. of lead to screen out the background gamma flux in which the system is designed to operate, and this background is almost completely attenuated before it can reach the detector. The detector itself is sensitive to both beta and gamma radiation—its mica window is 1.4 mg/cm^2 thick and its stainless steel cathode is 0.047 in. thick. A passageway cut through the lead shielding conducts the sample of air from the external atmosphere to the filter paper and out again. The air port is located so that the air sample is passed through the paper just before the paper passes in front of the tube, giving a minimum of delay in detection of any change in atmospheric contamination. The passageways have been designed aerodynamically to prevent particles from being dropped out of the air stream and have been highly polished to facilitate cleaning in the event of contamination.

The integrator circuit is shown in Fig. 12. The pulsating current produced by the tube passes through the *RC* network (R_2, R_3, C_2) and because of the long *RC* time constant (44 seconds) is further integrated and results in an average dc current which produces across the integrating network a dc voltage the amplitude of which is determined by the strength of the radiation field which the tube sees.

The 4 and 7 megohm resistors (R_2, R_3) are used instead of a single 11 megohm resistor to provide a voltage divider such that 4/11 of the total dc voltage developed can be applied to the vacuum tube voltmeter and alarm circuits which are discussed in detail later.

A typical characteristic curve for the Type 214 integrator tube and integrator circuit is shown in Fig. 13. The dc voltage developed across the integrating network is proportional to the logarithm of the ionization intensity. This relationship between the dc voltage developed across the integrating circuit and the ionization intensity may vary within limits from tube to tube for a given R_1C_1 combination. Because the R_1C_1 combination aids in the shaping of the final calibration curve, it is possible to accept integrator tubes whose individual responses vary within rather broad limits, and by suitably choosing the values of R_1 and C_1, bring the resulting calibration curves into agreement within ±5%.

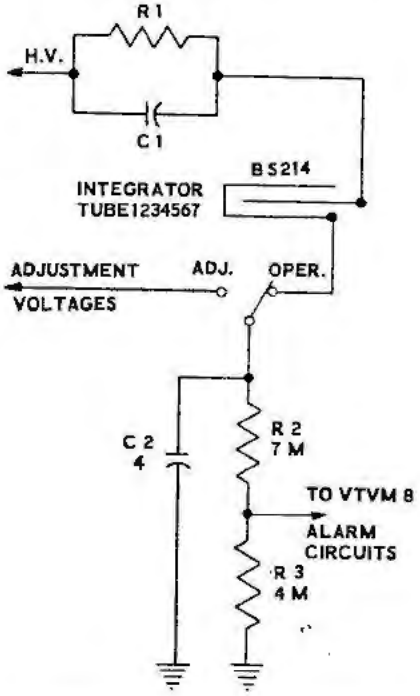


Figure 12. Integrator tube circuit

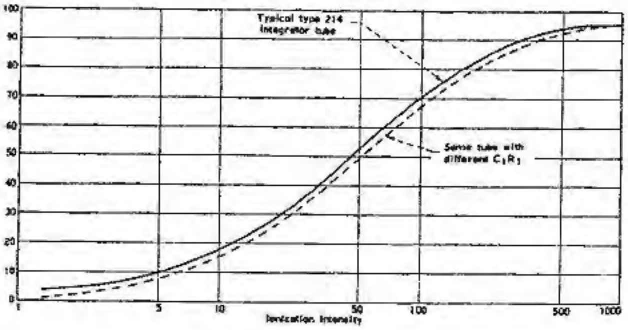


Figure 13. Typical characteristic curve for type 214 integrator tube and integrating circuit

In the present equipment the 214 tube is supplied with its R_1C_1 combination as a calibrated unit and this unit is therefore always directly interchangeable with any other such unit.

Electronic System

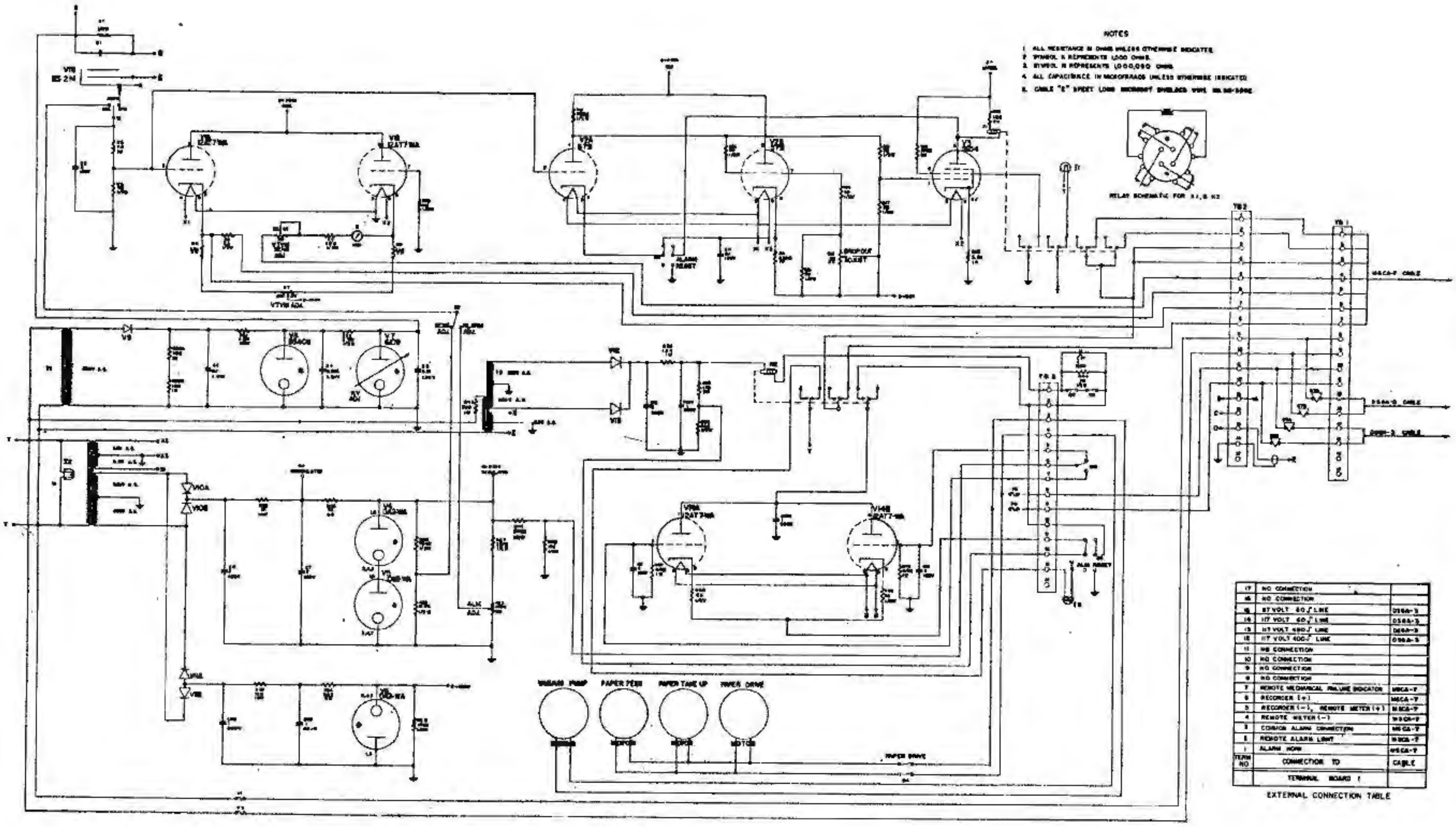
The electronic system consists of the following four components: (1) the vacuum tube voltmeter which receives the dc voltage signal from the integrator circuit and produces a corresponding scale deflection on a calibrated meter; (2) the alarm circuit, which also receives the dc output voltage from the integrator circuit—when this voltage reaches a preset level, the alarm is activated, giving local and remote alarm indications; (3) the power supply, which supplies a regulated positive high voltage to the integrator circuit as well as positive regulated and unregulated and negative regulated voltages to the vacuum tube voltmeter and alarm circuits; and (4) the mechanical failure alarm system which gives an indication if either the air flow or paper drive systems fail to operate correctly.

A brief description of each of these major components follows. A complete schematic diagram of the complete equipment is shown in Fig. 14.

Vacuum-Tube Voltmeter

The vacuum-tube voltmeter is a differential type cathode coupled vacuum-tube voltmeter using a 12AT7WA dual triode. Both sections of the 12AT7WA are connected as cathode followers with the indicating meter circuit connected between cathodes. The general circuit arrangement is shown in Figs. 15 and 16.

Because of the cathode follower connection, the cathode voltage of each section is equal to the zero input bias level plus a voltage approximately equal to the voltage applied to the control grid. If V_1 and V_2 have the same characteristics, with no input to either grid, the cathode voltages are the same. When a positive voltage is supplied to the grid of V_1, a corresponding rise in the voltage at the cathode of V_1 occurs. A difference of potential between the cathode of V_1 and that of V_2 is therefore established



17	NO CONNECTION	
16	NO CONNECTION	
16	117 VOLT 60 ∫ LINE	D58A-3
14	117 VOLT 60 ∫ LINE	D58A-3
13	117 VOLT 400 ∫ LINE	D58A-3
12	117 VOLT 400 ∫ LINE	D58A-3
11	NO CONNECTION	
10	NO CONNECTION	
9	NO CONNECTION	
8	NO CONNECTION	
7	REMOTE MECHANICAL FAILURE INDICATOR	MSCA-7
6	RECORDER (+)	MSCA-7
5	RECORDER (−), REMOTE METER (+)	MSCA-7
4	REMOTE METER (−)	MSCA-7
3	COMMON ALARM CONNECTION	MSCA-7
2	REMOTE ALARM LIGHT	MSCA-7
1	ALARM HORN	MSCA-7
TERM NO	CONNECTION TO	CABLE
	TERMINAL BOARD 1	

EXTERNAL CONNECTION TABLE

Figure 14. Over-all wiring diagram for air particle detector

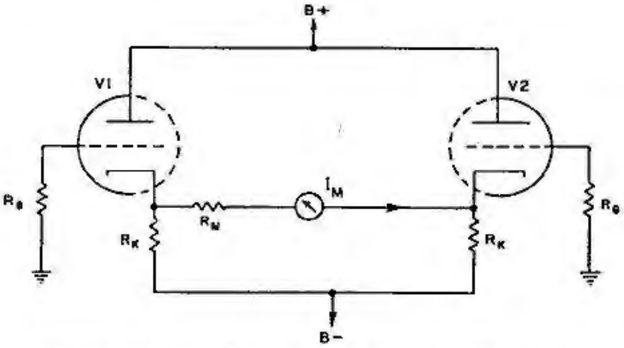


Figure 15. Generalized vacuum tube voltmeter circuit

which is directly proportional to the input signal voltage which was applied to the grid of V_1. The indicating meter circuit is essentially a voltmeter which measures this difference of potential. In order to insure reliable and stable operation of the equipment operation which would not be affected by change in tube characteristics because of age and severe environmental conditions—it is necessary to design this circuit so that these parameters will have a minimum effect.

