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TECHNICAL PROGRESS REVIEW

ISOTOPES AND RADIATION TECHNOLOGY

VOLUME 1, NO. 1

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DRAFT

TECHNICAL PROGRESS REVIEW
ISOTOPES AND RADIATION TECHNOLOGY
VOLUME 1, NO. 1

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DATE ISSUED

JUL - 3 1963

Prepared for U. S. Atomic Energy Commission
by
Isotopes Development Center

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee
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for the
U. S. ATOMIC ENERGY COMMISSION

[redacted]

.....The uses of radioisotopes as a whole give these by-products of the liberation of atomic energy a general importance that is greater for the real progress of our civilization than the production of atomic energy itself.....

Professor Perrin

French High Commissioner
for Atomic Energy

A new Quarterly Technical Progress Review, "Isotopes and Radiation Technolgoy," was approved by the U. S. Atomic Energy Commission in the summer of 1962 and its preparation was assigned to ORNL as a part of the general program of the Isotopes Development Center. The assembly of a staff began almost immediately. By early fall, under the auspices of the AEC's Divisions of Technical Information and Isotopes Development, the preliminary drafts which are now the first issue of this periodical were being developed. "Isotopes and Radiation Technology" is the fifth of a series of Commission-sponsored quarterly technical progress reviews which cover broadly the field of nuclear science and technology. The others are Nuclear Safety, Power Reactor Technology, Reactor Materials, and Reactor Fuel Processing.

This new review will emphasize the reporting of DID-sponsored research and development. By publicizing present applications, it can help to expand the utilization of isotopes; and by helping the Commission and contractors to keep abreast of related activities within the AEC complex it can help prevent overlap of research effort and can aid in predicting isotope usage so as to point the direction for future isotope production efforts.

"Isotopes and Radiation Technology" is a technical progress review. Even guest articles will be of a review nature.* The editors and the AEC have no intention of duplicating the present efforts of commercial publishers of abstract journals and periodicals covering original research, and overlapping will be avoided. Books, monographs, and bibliographies that are considered pertinent to the areas routinely covered will be noted and occasionally reviewed.

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*e.g., see report by G. H. Cartledge on technetium in this issue, p. 3.

The ultimate success of the review will, of course, depend upon its value to the readers; a little time and experience will be needed to evaluate this. Therefore, we shall make no attempt to fix the format of the quarterly at the present time, and the editors will be counting heavily on reader criticism and suggestions to help them out.

TECHNICAL PROGRESS REVIEW

Isotopes and Radiation Technology

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FEATURE ARTICLE

Recent publicity[1] has pointed out that one of the most interesting properties of technetium is the "anticorrosive" characteristic of its pertechnetate ion, TcO_4^-. G. H. Cartledge of ORNL, who has pioneered in this field, has written the accompanying article to bring us up to date on the status of the theories behind this particular use of technetium. The timeliness of such a report is emphasized by the fact that the slow burning (rusting) of iron and steel alone costs the U. S. more per year than all the faster burning forest, factory, and city fires put together.[1a]

Technetium was not discovered until 1937, although many investigators, starting with Osann and his "polinium" in 1828, claimed to have isolated it.[2] However, in 1937 Perrier and Segré irradiated molybdenum in a cyclotron and reported Tc^{95m} and Tc^{97m} with half-lives of 80 and 90 days, respectively.[3] More recently Tc^{99}, with a half-life of 2×10^5 years, has been isolated from fission-product wastes. A few years ago it was priced at from $2800 to $1600 per gram, depending upon quantity, but can now be purchased from the Isotopes Sales Department of ORNL for less than $100 per gram.

Other applications of technetium include its use in superconductors,[4] and it is being considered for catalysis, as an anticorrosive agent in high-pressure boiling water reactors, in semiconductors, and as a source of low-energy radiation for radiography.*

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*See p. for the report on Tc^{99} sources.

INHIBITION OF CORROSION BY THE PERTECHNETATE ION

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Chemistry Division
Oak Ridge National Laboratory

Experiments have demonstrated that very effective inhibition of the corrosion of iron and steel can be achieved with low concentrations of pertechnetates under suitable conditions. Whereas there may be special circumstances in which the inhibitor may find practical application, its radioactivity and cost must necessarily restrict its widespread use. There is need, however, for further investigation of possibilities of using it in protective coatings or similar applications, and also of its relation to the corrosion of nonferrous metals. The distinctive properties of technetium compounds make the element an excellent new research tool and differentiate it sharply from chromium, which forms the inorganic compounds so extensively used as inhibitors.

Properties of Technetium Compounds Related to Inhibition

A comprehensive review of the chemistry of technetium has been published recently,[5] but the following comparison with the chemistry of chromium will suffice to illustrate the differences that are significant for studies in inhibition.

The important valence states of chromium and technetium are Cr(VI), Cr(III), Tc(VII), and Tc(IV), respectively. The bivalent chromate ion has basic and buffering properties:

$$HCrO_4^- \rightleftharpoons H^+ + CrO_4^= \quad (\underline{K} = 3.0 \times 10^{-7}),$$

whereas the univalent pertechnetate ion derives from a strong acid and lacks buffer capacity. When used at 10^{-3} f concentration and pH 6.0,

the respective reversible electrode potentials are:

$$CrO_4^{=} + 5H^+ + 3e^- \rightarrow Cr(OH)_{3(ppt)} + H_2O;\ \underline{E}_h = 0.59 \text{ volt} \qquad (2)$$

and

$$TcO_4^- + 4H^+ + 3e^- \rightarrow Tc(OH)_{4(ppt)};\ \underline{E}_h = 0.20 \text{ volt.} \qquad (3)$$

Polarization measurements at a passive iron electrode indicate that the exchange current density of the pertechnetate electrode under the conditions specified above is several orders of magnitude greater than that of the chromate electrode. Unlike the chromate ion, the pertechnetate ion does not oxidize ferrous ions in weakly acidic solution, although it is effective in inhibition under the same conditions. Oxidation does occur, however, in alkaline solution.

The long half-life of Tc^{99} (2.15×10^5 y) and its 0.29-Mev beta radiation make it eminently suitable for tracer or autoradiographic techniques. At the low concentrations required in studies of inhibition the radiation hazard is of no consequence. The strong absorption bands of the aqueous pertechnetate ion at 2440 and 2875 A are very useful for analytical purposes, the extinction coefficients being 6080 and 2310, respectively. The best value of the ratio of these coefficients is 2.63, and this is a useful test of the purity of pertechnetate solutions. The only electrode potential that has been directly measured[6] is that of the couple,

$$TcO_4^- + 4H^+ + 3e^- \rightarrow Tc(OH)_{4(ppt)};\ \underline{E}_h = 0.738 \text{ volt.} \qquad (4)$$

Other important measured or calculated quantities are:[7]

Solubility of $KTcO_4$ at 25°C = 0.1057 $\underline{M}$

$\underline{S}°$ for TcO_4^- = 47.9 ± 0.3 e.u.

$\Delta\underline{S}°$ for $TcO_{4(aq)}^-$ = -73.3 e.u.

$$\Delta \underline{F}^\circ \text{ for } Tc(OH)_{4(ppt)} = -202.3 \text{ kcal/mole}$$

$$\Delta \underline{H}^\circ \text{ for } Tc(OH)_{4(ppt)} = -246.3 \text{ kcal/mole}$$

Technetium is recovered from fission products by solvent-extraction procedures and converted to the salts NH_4TcO_4 and $KTcO_4$.[8] Metallic technetium is obtained in the form of powder by a two-stage reduction of NH_4TcO_4 by hydrogen. In the first stage, TcO_2 is produced at 200°C, and this is converted to metal at 700-800°C. The powder may be melted in an induction furnace (m.p. 2150°C). A few Fe-Tc alloys have been prepared for metallurgical and electrochemical study.

Studies of Inhibition

When iron or low-carbon steel is immersed in a solution of potassium or ammonium pertechnetate at a pH of 5 or higher, there is an initial reaction wherein the inhibitor ion is reduced with formation of a film of mixed iron and technetium hydroxides. The extent of this reaction depends somewhat on the composition and surface activity of the metal; it may be measured by counting the beta activity of the water-rinsed specimen, since absorption of the weak betas is slight in the thin film produced. The precipitated $Tc(OH)_4$ may be reduced to much less than a monomolecular layer if an oxidizing agent such as hydrogen peroxide is added to the pertechnetate solution. Once passivation is achieved, no further consumption of the inhibitor is required if the solution is exposed to air. Two specimens of S.A.E. 1010 carbon steel have been immersed in a 5×10^{-4} $\underline{f}$ solution since January 1953 (Fig. 1). The beta activity of the rinsed specimens has been measured 30 times over the 10-year period. It rose to a maximum within 2 hr after exposure at 95°C, and then slowly fell about 10 percent owing to

reoxidation of $Tc(OH)_4$ by oxygen. After the activity leveled off, the counts on one specimen showed the presence of 0.146 ± 0.005 μg of technetium on 4 cm^2 throughout the long experiment. The surface of the specimens is still bright, the solution is clear, and the weight is unchanged within 0.1 mg.

ORNL Photo 10803

The film of reaction products first formed is not sufficient for maintaining resistance to corrosion, however, even when heavy deposits are induced. Some minimum concentration of pertechnetate ions must remain in contact with the metal for inhibition to persist. This seems to be about 5 ppm of Tc (5×10^{-5} f TcO_4^-) with electrolytic iron and somewhat higher with carbon steels or cast iron.

Tests demonstrated that the perrhenate ion, ReO_4^-, which is similar in charge, size, and geometry, had no inhibiting properties at any concentration at temperatures between 5° and 95°C. In order to determine whether the radiation or radiolytic products from technetium were involved in its action, a radioactive form of the perrhenate ion was prepared containing $Re^{186,188}$. When this was used at a specific activity corresponding to that of the inhibiting pertechnetate solutions, there was still no inhibition.