It may be shown that I_m, the current in the output meter in the circuit of Fig. 15, where two identical tubes are used is given by:

$$I_m = \frac{g_m R_k E_i}{2R_k + R_m\left[1 + \left(\frac{1}{r_p} + g_m\right)R_k\right]} \qquad \text{(I)}$$

where g_m = mutual transconductance, r_p = plate resistance, and E_i = voltage input between the grid of V_1 and ground.

The assumption of identical tubes is one of analytic convenience and is not a necessity.

In general $g_m >> 1/r_p$ so that:

$$I_m = \frac{g_m R_k E_i}{2R_k + R_m\,[1 + g_m R_k]} \qquad \text{(II)}$$

If R_k is now chosen sufficiently large so that $g_m R_k >> 1$, the expression for I_m reduces to

$$I_m = \frac{E_i}{R_m + \dfrac{2}{g_m}} \qquad \text{(III)}$$

Finally, if R_m is large as compared with $2/g_m$ the meter indication is approximately equal to E_i/R_m and is essentially free of error caused by variation in tube characteristics. Component values have been chosen so that these conditions are attained in the actual equipment.

In addition to indication on the front panel meter provision has been made for insertion of a remote 1-milliampere full-scale meter whose resistance can be compensated for by adjustment of the sensitivity potentiometer. A precision 50-ohm resistor R_5 has also been provided to make available a voltage proportional to I_m for a remote 50-millivolt recorder.

Alarm Circuit

As shown in Fig. 17, the alarm circuit consists of a 5751 dual triode tube (V_2) and a 5654 pentode (V_3). The 5751 is connected as a Schmitt-Trigger circuit which converts the dc voltage from the integrator circuit to a dc voltage whose magnitude decreases abruptly when the integrating network output voltage reaches a pre-determined level. The 5654 is used as a switching tube to which is applied a portion of the abrupt change in voltage from the Schmitt-Trigger circuit causing a relay to drop out and activate local and remote alarms.

The Schmitt-Trigger circuit (V_2) is a cathode coupled bistable multivibrator (a circuit in which there are two stable states). In the normal state with no input voltage on the grid of V_{2A}, V_{2B} is conducting and V_{2A} is held cutoff by the cathode bias developed by the current of V_{2B} through the common cathode resistor R_{11}. As the voltage on the grid of V_{2A} becomes more positive a point is reached at which the cathode bias is overcome to a sufficient extent to allow V_{2A} to start conducting. When V_{2A} conducts there is an additional voltage drop across R_{12} and the voltage on the plate of V_{2A} becomes less positive. A portion of this drop in voltage is transmitted to the grid of V_{2B} through the voltage divider (R_{13}, R_{14}, R_{15}, R_{31}). The plate current of V_{2B} will therefore decrease and will cause a resultant decrease in the cathode bias which will then cause a further increase in the conduction of V_{2A}. This is a closed cycle which will continue until V_{2B} is cut off and V_{2A} is conducting. This is the second stable state of the Schmitt-Trigger circuit and the circuit will remain in this state until the voltage at the grid of V_{2A} goes below a certain level at which time the circuit will flip back to its original state. The dc level at which the flip back occurs is less positive than the level required to start V_{2A} conducting due to a hysteresis effect.

The 5654 switch tube (V_3) is connected as a pentode amplifier and a relay (K_1) is connected in its plate circuit. The grid voltage of V_3 is determined by the voltage at the plate of V_{2A} through the voltage divider (R_{16}, R_{17}). When V_{2A} is cut off, that is, the normal condition, the grid voltage of the switch tube

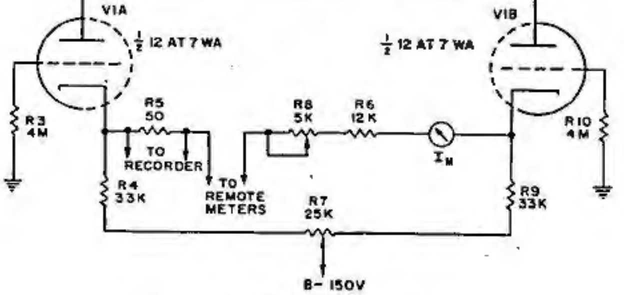


Figure 16. Actual VTVM circuit

is such that the tube will conduct heavily enough to hold the relay K_1 in the energized condition. When the Schmitt-Trigger circuit flips to its other stable state and V_{2A} conducts, part of the drop in plate voltage of V_{2A} is transmitted to the grid of V_3. This drop in dc level is sufficient to decrease the plate current of V_3 to a point below the holding current of relay K_1 and the relay will drop out. K_1 is a four-pole double-throw relay. Three of the poles are wired in such a way that when the relay is de-energized, circuits controlling the local and remote alarms are closed and the alarms are actuated indicating an excessive radiation level or a circuit failure, i.e., open relay coil, open heater on V_2 or V_3, etc. The fourth pole of the relay contacts is used as a holding contact to keep the relay de-energized after it has dropped out by grounding the screen of the 5654 thus ensuring that plate current will not reach the level required to pull the relay in until the alarm reset switch S_3 is depressed. In this way when an alarm indication is given, the indication will remain even if the Schmitt-Trigger circuit should flip back to its original state. The alarm reset switch serves a two-fold purpose. It grounds the plate of the switch tube causing sufficient current to flow through K_1 to pull it in. It also opens the cathode circuit of V_{2A} insuring that the Schmitt-Trigger circuit is returned to its normal state by allowing V_{2B} to conduct again. This is necessary because of the hysteresis effect mentioned earlier in the discussion of the Schmitt-Trigger circuit.

The filaments of V_2 and V_3 are wired so that if either half of the filament of V_2 or V_3 should open the alarm will indicate, thus making the alarm circuit fail-safe.

Power Supplies

High-Voltage Supply

This supply is a half-wave rectified power supply which is capacitance filtered together with a two-stage regulating circuit to provide regulation and adjustability of the output voltage for the integrator tube. This power supply is shown in detail in Fig. 14.

The anode voltage of V_8 is 750 v and is constant within ±3 volts or 0.4% for a line voltage change within ±10%. This degree of regulation is, however, not entirely adequate because of the sensitivity of the integrator tube current to changes in applied anode voltage. The anode voltage of V_8, the first regulator tube, is, therefore applied to the second regulating circuit, which consists of a series resistor (R_{22}), corona voltage regulator tube (V_7) (Anton Type 6109 adjustable corona voltage regulator) whose operating voltage is adjustable between 645 volts and 705 volts by means of a screwdriver adjustment built into the tube. The cascaded regulator circuits make it possible to hold the anode voltage of V_7 constant to within ±0.15 volts for the above mentioned ±10% change in line voltage. The output ripple in the anode voltage of V_7 is less than 10 mv. This degree of regulation makes the average output current and consequently the output voltage of the integrator assembly almost completely independent of changes of line voltage.

B^+ 255-Volt Supply

The B^+ 255-volt supply is a full-wave rectified supply with two stage RC filtering. The regulation of the output voltage is accomplished by means of two glow VR tubes in series. This supply is regulated to within ± 2 volts or less than ± 1% for a ± 10% change in line voltage, and with an output ripple of less than 10 mv. Resistors R_{25} and R_{26} provide a 42.5 v tap from the +255 v regulated supply to check the sensitivity of the Vacuum Tube Volt Meter. Resistor R_{27} and potentiometer R_{32} provide a regulated voltage which is variable from zero to about 60 v so that the tripping level of the alarm circuit may be set.

B^- 150-Volt Supply

The B^- 150-volt supply is also a full-wave rectified supply, with two stage RC filtering and regulation of the output voltage by means of a single glow VR tube.

The same secondary winding from which the B^+ voltage is obtained is used for economy of space to derive the B^- voltage by using a rectifier with reversed polarity.

The B^- 150-volt supply is regulated to within ± ½ volt or about ⅓% for a ± 10% change in line voltage and with an output ripple of less than 5 mv. From this power supply is obtained B^- 150 v regulated to supply the cathode returns for the vacuum tube voltmeter tube (V_1), and the Schmitt-Trigger tube (V_2).

Mechanical Failure Alarm System

The mechanical failure alarm system has its own power supply which operates off transformer T_3. The selenium rectifiers (V_{12} and V_{13}) form a full wave rectifier which supplies a pulsating dc voltage to filter capacitor C_{13}. Additional filtering is obtained from R_{34} and $C_{10}A$. The output voltage of this power supply is about +250 v and is unregulated.

The mechanical alarm system utilizes a circuit wherein a twin triode (V_{14}) whose anodes are tied together and have a relay coil for a common plate load. The grid circuits of V_{14} each contain a long time constant tank circuit (C_{11}, R_{37} and C_{12}, R_{40}). These tank circuits are charged alternately to a regulated voltage determined by the voltage divider R_{41} and R_{42} from the B^+ 255-volt supply, through the rotary commutator switch (S_{10}). The commutator is driven by the paper through the driven shaft and its associated gears. The biases of V_{14} have been chosen so that if both tank circuits are fully charged or near full charge the current through V_{14} is more than the drop out level of K_2 and the relay is held in. If however, either of the tank circuits becomes fully discharged the tube current decreases below the drop out level of K_2 and a mechanical failure alarm indication is given. The rotary switch is adjusted so that as it rotates the grid tank circuits are alternately charged. During normal operation each tank circuit will maintain a sufficient amount of its charge to hold in the relay. If, however, the paper stops thus stopping the rotation of the commutator one of the tank circuits is discharged completely; relay K_2 drops out and a mechanical failure alarm indication is given. S_5 and S_6 (the differential pressure microswitches) are in series with K_2, and thus if either of them opens the relay will drop out and give an alarm. To reset the alarm circuit, the alarm reset switch button must be pushed, thus connecting the relay coil across the power supply through S_5 and S_6. If, while this button is depressed, the mechanical failure alarm lights do not extinguish, it is an indication that the air flow system itself is the cause of the trouble as either S_5 or S_6 must be open.

The commutator system which has just been described was developed by Anton Laboratories specially for this equipment. No commercially available devices could be used because of the very low rate of travel of the paper tape.

Performance and Other Unusual Features

A series of equipments were built along the lines described above—the main differences being in the total range of detector as has already been related. These equipments were subjected to very complete valuation tests and were found to perform well within the target requirements. A final test performed at the Naval Research Laboratories wherein the detectors were actually tested in an atmosphere containing known concentrations of sodium-24 revealed that the equipments were accurate to within 5% of true dosage rate.

Among unusual features incorporated into these equipments and which have not been described are:

Self calibration: Throughout the design of these equipments care has been taken to devise circuitry which is dependent to a minimum on those characteristics of tubes and other components which are affected by aging, operation, etc. In spite of all precautions it can be assumed that some change in the calibration may take place with time. In order to check the entire system under actual operating conditions a Ra D + E calibrated beta source is incorporated into the equipment both for determining that the equipment is in operating condition and also for adjusting the calibration to its original condition. Tests wherein the calibration was purposely upset have been performed and the results have shown that the calibration can be readjusted to within a few per cent of the original value over the entire scale. This latter feature is made possible through the use of the Anton Adjustable voltage regulator tube which supplies the voltage to the integrator tube. A typical example of how this adjustment of the calibration is accomplished follows: A radium D + E calibrating source is positioned in the calibrating block provided. When the source is in this position, the meter reading should be for example, 12.5×10^{-7} microcurie/cm^3. If, after the vacuum tube voltmeter is zeroed and the sensitivity checked, the meter reading is different from 12.5×10^{-7} microcurie/cm^3, the anode supply voltage of the integrator tube is adjusted by means of the screw driver adjustment on V_8, the 6109, until the meter reading is 12.5×10^{-7} microcurie/cm^3.

Beta-gamma discrimination: A small metal slide has been provided in the lead shield which can be inserted between the mica window of the detector and the filter paper tube. By use of this slide beta-gamma discrimination is possible.

Immediate alarm shut-off: Once an alarm has been sounded because of a high concentration of radioactivity, it is impossible to reset the alarm until the contaminated filter paper has been removed

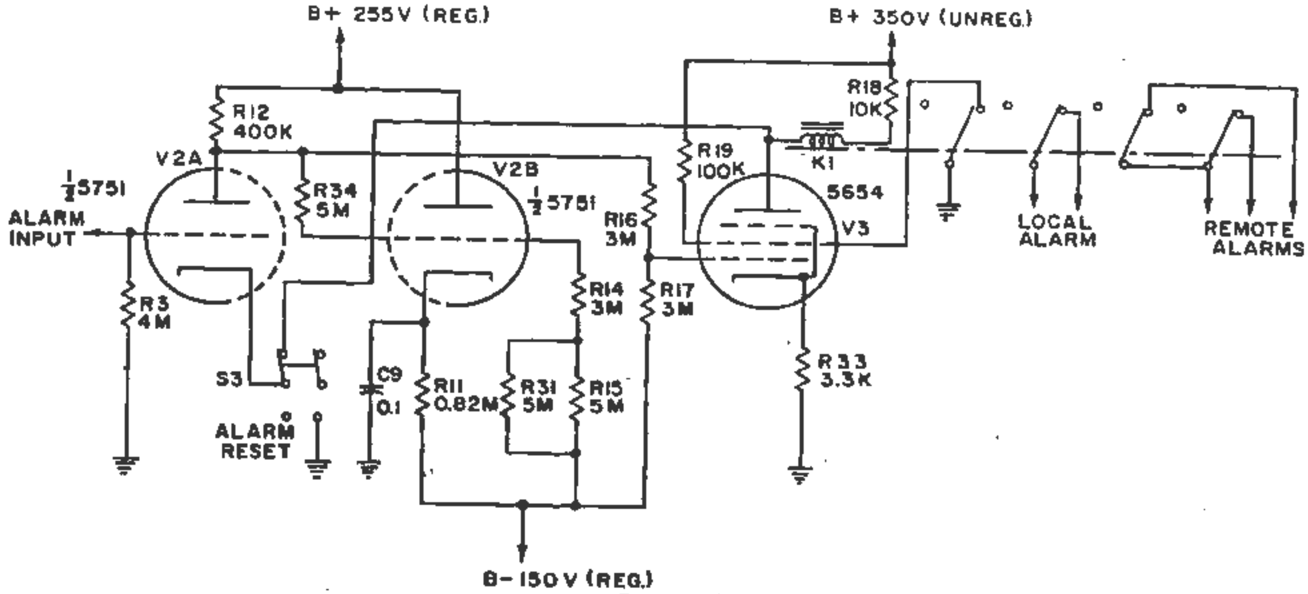


Figure 17. Alarm circuit

from in front of the detector tube. This may require quite a length of time because of the low rate of travel of the tape. Some of the equipments described in the literature were very annoying to the laboratory personnel because it often required hours of waiting or breaking the tape in order to shut off the alarm. In the Anton equipments a paper release switch has been incorporated to make it possible to move contaminated sections of paper through without disruption or delay.

VII. AIR PARTICLE MONITORS FOR OTHER ISOTOPES

The maximum permissible concentrations of the common radioisotopes in air for continuous exposure have been tabulated by the National Bureau of Standards in Handbook 52. The tolerance dose for natural uranium, for example, is as low as 1.7×10^{-11} microcuries per milliliter of air, which requires an increase in sensitivity of a factor of 10^4 over the equipment which has been discussed above. Equipment similar to that just described has been extended by even a greater factor in several ways:

1. An air flow system of greater capacity has been utilized. Larger air ducts have been combined with a wider filter paper tape. A new detector similar to the Type 1007 (see Section IX) but which has greater sensitivity as well as a larger mica window has been employed. The sampling time has also been increased in order to collect a greater amount of activity on the filter paper.
2. Where the isotope to be detected contains an appreciable component of gamma radiation and greater sensitivity is required, a sodium iodide thallium activated crystal and a photomultiplier have been utilized successfully.
3. Multichannel equipments have been built wherein the outputs of the channels are combined prior to amplification.

This method has been utilized to extend the scope of the airborne particle monitors to the analysis of the energy spectra of the beta and alpha activities deposited on the filter paper. The filter paper is allowed to pass consecutively in front of a number of detectors which are chosen and adjusted with absorbers to measure only a specific and discreet activity and energy range band. The output of each channel is "weighted" electronically in accordance with a prearranged scheme by a computor and the final output is utilized to sound alarms or perform automatically certain required tasks.

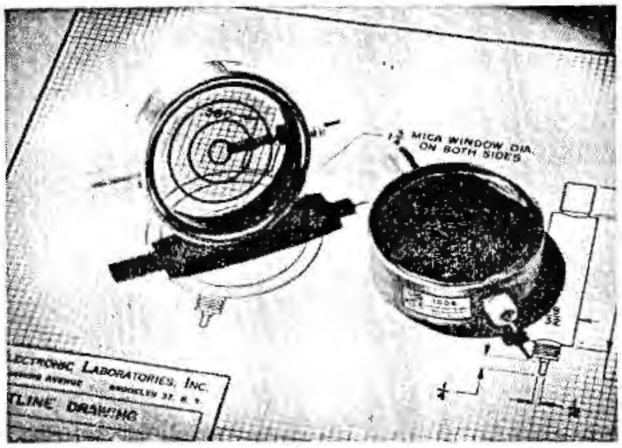


Figure 18. Double end mica window "pancake" type detector for use with coincidence air particle detector systems

Patent No. 2649554 entitled "Radiation Indicator" has been granted to the writer covering a multiple beta detector suitable to the determination of beta spectra. If the activity on the paper would be caused to pass in front of this detector the spectra could be automatically recorded.

VIII. DIRECT AND COINCIDENCE SYSTEMS

A fairly recent counter tube development makes possible construction of equipment for determining the radioactive condition of air without the collection of airborne particles *per se*. This tube, identified as the Anton Type 1008 "pancake" type detector is shown in Fig. 18. As can be seen from the illustration, the tube has the form of a round pillbox approximately two inches in diameter and one half inch deep. The thin windows on each large face are made of metallized mica which assists in creating a very uniform electric field within the counter. The tube responds to alpha or beta particles and has a low gamma efficiency of 0.2% for cobalt-60 radiation. The direct method can be best utilized for the determination of beta emitting isotopes such as Na^{24} and Ta^{182}.

As an example we will discuss the determination of the concentration of Na^{24} particles in air for which the tolerance dosage is 2×10^{-7} microcuries per cubic centimeter. The maximum energy of the beta-spectrum of Na^{24} is 1.390 Mev. The average energy (kinetic plus rest energy) of the emitted beta-particles, E_{avg} in units of m_0c^2, can be calculated for a known distribution $P(E)dE$ by

$$E_{avg} = \frac{\int_1^{E_0} EP(E)dE}{\int_1^{E_0} P(E)dE} \qquad \text{(IV)}$$

For an allowed transition, the distribution $P(E)dE$ is given as

$$P(E)dE = \left(\frac{G^2}{2\pi^3}\right)\frac{m_0c^3}{h}|M|^2F(Z,E)\ (E_0 - E)^2 (E^2 - 1)^{1/2}EdE \qquad \text{(V)}$$

where E_0 is the maximum total energy in the relativistic units.

By substituting equation V into IV one obtains

$$E_{avg} \cong 0.542 \text{ Mev} \qquad \text{(VI)}$$

The range of an electron of 0.542 Mev is approximately 180 mg/cm² or 200 cm in air which indicates that ample space should be reserved unobstructed around the G-M counter in order to detect as many beta-particles as possible.

Consider the 1008 counter supported in such a way that it can see on each side of its thin mica windows 2 meters of free space, then if for simplicity sake the dimensions of the tube were as given in Fig. 19, the estimated β-counts would be

$$N_\beta =$$

$$\frac{2 \cdot 10^{-7} \cdot 3.7 \cdot 10^{4}}{\text{disintegration/cm}^3} \cdot \int_0^{200} \underbrace{4\pi r^2 dr}_{\substack{\text{Volume of a}\\ \text{spherical shell}\\ \text{of r to r + dr}}} \cdot \underbrace{\frac{\pi(2.5)^2}{4\pi r^2}}_{\substack{\text{Solid angle}\\ \text{subtended}\\ \text{by the}\\ \text{counter}}} \cdot \underbrace{\frac{1}{3}}_{\substack{\text{Correction}\\ \text{for the}\\ \text{oblique}\\ \text{effect}}}$$

$$\therefore N_\beta = 7.4 \cdot 10^{-3} \cdot \frac{\pi(2.5)^2}{3} \cdot 200$$
$$= 7.4 \cdot 10^{-3} \cdot 1.3 \cdot 10^{3} \cong 10 \text{ cps}$$

It is evident from the above calculation that the counting rate of 10 cps is sufficiently high to permit us to determine the presence of the tolerance dose of Na^{24} if the background remains at a reasonably low value such as one would encounter in an establishment utilizing Na^{24} isotope for chemical purposes.

Other beta isotopes having tolerance doses ranging down to 1×10^{-8} μc/cm^3 can be similarly detected. Absolute determinations at these low levels are difficult. Experimental tests indicate that the order of magnitude of the above estimates are correct.

The situation changes rapidly when a variable gamma background (due to the proximity of the equipment to some high level machine) is present. If we assume that the gamma field can reach a value of say 60 mr/hr and possesses an average energy of 2 Mev the number of counts in the 1008 tube (which as was mentioned before has a gamma efficiency of 0.2%) will be approximately 200 counts per second.

$$N_\gamma = 3.7 \times 10^{10} \times \frac{\pi(2.5)^2}{4\pi(100)^2} \times 60 \times 10^{-3} \times 0.2 \times 10^{-2} \doteq 700 \text{ cps}$$

The above computation is based on the fact that one curie of gamma emitter produces approximately 1 r/hr at one meter.