That adsorption is involved in the overall corrosion-inhibition process was indicated by experiments in which foreign ions, such as sulfate, were added to inhibited systems. It was shown that all the XO_4^{n-} inhibitors fail when the added ions exceed some concentration, which increases as the concentration of inhibitor is increased.

The failure of the perrhenate ion to inhibit corrosion is particularly significant for adsorption theories, since it is externally similar to the pertechnetate ion. Since the electrochemical polarization theory ascribes the inhibiting properties of chromate and, presumably, pertechnetate ions to their reducibility, polarization measurements have been made to determine the relative rates of reduction of oxygen and reducible inhibitors on a passive iron electrode. Experimental results[9] for oxygen, technetium, and two other inhibitors (Fig. 2) show that the only inhibitor that exceeds oxygen in its rate of reduction at potentials in the neighborhood of the Flade (passivating) potential is OsO_4. These measurements disclosed an interesting feature of the electrodes exposed to pertechnetate ion and osmium(VIII) oxide. In both cases, the reduction products, $Tc(OH)_4$ and $Os(OH)_4$, accelerated the cathodic process, whether oxygen or inhibitor was being reduced. The data from an experiment[10] involving reduction of pertechnetate ions (Fig. 3) again show the favorable properties of technetium compounds. Any $Tc(OH)_4$ cathodically produced on the surface may be removed easily by brief anodic polarization so that effects associated with its presence may be identified.

Theories of Inhibition

In modern studies of the mechanism of corrosion and its inhibition, the radioisotope principally used has been Cr^{51} with a 27.8-day half-life. This nuclide has been used, for example, in studies of the composition of passive films and the rate of deposition of such films on iron. Since the chromate ion is the most useful of the inorganic inhibitors of the passivating type, it is largely around this ion that theories of the action of such inhibitors have been developed.

These theories have considered rather varied conceptions as to the function of the inhibitor, the simplest being that the buffering and oxidizing properties of the chromate ion permit it to oxidize ferrous ions emerging through imperfections in the film and thus precipitate a plug directly in the pore or crack. Evans,[11] who advanced this hypothesis years ago, now attempts to give a more generalized description of the conditions that give rise to corrosion, on the one hand, or formation of a protective film, on the other. For this purpose, he considers the nature of the barrier to anodic dissolution which is interposed by whatever species is preferentially adsorbed on the metal surface from the solution. If this is water or hydroxide ions, emerging ferrous ions first meet the oxygen (negative) side of the adsorbed layer and further migration is impeded by the more remote (positive) side of the layer. Protons are then ejected and carry the anodic current away from the surface, leaving an oxide film that may be protective. Evans then treats the chromate ion analogously, postulating that both it and the sulfate ion may have a positive region, XO_2^{++}, analogous to that shown by the nitronium ion, as in crystalline $NO_2^+ClO_4^-$. Both Uhlig at M.I.T. and Kolotyrkin in Russia have postulated electrical effects at the surface due to adsorption, but without developing the concepts to the degree that would permit a real differentiation between inhibiting and noninhibiting species.

Another theory[12] of the action of reducible inhibitors like the chromate ion is that reduction can occur at a sufficiently high current density and potential to polarize the metal anodically above that potential required for passivation. In this theory, no specific properties

are assigned to the inhibitor other than its oxidation-reduction potential and exchange current density. Although this explanation may suffice in particular cases, it takes no account of specific interactions between the surface and aqueous ions or molecules which certainly are present. Nor does the theory explain how certain nonoxidizing inhibitors make it possible for oxygen to induce passivity in their presence, whereas it would accelerate corrosion in their absence. The polarization theory without supplementary considerations cannot, therefore, be considered a general theory of inhibition.

There are also reasons why oxidation and buffering do not appear sufficient as a general explanation of inhibition.[13] Further, the ideas regarding surface effects of adsorption were unable to explain why chromate ions inhibit corrosion, whereas sulfate ions do not, although the two ions have the same charge, geometrical configuration, and nearly the same size. It was this situation which led to the suggestion that the differentiation must lie <u>within</u> the ion, rather than depend upon its gross charge, size, or geometry.

A crude electrostatic calculation made in 1952[13] showed that chromate and sulfate ions will induce space charges of opposite polarity when adsorbed on a dielectric surface, if, as seemed likely, the S-O bonds are essentially covalent and the Cr-O bonds largely ionic. This difference in polarity should affect the kinetics of charge transfer across the interface. If the space charge is indeed related to the difference in corrosion rates, one should then be able to describe conditions favorable to inhibitory properties in the XO_4^{n-} structure: (1) a high central positive charge; (2) ionic bonding X-O; and (3) an

oxidation-reduction potential low enough to prevent rapid reduction of the XO_4^{n-} particles past the point of forming a surface film of reaction products. The last criterion excluded the powerful oxidant MnO_4^-, and it was therefore apparent[14] that technetium, in the seventh group of the periodic system was the most likely of all elements to have the desired combination of properties. There were reasons for anticipating that the ReO_4^- ion would be less inhibitive, but no way to predict how much less.

Experiments[15] with electrolytic iron and carbon steel in November 1952 immediately demonstrated the correctness of the prediction regarding the pertechnetate ion. It was found to be more effective both at lower concentrations and at higher temperatures than the chromate ion. Thus, 5×10^{-5} f $KTcO_4$ gave excellent protection to S.A.E. 1010 steel at 250°C in a test lasting 116 hr; a visible film formed on the specimen, but the solution remained clear, and the weight of the metal was unchanged within 0.1 mg.

Conclusion

The experiments cited illustrate how technetium may be used to evaluate the various theories of inhibitor action. For example, the absence of buffer action and the mild oxidation potential argue against the early chemical hypothesis. The extreme contrast between the characteristics of TcO_4^- and ReO_4^- disproves any idea of adsorption that considers only the external features of the ions. The theory of purely electrochemical polarization is doubtless a valid description of action that could account for passivation, although the measurement of the comparative contributions of oxygen and reducible inhibitors shows that

oxygen is the chief source of cathodic current with either TcO_4^- or $CrO_4^=$ as inhibitor. The action of sulfate ions in destroying inhibition also shows the incompleteness of the polarization hypothesis.

There is considerable evidence to support the assumption of the electrostatic polarization hypothesis which looks inside the various ions for differentiating characteristics among them.[13,16] The conclusion most likely to be reached ultimately is that each of the ideas thus far proposed is meaningful for some aspect of the inhibitory process, the complete understanding of which will necessitate the combination of the points of view of the electrochemist and the solid-state physicist.

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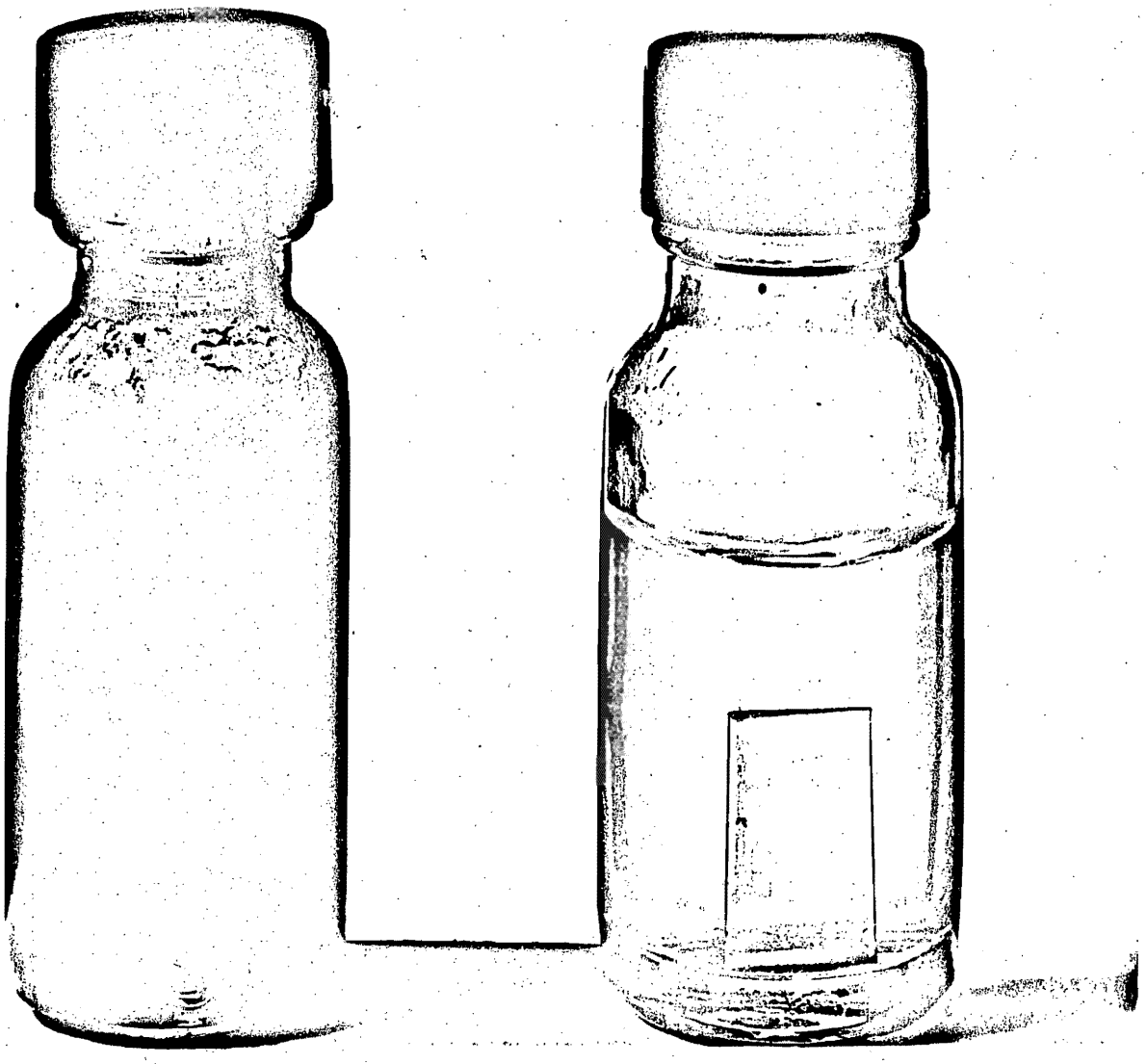