One also has to consider the secondary electrons produced by the gamma radiation in the free air around the counter which will enter the tube and cause additional counts to be recorded. The beta efficiency of the 1008 is high and actual experimental data shows that the response of this tube in a 60 mr/hr field averages 1400 cps. The normal background of these tubes averages 31 cpm.

Obviously then the direct method can be used only in conditions where the gamma background does not exceed 3 or 4 times normal background.

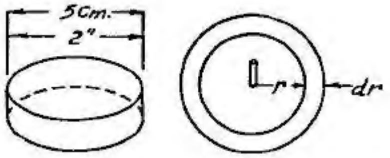


Figure 19

A differential method has been used where gamma backgrounds up to the maximum weekly gamma tolerance dose of 300 mr have been encountered. The differential system is a two channel system wherein the output count of a tube exactly equal in response to gamma radiation but insensitive to betas is subtracted from the beta plus gamma count of the 1008.

For the sake of continuity we will carry forward the measurement of tolerance dose of Na^{24} in the presence of a gamma field reaching maximum values of 1 mr/hr and 7 mr/hr.

The gamma count in the 1008 at 1 mr/hr is 1400 cpm. Assuming our required response time to be one minute then the most probable error is $0.67 \sqrt{1400} = \pm 25$ cpm. The addition of the most probable errors of the differential system are ± 50 cpm which will affect the beta tolerance component count by ± 12%. At 7 mr/hr the gamma count is 9800 cpm and under the same condition as above we obtain a most probable error of ± 65.5 cpm and ± 31% as the value of the error introduced in the beta count by the gamma background. At longer integration times or at higher than tolerance values of Na^{24} the error due to the variable gamma field will be proportionately lower.

The 1008 type tube makes possible another system by which relatively low intensities of beta radiations can be determined in the presence of variable levels of gammas: the coincidence system. Consider stacking 2, 3 or 4 tubes on each other with the large mica window faces parallel. The output pulses are connected into a coincidence circuit. This circuit has been designed and constructed so that it will record coincidences only when two pulses are separated by a period shorter than 1 microsecond. Each tube dead time has been artificially shortened to 8 microseconds by a polarity inverting circuit which makes the anode negative for a period of 5 microseconds, 3 microseconds after the initiation of a pulse.

In order to test the circuit the accidental coincidence of 2, 3 and 4 1008 tubes arranged in planar array and exposed to 60 mr/hr was determined and found to be respectively 1000, 10 and zero cpm. When the tubes were stacked with no space between the mica windows and exposed to the 60 mr/hr flux the triple coincidence response was 16,000 cpm and the quadruple 4000 cpm. It is evident that secondary betas originated by the action of the gamma radiation on the first and fourth tube walls were firing the coincidence system which was also counting the sec-

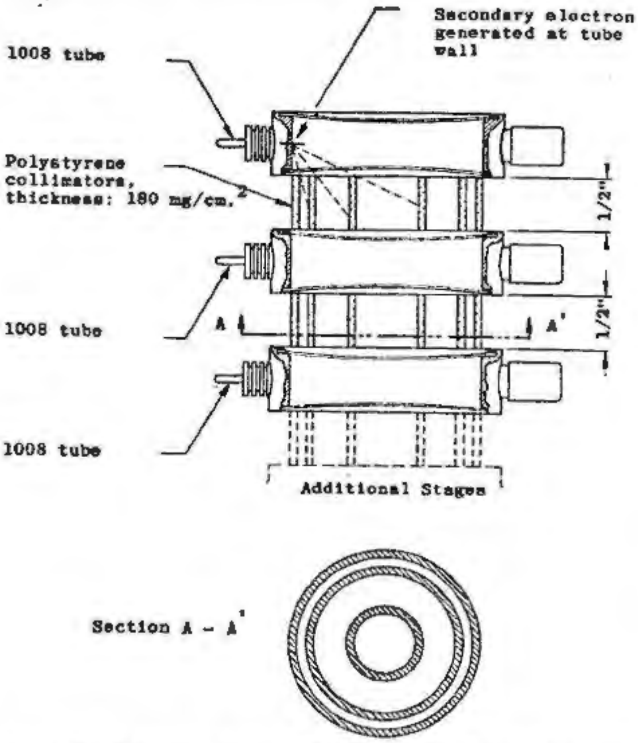


Figure 20. Schematic diagram illustrating use of polystyrene collimators with 1008 tubes to inhibit secondary electrons (generated at tube walls) from traveling in a straight line between active volumes of tubes

ondaries generated in the free space in such a direction as to pass through the four tube stack.

Increasing the air space between tubes reduced the accidental coincidence count considerably and achieved the following values at separations of 1½ inches. Double coincidence: 4000 cpm; triple: 2300 cpm; and quadruple coincidence: 300 cpm. By the addition of 3 collimators (as shown in Fig. 20) each constructed from 3 concentric cylinders of 180 mg/cm^2 polystyrene of suitable diameters to inhibit straight line travel between active volumes of the tubes for secondaries generated at the tube walls and reducing the spacing between tubes to 0.500 inches the accidental coincidences were further reduced by a factor of 2. Although no actual complete commercial model of an equipment was constructed utilizing the coincidences system it is evident from the above and experimental data obtained at Anton Laboratories that this principle can be utilized to measure tolerance doses of betas in the presence of gamma fields of higher intensities than with the other direct systems described above. The coincidence system depends for its operation on the fact that the energy of the betas must be high enough to permit them to enter the active volumes of the tubes. The window thickness of the 1008 is 3 to 5 mg/cm^2 and 7 windows do not usually exceed 30 mg/cm^2.

From the above it can be seen that systems which do not require the collection of activity on filter paper for measuring low concentrations of relatively high energy betas in air are useful if the simultaneous and variable gamma fields do not exceed values of 1 mr/hr for the direct method, 7 mr/hr for the differential method and 12 mr/hr for the coincident method.

IX. LABORATORY RADIOACTIVITY COMPARATOR

In Section V of this paper are described air particle detectors in which the activity is collected on discs of filter paper which are subsequently scanned by a measuring equipment of varying degrees of complexity. Anton Laboratories has perfected a simple laboratory radioactivity comparator which enables a relatively inexperienced laboratory technician to quickly and accurately evaluate the level of activity contained on these discs in terms of a laboratory sample whose activity is known to a high degree of precision. This equipment (Figs. 21, 22, 23) consists of a detector and a novel electronic integrator which is described below.

The particular equipment illustrated in Figs. 21, 22, and 23 has been adapted for comparing the activity of contaminated water with acceptable civil defense limits for drinking water. Such an equipment is useful in determining the degree of contamination in river water which is possibly being contaminated by waste products from a nuclear activity. The comparison standard is contained in the cover of a 4-ounce ointment tin—the unknown sample is contained in the lower half of the tin. A small jig is provided for accurately locating the level of both samples with respect to the counter tube window.

Detector

The detector utilized is an Anton Type 1007 which has been specially designed for the measurement of weak alpha and beta sources. This detector is illustrated in Fig. 24. The flat "pancake" shape of this detector offers the advantage of a large diameter window without the disadvantage of a large volume and resulting high background. The mica window is 1⅛ inches in diameter and is only 1.4 mg/cm^2 thick. Counter tubes with large mica windows have in the past found disfavor because the windows

Figure 21. Front view of radioactivity comparator showing probe in position for testing unknown sample

tended to become charged and thereby distorted the field and response of the tubes. To overcome this difficulty Anton has metallized the windows and added the center post to the cathode. This construction effectively grounds the window when the internal pressure of the tube is less than atmospheric pressure, for then the window is caused to bow in and rest against the center post creating a positive electrical contact. This construction also permits the use of even thinner windows than were hitherto possible. Versions of this tube have been built with their mica windows up to 6 inches in diameter.

The tube has a low dead time (approximately 20 μsec) and produces large amplitude pulses which obviate the need for a preamplifier thus simplifying the resultant equipment.

Electronic Integrator

The pulses produced by the tube trigger a monostable multivibrator. The multivibrator output pulse is fed through a cathode follower and a pulse-shaping network to an integrating capacitor. There an average voltage is built up on this capacitor by a succession of such pulses which are initiated in a random manner by the beta or gamma emanations from the radioactive sample. A pre-set electromechanical timer governs the interval if time over which the capacitor is permitted to accumulate charge and at the end of this interval it automatically switches a vacuum tube voltmeter into the circuit which reads the voltage across the capacitor. For a given counting interval this voltage will depend upon the level of activity of the sample. The capacitor itself has extremely low dielectric hysteresis so that it can be completely discharged for subsequent measurements within a very short interval.

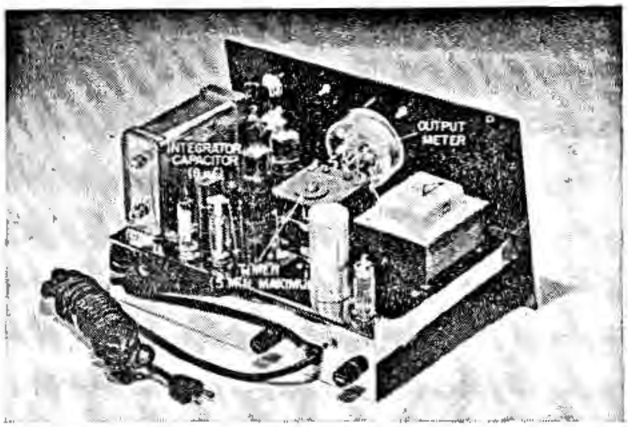


Figure 22. View of comparator chassis showing construction details

Once the vacuum-tube voltmeter has been standardized with a known source by means of the provided calibrating adjustment it will indicate the per cent relative activity of an unknown radioactive sample in terms of the standard. The presence of background does not appreciably affect the measurements because the vacuum tube voltmeter can be adjusted to read zero at background.

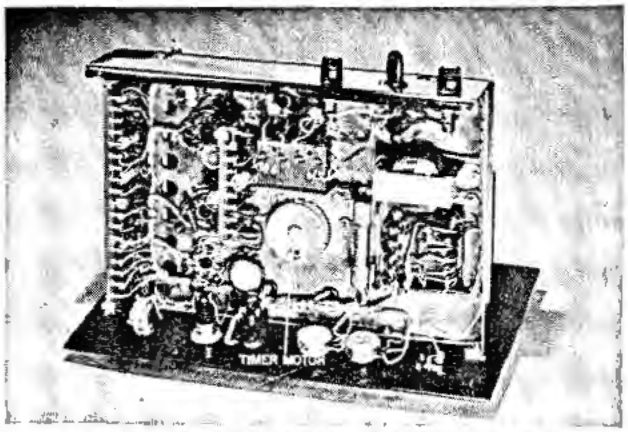


Figure 23. View of underside of comparator chassis

The integrator is packaged in a cabinet which is 14 inches wide × 10 inches deep × 9 inches high and weighs 25 pounds. The cabinet is fabricated of high polished stainless steel to facilitate decontamination. The equipment is rugged enough to withstand the shock and vibration which portable equipment can be expected to encounter.