A B

A-CORROSION IN DISTILLED WATER
B-INHIBITION OF CORROSION BY PERTECHNETATE IONS

10 ppm CHLORIDE ION PRESENT

Fig. 1

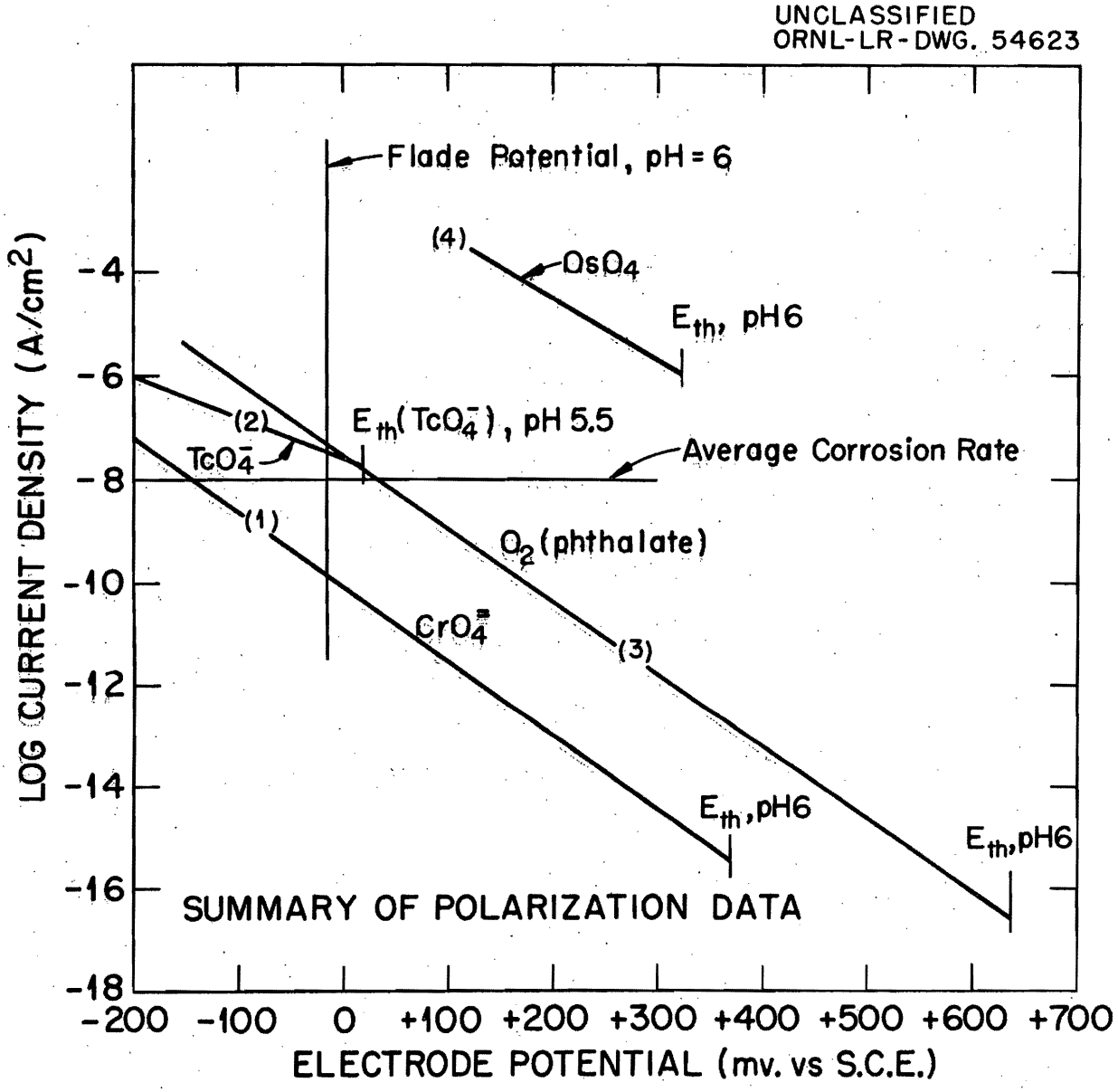
UNCLASSIFIED
ORNL-LR-DWG. 54623
Flade Potential, pH = 6
(4)
OsO4
Eth, pH6
(2)
TcO4-
Eth(TcO4-), pH 5.5
Average Corrosion Rate
(1)
O2 (phthalate)
CrO4=
(3)
Eth, pH6
Eth, pH6
SUMMARY OF POLARIZATION DATA
LOG CURRENT DENSITY (A/cm2)
-4
-6
-8
-10
-12
-14
-16
-18
-200 -100 0 +100 +200 +300 +400 +500 +600 +700
ELECTRODE POTENTIAL (mv. vs S.C.E.)

Fig. 2

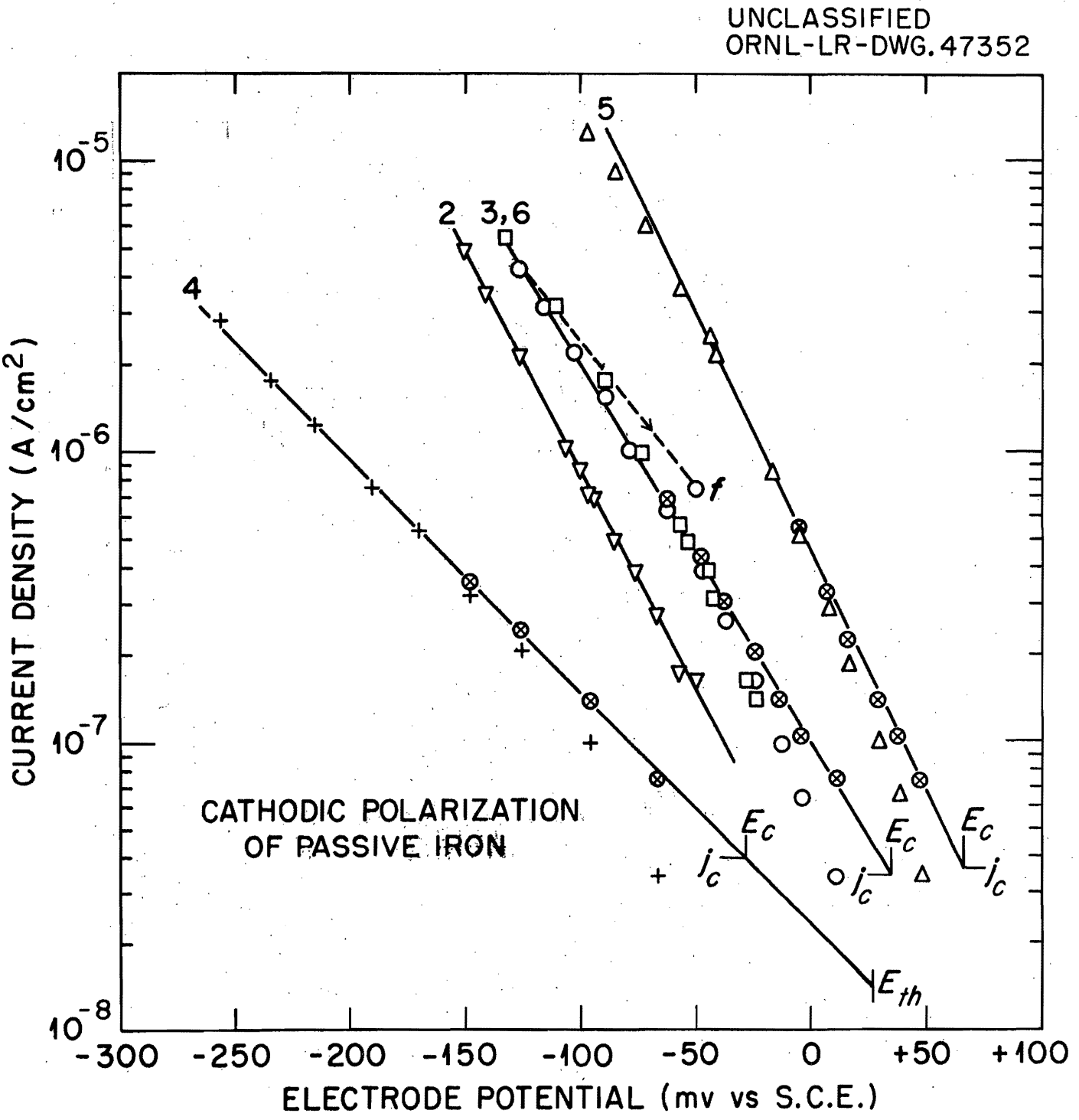
UNCLASSIFIED
ORNL-LR-DWG. 47352
CATHODIC POLARIZATION
OF PASSIVE IRON
CURRENT DENSITY (A/cm²)
ELECTRODE POTENTIAL (mv vs S.C.E.)
10⁻⁵
10⁻⁶
10⁻⁷
10⁻⁸
-300
-250
-200
-150
-100
-50
0
+50
+100
4
2
3,6
5
f
Ec
jc
Eth