Performance

The electronics of this equipment have been designed to insure stability and accuracy which is not attainable by other comparators. For example:

1. The sensitivity of the detector is great enough to produce adequate counts in a relatively short interval and thereby insure good statistics.

2. The large mica window provides large sampling area and by using two of these tubes—one on each side of the filter paper disc—almost 4π geometry can be attained.

3. The linearity of the scale permits the user to set the meter pointer at any position of the scale when the detector is exposed to a standard one millimicrocurie source. The timer in the comparator allows adjustment of the integrating time up to 5 minutes. When one combines this time variable with the time of sample collection of the radioactivity in the filter paper disc, one can see that the range of the comparator can be extended to cover the tolerance doses for ingestion of isotopes.

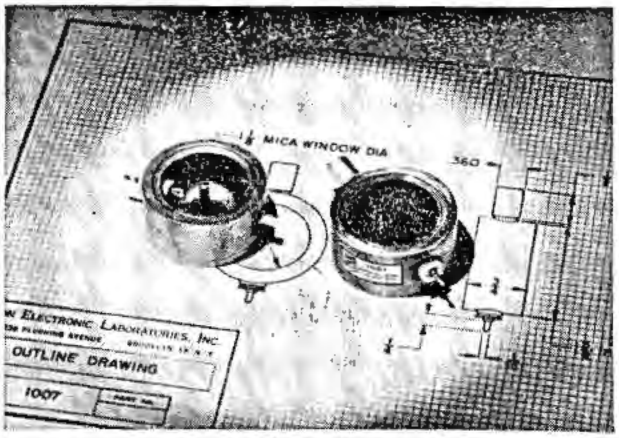


Figure 24

One of the most useful applications of this comparator in the laboratory is for quickly sorting a great number of samples to cull out those whose activity is within a specified range. When used with a set of filters this comparator measures not only the number of events but can also yield measurements which reflect the nature of the events as well.

ACKNOWLEDGEMENTS

The author is indebted to M. Youdin, Drs. J. H. Mulligan and S. Z. Lewin of our staff for their valuable advice and stimulating discussions concerning many phases of the topics treated.

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Combination of a Spark or Geiger-Müller Counter with a Photomultiplier Tube for the Detection of Nuclear Radiations

By A. M. Baptista and A. J. G. Ramalho,* Portugal

In 1945, Chang and Rosenblum[1] presented a type of radiation detector based on electrical spark discharge, where an ionizing particle induces considerable specific ionization between a wire and a plate after a potential difference of some thousands of volts has been established between them.

Many studies were carried out after them on the properties and applications of the detectors, resting on the same principle. But, recently, Blanc[2] brought the whole matter into focus.

All these detectors, in so far as we know, operate under such conditions that, following the application of a potential difference, a corona effect is produced, which creates a current through the space between the electrodes.

Under those conditions, when there passes an ionizing particle which induces large specific ionization between the electrodes, a spark is created. The potential variation of one of the electrodes, as brought about by the spark, is what is actually detected by means of the conventional counting devices.

Under such operating conditions, we can state that the only particles detected, with a substantial counting efficiency, have been those ionizing particles which have considerable ionizing power, such as alpha particles or fission products.

COMBINATION OF A SPARK DETECTOR WITH A PHOTOMULTIPLIER

The first experimental results obtained with devices similar, up to a point, to those used before, but for which the operating voltage is much lower than that necessary to give the corona effect (electrical effluvium) under the conditions imposed by the experiment, will be presented.

The passage of an ionizing particle could create a spark (a spark, in this paper, is taken to mean a brief intermittent luminous phenomenon associated with an electrical discharge between two points—following the creation of a potential difference) without bringing about the corona phenomenon.

We now propose to detect this spark, not, as was done so far, by using the potential variations of one of the electrodes, which would be extremely difficult under our operating conditions, but rather directly, by means of a photomultiplier which "sees" the photons emitted by the spark and transforms the luminous excitation into electrical impulses which are counted following amplification.

Figure 1 shows the counting device which we used. It consists of the spark generator, placed in front of a photomultiplier in such a way that the light emitted during the discharge can easily reach the photocathode of the latter. The spark generator and dynodes are biased by stabilized voltage units.

The impulses at the output terminals of the photomultiplier (E.M.I. 6260, with an interdynode voltage of 155 v) are passed on, through a cathode follower to a linear amplifier (of the Harwell B 1049 type), which amplifies them. Thereafter, they go to an electronic counter (Harwell 1009 A type) which discriminates between them and counts them.

Figure 2 shows one of the spark generators used. A tungsten wire, 0.1 mm in diameter, is placed 1.5 mm away from a brass plate. The radioactive source faces it, 2 cm away.

Figure 3 shows the counting rate variation as a function of the voltage applied to the wire (positive with respect to plate) in the air, under a pressure of one atmosphere, for a source of Po^{210} (alpha-particle

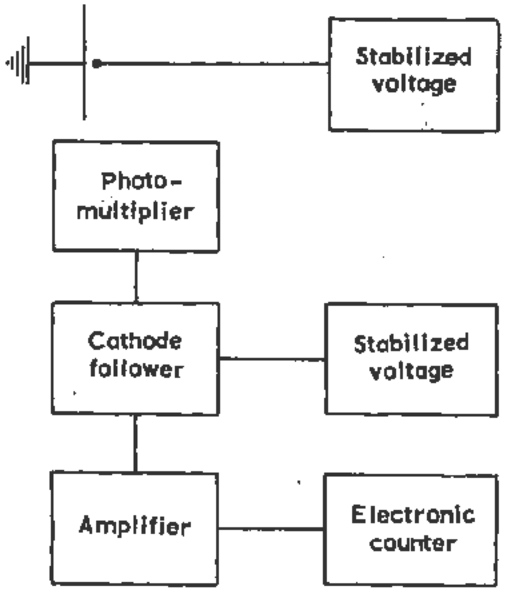

Figure 1. Block diagram of the equipment used

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* Centros de Estudos da Energia Nuclear, Laboratório de Física.

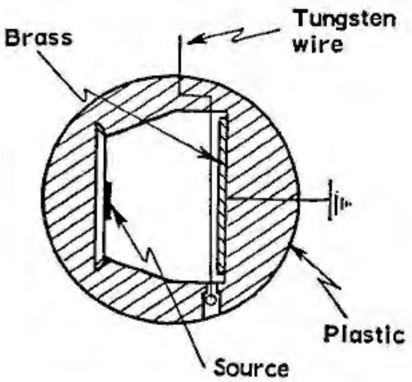

Figure 2. The spark detector used. The tungsten wire, which is biased with respect to the brass plate, has a diameter of 0.1 mm

energy 5.3 Mev). This particular range ends at a voltage lower than that which corresponds to the corona effect in the absence of a radioactive source.

Figure 4 shows a range obtained, under the same conditions, for the beta particles of Co^{60} (maximum energy, 0.3 Mev). This stops practically at a voltage level very close to that which corresponds to the corona effect in the absence of a radioactive source.

We can see, as an interesting result, the possibility of detecting beta particles with a substantial counting efficiency, for the radioactive source had an intensity of approximately 5×10^{-2} μc. It was circular and had a diameter of 5 mm. The curves on Figs. 3 and 4 also show that, with such a method of detection, it is possible to separate the alpha from the beta particles for, as was to be expected, in view of the difference in specific ionization between alpha particles from Po^{210} and beta particles from Co^{60}, the counting threshold for the alpha particles from Po^{210} is much lower than for beta.

A first approximate determination of the resolving time for the detector, when the resolving time of the electronic counter varies, gives a value under 500 μsec.

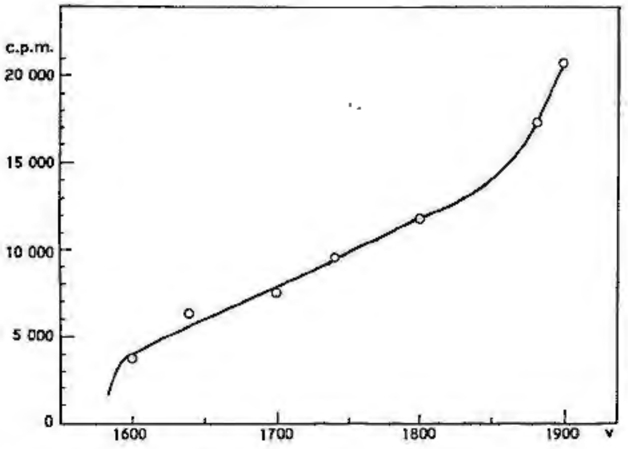

Figure 3. Counting rate variation curve as a function of the potential drop between the electrodes, for a source of p^{210}. Amplification system gain 757

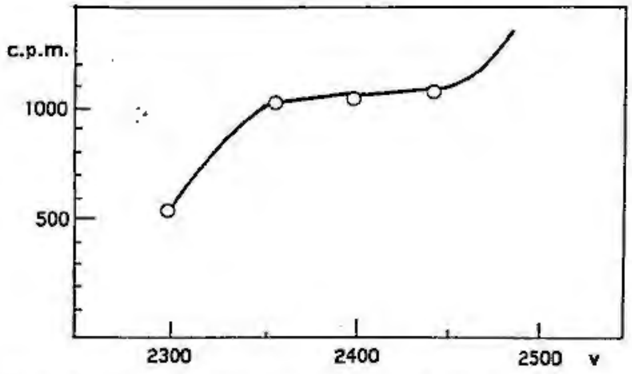

Figure 4. Plot of the variation in the counting rate as a function of the potential difference between the electrodes for a Co^{60} source of beta particles. Amplification system gain 757. Working conditions are the same as in Fig. 3

COMBINATION OF A GEIGER-MÜLLER COUNTER WITH A PHOTOMULTIPLIER

We have used another device which is made up of the combination of a Geiger-Müller counter (a spark generator in our case), placed on a suitable rest (Fig. 5) with a photomultiplier tube. The over-all scheme is the same as that shown in Fig. 1, but it must be completed by a device for counting the impulses in the Geiger-Müller tube. The window of the Geiger-Müller tube is exactly aligned with the photocathode and, 1.5 mm from the latter, the radioactive source, this time an emitter of alpha and beta particles, is placed on a transparent lucite plate, between the window of the Geiger tube and the photocathode, 1 cm from the latter. In the case of gamma radiation, we have used a collimated bundle from a Co^{60} source. The photomultiplier tube was protected by a 5 cm lead thickness.