Fig. 3

FIGURE LIST

Dwg. or Photo Number	Caption	Page
ORNL Photo 10803	Fig. 1. Uninhibited and Inhibited Steel Corrosion Specimens (10 ppm Chloride Ion Present). A. Corrosion in Distilled Water B. Inhibition of Corrosion by Pertechnetate Ions	
ORNL-LR-DWG. 54623	Fig. 2. Summary of Polarization Data, 25°C. Relative reduction rates of O_2, OsO_4, TcO_4^-, and $CrO_4^=$ on a passive-iron electrode at pH ca. 6 and 24°C. E_{th} represents the calculated reversible potential for each couple. [Reprinted with permission from J. of Phys. Chem., 65: 1367 (1961)].	
ORNL-LR-DWG. 47352	Fig. 3. Cathodic Polarization of Passive Iron. Successive polarizations of passive iron in phthalate or pertechnetate solutions (0.010 f at pH 5.6 - 5.9). (2) Oxygenated phthalate alone; (3) Oxygenated pertechnetate, points ⊙, ending at f; (4) Pertechnetate in helium, after point f; (5) Oxygenated pertechnetate with $Tc(OH)_4$ left on film; (6) Same after removal of $Tc(OH)_4$ by anodic polarization, points ⊡; at lowest current densities, data points require correction for an anodic current density of 4×10^{-8} amp/cm^2, points ⊕. [Reprinted with permission from J. of Phys. Chem., 64: 1884 (1960)].	

I. PRINCIPLES OF ISOTOPE TECHNOLOGY

In this first issue, a brief general review of the principles of isotope production and utilization will be followed by more detailed consideration of various applications in a number of areas. In subsequent issues, advances in technology and new applications will be covered but, in general, routine applications will be ignored. Since there are only certain utilization principles, we hope that novel disclosures in one area may suggest analogous techniques to users in other areas.

A chronological outline of the development of the isotopes program is included to provide background information. Many publications, including annotated bibliographies, may be used to supplement the reviews in the first issues of this quarterly by those who want more detailed information on early work and developments. Among these are the three-, five-, and eight-year summaries of isotope distribution and utilization;[1] the subsequent bibliographies published by the U. S. Atomic Energy Commission;[2] the list of special sources of isotope information;[3] the reports of the Atomic Energy Commission to Congress;[4] and various AEC and nongovernment publications.[5]

Only about 100 radioisotopes are now routinely available,[6] but more than 1000 have been isolated and more than 1500 identified. Certainly many of these have properties suitable for a wide variety of applications. We plan to keep readers abreast of progress in this

direction, particularly as enriched targets and higher fluxes are used, and to suggest profitable areas of utilization. The subject "Stable Isotopes" will be covered in a separate section.

That the program has grown -- and grown rapidly -- is illustrated by examination of information on shipping, licensing, and publications since 1946 (Table I-1) and by comparison of the number of labeled compounds available in 1949 with the number available in 1962 (Table I-2).

Table I-1. Growth of Isotopes Program

To (Date)	Total Shipments	Total Authorizations and Licenses	Cumulative Total Publications
Aug. 1949	11,400	5,000[a]	1,850
June 1951	18,900	13,100[a]	3,000
Dec. 1954	64,200	37,155[a]	7,000[c]
Dec. 1958	118,600	4,347	15,000[c]
Dec. 1962	170,000	5,881[b]	40,000[c]

[a]Until 1956, individual authorizations were processed for each isotope. After that, users were able to obtain licenses which could authorize any number of isotopes.

[b]The 1962 figure does not include licenses transferred to the jurisdiction of California (580), Kentucky (68), Mississippi (37), and New York (637).

[c]Estimated on basis of sampling from Nuclear Science Abstracts, Chemical Abstracts, and J. of Nuclear Medicine.

Table I-2. Number of Labeled Compounds Available in 1949-1962

Isotope	1949[a]	1962[b]
S^{35}	5	131
I^{131}	5	56
P^{32}	13	81
C^{14}	~100	~1300

[a] p. 68 of reference 1a.

[b] pp. 13-132 of reference 7.

Isotope Production

Any applications involving radioisotopes presume availability of suitable nuclides. Most radioisotopes are prepared by nuclear reactions involving neutrons from reactors, by cyclotron reactions, or by separation of gross fission products which remain after the processing of spent fuels to recover the unused fissile material (Tables I-3, 4, 5).