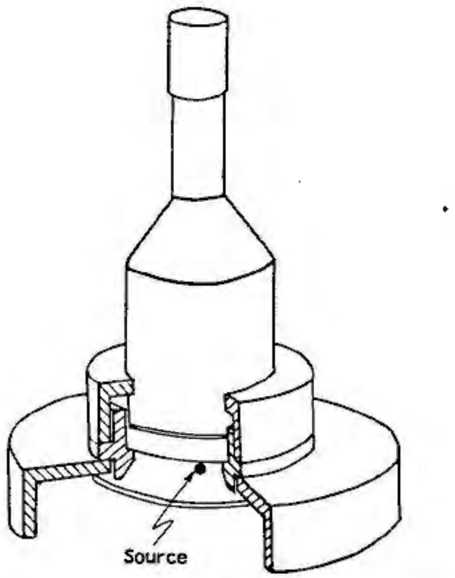

Figure 5. Geiger-Muller tube carrier, showing how the tube is placed, and the position of the radioactive source between the window of the Geiger tube and the photocathode of the photomultiplier tube

Figure 6 shows ranges, in the photomultiplier tube, which correspond to various degrees of discrimination, for a Co^{60} source, as well as the range traced at the same time by the conventional method for a Geiger tube (Phillips 18154 tube filled with argon, neon and a halogen). It will be seen that, as discrimination increases, the ranges become even longer and of lesser slope, so that, for a 50-v discrimination, the range obtained has the same length and slope as the Geiger tube but twice the sensitivity.

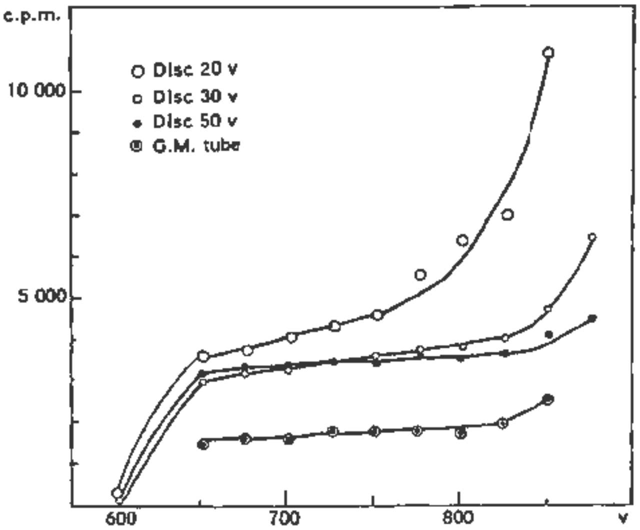

Figure 6. Curves showing the counting rate variation as a function of the voltage applied to the Geiger detector for a gamma ray source (Co^{60}). Amplification system gain 473. The conventional "plateau", or range, of the Geiger detector is indicated by the "G.M. tube" marks

Figure 7 shows the ranges obtained in the photomultiplier with a P^{32} source for various degrees of discrimination, as well as the Geiger-Müller tube range, obtained at the same time. The figure shows that, in a fashion similar to that obtained with gamma radiation, the ranges obtained with beta particles become ever longer and less inclined as the discrimination increases and that, when it is equal to, or greater than, 40 v, a range is obtained of the same size and inclination as that of the Geiger counter, but twice as sensitive.

Figure 8 shows the ranges obtained in the photomultiplier with alpha particles form a source of Po^{210} for two different values of discrimination (15 v and 25 v), and the Geiger tube range plotted at the same time. It will be seen that the two ranges obtained in the photomultiplier tube practically coincide, and that their length and slope are approximately the same as in the Geiger tube, while they start 75 v below and end up 50 v below those, also that the sensitivity is double, as in the case of the beta particles. We shall see, however, that the initial counting voltage, for the alpha particles from Po^{210}, is well under (about 300 v) that which corresponds to the range obtained with the Geiger tube used in the conventional manner.

The ranges obtained with the photomultiplier tube and shown on Figs. 6, 7 and 8, all have been plotted under the same conditions, with an amplification of 473 for all of them, and amplification of 0.064 for the cathode follower, and 7400 for the linear amplificator.

Figure 9 also shows ranges obtained in the photomultiplier tube with the same source of Po^{210} and the Geiger tube range which has been traced at the same time. We have worked, in both cases, with two different discriminations (5 v and 50 v), and with an amplification of 200. Figure 9 shows that the three ranges (two obtained with the photomultiplier tube and the third with the Geiger tube), practically coincide. However, the first two begin 75 v under the last and, in addition, in that working range, the "background" is nil. This shows that the ranges of the photomultiplier tube, aside from being longer than those of the Geiger tube, have an area of some 75 v in which background is eliminated. Let it also be noted that the initial counting voltage for the alpha particles, here again, is much lower than in the Geiger tube.

In each and every case, the amplification sensitivity at the input of the electronic counter was 0.7 v, and the resolving time 240 μsec for the conventional system and 200 μsec for the system combined with a photomultiplier.

The first results obtained with a window type detector filled with a mixture of alcohol under 1 cm Hg and argon under 9 cm Hg seem to indicate a different behavior: the sensitivity achieved is about 4 times that obtained with a detector used in the conventional manner.

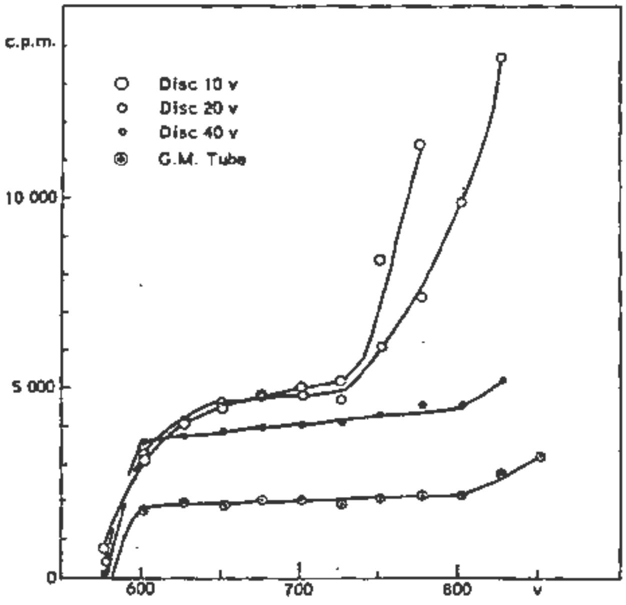

Figure 7. Curves showing the variation in the counting rate as a function of the voltage applied to the Geiger detector for a P^{32} beta source. Amplification system gain 473. The conventional Geiger "plateau" or range is identified by "G.M. tube"

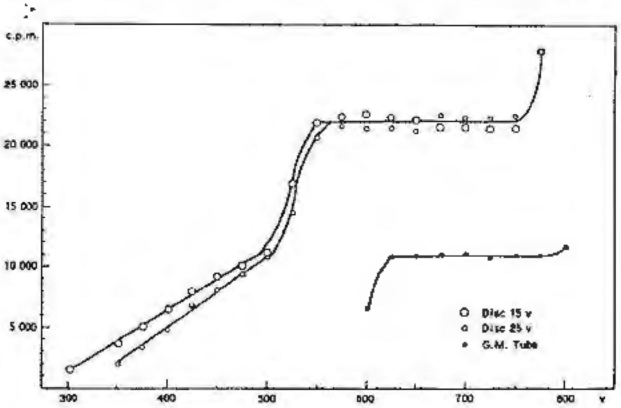

Figure 8. Curves showing the variation in the counting rate as a function of the voltage applied to the Geiger tube anode for a Po^{210} alpha source. Amplification system gain 473. The Geiger detector "plateau" or range is identified by "G.M. tube"

CONCLUSION

As will be seen from the results presented, a spark detector and Geiger counter, when combined with a photomultiplier, behave in similar fashion.

In both cases, alpha particles can be distinguished from beta particles. Figure 8 seems to show that, in the Geiger area, with a halogen type detector, the sensitivity increase for the detection gain is a consequence of the appearance of new luminous discharges of lesser intensity from a voltage of 500 v: these cannot be detected as the gain decreases (Fig. 9).

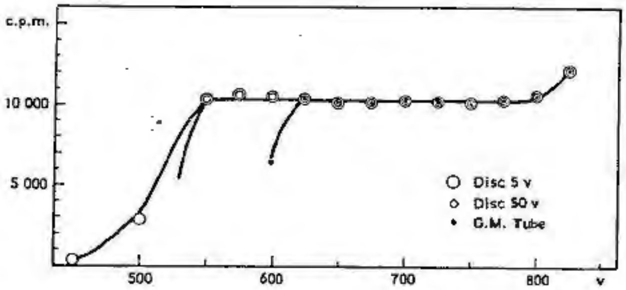

Figure 9. The same curves as on Fig. 8 (Po^{210}), but with an amplification system gain of 11.6

It is interesting to note that, for variable gains at the amplifier, ranges of different heights are obtained.

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1. Chang, W. Y. and Rosenblum, S., "*A Simple Counting System for Alpha Ray Spectra and the Energy Distribution of Po Alpha-Particles,*" Phys. Rev. *67*, 222 (1945).
2. Blanc, D., "*Le Fonctionnement Des Compteurs a Etincelles,*" J. Phy. Rad. *15*, 483 (1954).

Record of Proceedings of Session 20C

THURSDAY AFTERNOON, 18 AUGUST 1955

Chairman: Mr. E. Saeland (Norway)

Vice-Chairman: Mr. H. Hulubei (Romania)

Scientific Secretaries: Messrs. A. Milojevic and C. A. Sastre

PROGRAMME

20C.1. General aspects

P/150 Low-level counting and the future of isotopic tracers..... W. H. Johnston

DISCUSSION

20C.2. Dosimetry of gamma rays

P/386 Dosimetry of cobalt-60 gamma rays and thermal neutrons in heavy water piles.............................. J. Sutton *et al.*

P/70 Absolute dosimetry of cobalt-60 gamma rays........... J. S. Laughlin *et al.*

DISCUSSION

20C.3. Neutron dosimetry

P/154 Dosimetry of reactor radiations by calorimetric measurements .... D. M. Richardson *et al.*

P/63 Methods of measurements of neutron flux at low levels .... F. P. Cowan and J. O'Brien

P/60 A gamma insensitive semi-conductor fast neutron dosimeter using single crystal germanium........................ B. Cassen

DISCUSSION

20C.4. Counting and electronics

P/71 Dosimetry of ionizing particles............................... G. Failla

P/61 Recent developments in the scintillation counter field......... G. A. Morton

P/66 Methods of pulse analysis............................... G. G. Kelley

P/159 High resolution radiation detectors in research .... H. J. Gomberg and M. J. Schlesinger, Jr.

P/930 New Geiger tube design—hollow anode and parallel plate counters .... J. Hermsen *et al.*

DISCUSSION

The CHAIRMAN: This afternoon we are going to discuss the subject of radiation dosimetry. The term dosimetry is generally attributed to the theory and practice of determining radiation dosage either as energy flux or as energy absorbed in some medium. The art of dosimetry which we take over from the pioneers in the field of X-ray and natural radioactivity has become extremely important, but also very complicated in view of the great variety of radiations both as to quality and quantity which the advent of atomic energy, high energy physics and medical isotope applications have incurred.

Today we will therefore deal with dosimetry problems ranging from that of kilocuries cobalt sources and radiations in nuclear reactors down to the radiations hardly detectable from natural radioactive carbon. A great variety of physical and chemical processes which can be used for the determination of dose will be discussed.

It might be added that some of the papers allotted to this session are not directly related to dosimetry in the usual sense as they are concerned more with the detection and localization of radiations and radioactive substances than with a measurement of dose. This adds to the interest and variety of the topics to be dealt with this afternoon, but we must at the same time regret that so many of the outstanding papers allotted to this session cannot be presented and probably cannot be discussed during this meeting.

Mr. W. H. Johnston (USA) presented paper P/150.

DISCUSSION OF PAPER P/150

Mr. R. F. Chaudhari (Pakistan): With a view to improving the efficiency of detecting gamma radiation with a Geiger-Müller tube we have been investigating if it could be used in place of a photomultiplier to respond to light given out by phosphors. The work has been carried out with nickel cathode counters enclosed in Pyrex glass envelopes. A fresh Geiger tube with cathodes of high work function is normally not much photosensitive to wavelengths of 3300 Å and above. We have found, however, that it can be made photosensitive to longer wavelengths by passing in it a glow discharge at low pressure in pure alcohol or in an argon-alcohol mixture. This processing of the counter makes it photosensitive to all wavelengths up to 8000 Å and longer, but we have found that its photosensitivity to wavelengths over 5000 Å to 6000 Å is very short-lived. It lasts very much longer to band around 4000 Å. It is observed that the sensitivity power of a processed counter lies at about 3400 Å. The photosensitivity of the counter to the range of wavelengths 3400–4000 Å increases still more if it is baked first at 500–600°C under a high vacuum for a few hours and then a glow discharge is passed into it at low pressure before filling it with 10 per cent argon-alcohol mixture at 10 cm pressure. I may point out that the alcohol in the mixture is primarily responsible for the photosensitivity.

We have also observed no marked changes in the counter's photosensitivity for longer wavelengths by baking.

A photosensitive counter has been successfully used in detecting radiations from the ionisation and excitation of mercury atoms caused by the impact of positive ions of potassium of as low energies as 200 volts. In a similar way we have detected ionisation of mercury atoms by mercury positive ions of low energies. A counter processed in a manner described above, when placed against an NaI(Tl) crystal exposed to gamma radiations gives a greater number of counts than obtained with direct counting.

We have also made an attempt to detect neutrons with such Geiger counters by enclosing them in a flask whose interior is covered with a thin layer of paraffin wax and is filled with hydrogen at low pressure. The ionisation radiations of hydrogen caused by protons knocked out by incoming neutrons are detected by the Geiger counter.

Mr. Roucayrol (France): In his very interesting statement, Mr. Johnston seems to take the view that it is at present necessary to use two photomultipliers when counting low-level beta activity in a liquid medium by the scintillator technique. We, however, think it possible to use a single photomultiplier; in fact, Mr. Johnston suggested it himself in his paper. All that is necessary is to select a photomultiplier having an extremely low level of background noise. A further possible improvement is to coat the outside of the photomultiplier first with a dull black absorbent substance and then with a second, waterproof, coating. By using these two techniques we have achieved a 50 per cent reduction of the dark current at an operating temperature of 14°C. It will also help to pay particular attention to the optical coupling between the tank containing the scintillator and the photocathode; again, it is possible to work with a light trap so as to obviate the necessity of switching off the high-voltage supply every time the sample is changed.

By taking these precautions we have been able, with a sample-charger which we showed at the exhibition of the French Physics Society in June, to obtain an inherent activity in our system equivalent to the number of counts per minute furnished by one millimicrocurie of C^{14} dissolved in a solution of 2–5 diphenyl oxazole in toluene.

Detection by our system is 100 per cent efficient in the case of carbon-14 and sulphur-35, and we have carried out a number of absolute counts of activity which have just been published. In the case of tritium, on the other hand, we have not yet exceeded an efficiency of 30 per cent. Our apparatus is, however, quite appreciably simpler and cheaper than those commercially available, which contain two photomultipliers, and the system has proved entirely satisfactory for activities which are, perhaps, not of the same order of magnitude as those in which Mr. Johnston is interested.

Mr. Guéron (France) presented paper P/386 and made the following remarks: I wish to present a paper prepared at the Chemico-Physical Laboratory of the Commissariat à l'Energie Atomique by Mr. Sutton and by Mr. Draganic, a member of the Yugoslav Atomic Energy Commission. It concerns dosimetry in chemical studies carried out under radiation in piles.

I wish to add that those solutions are presenting us with interesting problems concerning the mechanism of radiolysis, as radiolysis is quite different for oxalic acid and for $C_2O_4^-$ ions. Results of the work undertaken on this problem will be published in the future. Those questions belong rather to sessions devoted to radiochemical reaction mechanism than to a dosimetry session.

Mr. J. S. Laughlin (USA) presented paper P/70.

DISCUSSION OF PAPERS P/386, P/70

Mr. Sevchenko (Byelorussian SSR): With reference to the interesting papers we have listened to here on the dosimetry of gamma rays and thermal neutrons, I should like to add a few comments on the luminiscent method of dosimetry.

At a session of the Academy of Sciences of the USSR on the peaceful uses of atomic energy, a dosimeter for gamma and beta rays, and also for the recording of thermal neutrons, was proposed by Dr. V. V. Antonov-Romanovski and his associates.

The principle of the method is that a phosphor obtained from an alkaline-earth sulfide and activated by traces of europium and samarium, readily absorbs the energy of gamma rays, alpha particles, X-rays, photons, and so forth. Such phosphors, acting as an accumulator, retain the energy so absorbed for a long time, but emit it speedily under the action of infra-red light.

A linear dependence of the luminous intensity of the phosphor on the dose of irradiation over a wide range was established by the author. It comprises 5–6 powers of 10, and is unapproachable by existing methods of dosimetry. For example, doses of gamma radiation ranging from 0.005 to 1000 roentgens have been recorded with a luminescent dosimeter.

For the recording of thermal neutrons the phosphor is placed in a filter-case of cadmium. A thin layer of cadmium almost completely absorbs the thermal neutrons. In the process the cadmium itself becomes a source of gamma radiation which excites the phosphor. The sensitivity to neutrons of such phosphors in the cadmium filter-case reaches the level of sensitivity to gamma radiation.

Since the action of gamma radiation on luminescent phosphors is equivalent to excitation by secondary electrons, it is obvious that luminescent phosphors are able to record beta radiations also.

A luminescent dosimeter consists of a cake of the aforesaid phosphor in compressed powder form with a diameter of 1.6 cm and a thickness of 1.5 mm, cemented into a glass tube which, in turn, is housed in a metal filter-case one side of which consists of layers of cadmium, lead and mica. Before being put into operation, the luminescent dosimeter is placed in a thermostat in which it completely emits the energy it has stored up, and after cooling off in the dark it is placed in the corresponding filter-case and then goes into an electrophotometer for the purposes of its work. After irradiation, the readings are made in a darkened booth with diffused red lighting. By varying the voltage on the photomultiplier it is possible to determine the necessary sensitivity of the indicating dial of the photometer. When the operation is completed, the ampule containing the phosphor is closed with a metal cover and transferred to the thermostat for complete exhaustion of its radiation.

We have found by experiment that no more than one minute is needed to take the readings and enter them in the log-book.

The method of measuring doses of radioactivity with the luminescent dosimeter is very simple and sensitive and takes little time. One laboratory assistant can take 200 readings in one working day and take care of 100 workers.

A detailed description of the device and its operation was printed in the paper by Antonov-Romanovski which appeared in the published proceedings of the session of the Academy of Sciences of the USSR on the peaceful uses of atomic energy which was held from 1 to 5 July 1955.

Mr. C. J. Hochanadel (USA) presented paper P/154.

Mr. F. P. Cowan (USA) presented paper P/63.

Mr. B. Cassen (USA) presented paper P/60.

DISCUSSION OF PAPERS P/154, P/63, AND P/60

Mr. Gomberg (USA): I have a question for Dr. Hochanadel. In Figure 2 of paper P/154 for the neutron flux spectrum it was shown that the neutron flux peaks at 2 Mev approximately. I understood that this was a graphite moderated reactor and I wondered where within the reactor this flux was measured.

Mr. Hochanadel (USA): The flux is measured very nearly at the center of the reactor—within a few feet of the center of the reactor.

Mr. Gomberg (USA): I was asking whether it was in close to the fuel because of the remarkably large number of fast neutrons.

Mr. Hochanadel (USA): I wish there were an expert on reactors here. Perhaps Dr. Snell can help me? This is an experimental tube removed vaguely from the top of the reactor. It is necessarily close to the fuel elements—actually in the center.

The Chairman: Are there any more questions relating to these three papers? If not I will take this opportunity also to comment on the paper presented by Mr. Hochanadel.

I would like to compliment Dr. Richardson and his co-workers for the good agreement they obtained between calculated pile neutron doses and experimental calorimetric values. This agreement is very comforting because work done at the Kjeller reactor and published in the Journal of Nuclear Energy used that sort of calculation for determination of dose of pile-irradiated seed material. We could not verify directly whether these calculations were correct but the biological effects of the pile neutrons, the dose of which had been determined by such calculations, agreed within 10 per cent with that of (d,n) produced fast neutrons, the dose of which was determined actinometrically by the ferrous system. This already indicated that such calculations were not too far off.

Otherwise, I have the same comment as the previous speakers. The neutron spectrum seems to be very rich in fast neutrons. According to our experiments there is a large deficiency in fast neutrons for the high energy part of the pile neutron spectrum say beyond 8 Mev, as compared with the fission neutron spectrum. I refer, however, to a heavy-water pile.

Are there any other questions? We have still some time on our schedule.

May I take the opportunity to ask Mr. Cowan about the cross section of 230 millibarns mentioned for the (n,p) reaction on sulphur? What neutron energy or energy spectrum does this cross section refer to?

Mr. COWAN (USA): I cannot give you the details on this without looking up the detailed information, but we tried to take the average value, discounting any peak that might occur in the low neutron energy region, and actually these threshold detectors are subject to considerable uncertainty because of lack of cross-section information in really high energy regions. The cosmotron has a spectrum extending up to 3-billion electron volts and I am sure it will be some time before we have the cross-section information really needed to ascertain these values and to check how accurate they really are.

Mr. G. FAILLA (USA) presented paper P/71.

Mr. G. A. MORTON (USA) presented paper P/61.

Mr. SNELL (USA) presented paper P/66 as follows: The approach to this subject has developed simultaneously along several different lines. The simplest and cheapest analyzers make use of the deflection of the electron beam in a cathode ray tube, the amount of deflection being determined by the size of the voltage pulse that is applied to the deflecting plates. Recording is accomplished photographically, either on a moving film or on stationary film using a "gray wedge" that renders the pulse heights proportionally with the logarithm of their intensity. If the oscilloscope has been used with vertical sweep and horizontal displacement proportional to pulse amplitude, there results a sharply-defined envelope of the peaks of the spectrum, displayed in the conventional orientation. Devices of this kind all have the advantage of yielding a permanent record, but some experimenters believe that they have undesirable limitations in accuracy, and in the fact that there is no "instantaneous readout" that can be examined as the readings are being taken.

A second class of analyzers depends upon the conversion, typically by the use of a saw-tooth wave, of pulse amplitude information into time interval information. This principle is successfully used in the well-known Hutchinson-Scarrott pulse height analyzer. In this machine the storage of information makes use of pulse circulation in an acoustic delay line. The delay line can be made of either mercury or quartz. The stored information is repeatedly read out and displayed on an oscilloscope screen, so the spectrum dramatically builds up before your eyes as a semi-logarithmic array of dots. There is an advantage in simplicity in having only one storage device to serve perhaps 100 pulse height channels, but this advantage is purchased at the price of speed, because any one channel has access to the storage unit only once in the acoustic circulation time, which is 1200 microseconds.

The requirement of speed in a multi-channel pulse height analyzer perhaps deserves some comment. Speed is needed not so much for the purpose of recording data quickly as to prevent pulses in an intense part of the spectrum from choking up the analyzer so much that weaker details never get a chance to reveal themselves. Of course there are also special situations that demand speed in the analyzer, for example, when pulsed ionization sources are used, or when radiation of short half-life is being examined, so that a wide range in intensity is experienced perhaps within the course of one second.

With these considerations in mind Mr. Kelley and his collaborator Dr. P. R. Bell have adopted the philosophy that speed is very desirable in a pulse height analyzer provided that it can be obtained without excessive cost in dollars, in circuit reliability, or in accuracy. In seeking both accuracy and speed, Kelley has fallen back on the old principle of comparing the pulse height with a series of discriminators successively biased to higher and higher voltages. Let me review the principle with the modifications Kelley has introduced.

I shall try to indicate how the pulse height analyzer works.

The various discriminators are set at successive levels, perhaps 3 volts apart. An incoming pulse may have an amplitude that allows it to pass, say, the 15th discriminator, but not the 16th. They are spaced about 3 volts apart. The pulse has been previously lengthened and it has been inspected by an auxiliary circuit that would have rejected it if it had been confused with another pulse coming 2.3 microseconds before or afterwards. As the pulse is presented to the bank of discriminators, a so-called "interrogate pulse" is briefly flashed to all channels, and the circuits require both a coincidence between the incoming pulse and the interrogation pulse, and an anticoincidence between one of the discriminator levels and the other one, immediately above it. This can only happen at the top of the pulse, in discriminator 15 in our example, with the result that the interrogation pulse is passed on to a scaler attached to channel 15.

This arrangement demands one registering scaler per discriminator level, so for a 120-channel analyzer, 120 scalers would be required. One therefore is faced with a problem of expense. The problem can be met in two ways, first by providing an adjustment on the preceding window amplifier so that one can look at the 120 channels not all at once but in blocks of 20 at a time, so that only 20 discriminators and scalers are needed, and secondly by reducing the cost of the individual scaling units. By miniaturization of the scaling units, Mr. Kelley's colleagues, Messrs. Bell and C. C. Harris, have effected great economies.

I have here a plastic box containing one of their complete scale-of-two circuit elements, including the indicating lamp. Four of these side by side, comprise one of the scale-of-sixteen units built into the analyzer, and together they consume less than one watt. They feed into a commercial mechanical register that has been speeded up so as to be able to handle 75 impulses per second.

Slide 1 (Fig. 1 of paper P/66) shows the older complete 120-channel analyzer, taking 20 at a time, called the MC-3; this is commercially available in the United States.

Slide 2 (Fig. 2 of paper P/66) shows a recent modification, called the MC-4, which incorporates the miniature scalers and other refinements and which is just now coming into experimental use. Like the MC-3, the whole analyzer is contained in a single standard relay cabinet.

Table 1 (bottom of this page) compares some of the objective characteristics of a few different analyzers. In the table you see first of all that the beam deflection analyzers are relatively cheap and that they will be adequate if you do not mind waiting for your answer and if you are satisfied with moderate accuracy in the relative intensities of various parts of the spectrum. Although they are analogous devices, they can effectively be said to embody a finite number of channels because of such factors as the size of the beam spot that is photographed; the number of channels is here taken as about 100. Of the four examples of pulse height-to-time conversion, the original one developed by Wilkinson was so slow that it will henceforth probably be mostly of historical interest. Somewhat faster pulse height-to-time conversion analyzers are under development at Los Alamos and at the Argonne National Laboratory. Finally, we see Kelley's fast and accurate analyzers—MC-3, MC-4 and MC-5. The MC-5 is the same as the MC-4 except that it will register in all 120 channels at once. It is about half finished and is being built at Oak Ridge. Note how the introduction of the miniature scalers has reduced the price of the MC-4 as compared with the MC-3 from $400 per channel to $225 per channel, or from $8000 total price to $4500.

The things that are difficult to show in a table of this kind are matters of susceptibility to failure, diagnosis of faults, and their ease of indication, location and repair. Mr. Kelley has given attention to those factors, and he believes that in the MC-3, 4, and 5, a man reasonably experienced in electronics can diagnose, isolate and repair faults with comparative ease.

In closing, let me say that in this presentation I have stressed Mr. Kelley's contribution more than he himself has done in his own script of the paper. Many aspects of this subject are still controversial, and there is plenty of opportunity for experimenters to choose the kinds of circuit particularly suited to their needs.

Mr. H. J. GOMBERG (USA) presented paper P/159.

Mr. VAN DUUREN (Netherlands) presented paper P/930.

DISCUSSION OF PAPERS P/71, P/61, P/66, P/159, AND P/930

Mr. SNELL (USA): Speaking of multi-channel pulse-height analyzers, I should have remarked that the Los Alamos analyzer is on display in the United States Exposition in this building. It is actually in an operating state and is quite an impressive machine to look at.

The CHAIRMAN: May I ask Mr. van Duuren about the pressure of the filling gas of the flat parallel counter he showed.

Mr. VAN DUUREN (Netherlands): One hundred millimeters. The gas is a halogen neon-argon mixture of conventional type—10 centimeters mercury.

Mr. CHAUDHARI (Pakistan): May I ask Dr. van Duuren to tell me what is the proportion of neon

TABLE I. 1955 Comparison of Types of Multi-Channel Analyzers

Type	*Example*	*Speed*	*Number of channels*	*Accuracy*		*Time to get answer*	*Approximate cost*	*Cost per channel*
				Intensity	*Energy*			
Photographic beam deflection	Dots on film	Fast	Effectively $\sim$100	Fair	Good	2¼ hr	$2000–3000	$20–30
	Gray wedge	Fast	Effectively $\sim$100	5%*	$\sim$1%	1 min. to 30 min.†	$2000	$20
Pulse height to time conversion	Wilkinson	0.05 sec per pulse	100	Fair	Good	Instantaneous	Probably < $3000	$30
	Hutchenson-Scarrot (Emmer)	Average 600 μsec. per pulse	100	Good	Good	Instantaneous	$9000	$90
	Los Alamos	220 μsec per pulse	100	Good	Good	Instantaneous	$16,000	$160
	Argonne	Average 80 μsec per pulse	256	Fair	Good	Instantaneous	?	?
Multiple discriminators	MC-3	2.5 μsec	20(120)	1%	1%	Instantaneous	$8000	$400
	MC-4	2.5 μsec	20(120)	1%	1%	Instantaneous	$4500	$225
	MC-5	2.5 μsec	120	1%	1%	Instantaneous	$15,000	$125

* With densitometer analysis.

† With very crude intensity values.

and argon in the mixture in his flat counter and hollow anode counter? Also I would like to know the comparative value of the deadtime and the recovery time of the hollow anode counter as compared with the thin wire counter.

Mr. VAN DUUREN (Netherlands): The noble gas mixture is neon-argon with about 1 per cent of argon. The function of the argon is not quite critical. It is common opinion that it could be as well omitted as not. Your second question I did not quite get?

Mr. CHAUDHARI (Pakistan): Would you kindly tell me the comparative value of the deadtime and recovery of these counters as compared to the deadtime and recovery time in thin wire counters?

Mr. VAN DUUREN (Netherlands): This is quite difficult to compare, of course, because the deadtime and recovery time depend also on the gas fillings: but roughly, if one compares the deadtime of this parallel plate counter with a counter with the same window diameter, then it may be a factor of about four or five.

The CHAIRMAN: Are there any more questions or comments regarding the last five papers given?

Mr. SNELL (USA): Dr. van Duuren's paper reminds me of a counter that was built some years ago by Dr. Robley Evans, of the Massachusetts Institute of Technology. This counter had the anode in the shape of a spoon and the cathode in the shape of a fork. It would count, but I do not recall that Dr. Evans investigated the characteristics and showed that useful geometries can be made, such as Dr. van Duuren has done.

PEACEFUL USES OF ATOMIC ENERGY:

Proceedings of the International Conference in Geneva, August 1955.

The following is a complete listing of the sixteen volumes which comprise this publication, together with the main subjects covered by each volume.

VOLUME 1

The World's Requirements for Energy: The Role of Nuclear Power

World Energy Needs
Capital Investment Required for Nuclear Energy
The Role of Thorium
The Role of Nuclear Power in the Next 50 Years
Education and Training of Personnel in Nuclear Energy
Economics of Nuclear Power

VOLUME 2

Physics, Research Reactors

Special Topics in Nuclear Physics
Fission Physics
Research Reactors
Research Reactors and Descriptions

VOLUME 3

Power Reactors

Experience with Nuclear Power Plants
Fuel Cycles
Design of Reactors for Power Production
Power Reactors, Prototypes

VOLUME 4

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Equipment and Techniques Used in Measuring Cross-Sections Important to Reactor Design
Cross-Sections of Non-Fissionable Materials: Delayed Neutrons
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Effects of Radiation on Reactor Materials
Effects of Radiation on Liquids
Effects of Radiation on Solids

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Production of Metallic Uranium and Thorium
Analytical Methods in Raw Material Production
Production Technology of Special Materials

VOLUME 9

Reactor Technology and Chemical Processing

Waste Disposal Problems
Metallurgy of Thorium, Uranium and their Alloys
Fabrication of Fuel Elements
Liquid Metal Technology
Chemical Aspects of Nuclear Reactors
Chemical Processing of Irradiated Fuel Elements
Waste Treatment and Disposal
Separation and Storage of Fission Products

VOLUME 10

Radioactive Isotopes and Nuclear Radiations in Medicine

Isotopes in Medicine and Biology
Therapy
Diagnosis and Studies of Disease

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