Table I-3. Neutron Reactions

Reaction	Example
(n,γ)	$Co^{59}(n,\gamma)Co^{60}$
	$Fe^{58}(n,\gamma)Fe^{59}$
(n,p)	$N^{14}(n,p)C^{14}$
	$S^{32}(n,p)P^{32}$
(n,α)	$Ca^{40}(n,\alpha)Ar^{37}$
	$Li^{6}(n,\alpha)H^{3}$
$(n,\gamma;EC)$	$Xe^{124}(n,\gamma)Xe^{125} \xrightarrow{EC} I^{125}$
$(n,\gamma;\beta^{-})$	$Pd^{110}(n,\gamma)Pd^{111} \xrightarrow{\beta^{-}} Ag^{111}$

Table I-4. Cyclotron Reactions

Reaction	Example
(p,n)	$Li^{7}(p,n)Be^{7}$
(d,n)	$Ge^{72}(d,n)As^{73}$
$(d,2n)$	$Cr^{52}(d,2n)Mn^{52}$
(d,α)	$Mg^{24}(d,\alpha)Na^{22}$
(p,pn)	$Ca^{48}(p,pn)Ca^{47}$
(p,α)	$Mg^{25}(p,\alpha)Na^{22}$

Table I-5. Fission Products

Isotope	% Fission Yield	Half-life
Kr^{85}	0.3	10.3 y
Sr^{90}	5.9	28 y
Zr^{93}	6.5	1×10^6 y
Tc^{99}	6.1	2×10^5 y
Ce^{144}	6.1	0.8 y
Pm^{147}	2.6	2.6 y
Cs^{137}	5.9	30 y
I^{129}	1.0	1.7×10^7 y

Isotope Utilization

The principles of isotope utilization (Fig. I-1) have been reduced to three major types, or methods of use. The first two applications do not incorporate the activity into the process material and there is no possibility of its remaining in the final market product. In the third, a radioisotope is introduced into the substance under investigation -- usually a short-half-life isotope -- in small amounts, but nevertheless often in sufficient quantity to leave an objectionable residual activity. The three types of applications involve:

1. The Effects of Radiation on Materials. The radioisotope is used as a fixed source of radiation, comparable to ordinary radium or x-ray machines, and the radiation is allowed to impinge upon the material that is to be affected by the radiation. There are many important examples of this application. The target material may be a plastic, exposed in order to alter its properties (Fig. I-2); a patient with a malignancy, receiving therapy; food being irradiated to retard decay (Fig. I-3); or a phosphor being excited to luminescence by an admixed isotope.

2. The Effects of Materials on Radiation. Here the target material affects the radiation in such a way as to provide information about the material. The radiation, either reflected from or penetrating the material, is measured by a counter or photographic emulsion, depending on the information required. For example, in industrial or medical radiography a radioactive source can replace x-ray machines to photograph bones, teeth, or castings. Industrial utilization includes many important applications to process and product control: measuring blind (inaccessible) liquid levels (Fig. I-4); gaging thickness of moving sheets of paper, plastic, and metal (Fig. I-5); controlling cigarette densities; radiographing castings and internal structures of equipment (Fig. I-6); tracing oil flow in pipelines (Fig. I-7); and, in some of the more recent applications, using the isotopic heat resulting from the complete absorption of the radiant energy by a heavy metal such as lead or uranium (Fig. I-8).

3. The Tracing of Radioactive Atoms. Perhaps the most interesting and significant use of radioisotopes takes advantage of the continuous "broadcast signal" generated by the decaying radioisotopes. Suitable detectors can be used to follow even the most complicated courses of these atoms as they take part in chemical reactions or biological processes, or as they are involved in physical transfer. The radioisotope tracer may be used to study surface phenomena such as induced passivity; it may be incorporated in a compound added to water running through a leaking pipe; it may be an active atom in sugar (e.g., C^{14}) for studying body metabolism; or it may be a label in a fertilizer compound used to study the mechanism of fertilizer uptake.

CHRONOLOGICAL OUTLINE OF DEVELOPMENT OF ISOTOPES PROGRAM

1946, June	First public announcement of reactor-produced radioisotopes available under the distribution program appeared in Science (served as 1st radioisotope catalog)
1946, Aug.	First reactor-produced radioisotope shipment from Oak Ridge
1947, Jan.	Jurisdiction of program turned over to civilian Atomic Energy Commission
1947, Mar.	Initiation of reactor service irradiations; that is, a service whereby the radioisotope user may submit his own sample for irradiation or neutron bombardment in a nuclear reactor
1947, July	First compound labeled with radiocarbon (methyl alcohol) available from the Commission
1947, Sept.	Routine availability of chemically processed radioisotopes with standard specifications
	Initiation of a program for international distribution of radioisotopes
1948, Jan.	Formation of an Advisory Committee on Isotope Distribution (to replace interim policy committee set up under the Manhattan Project)
	Initiation of a program for distribution on a loan basis of more than 100 varieties of electromagnetically concentrated isotopes of about 30 elements
	Publication of regulations by the Interstate Commerce Commission for shipment of radioisotopes by common carrier (rail and truck)
1948, Apr.	Initiation of a program making available free of production costs three isotopes -- radiosodium, radiophosphorus, and radioiodine—for research, diagnosis, and therapy of cancer and allied diseases
1948, June	Opening of first Commission-sponsored training course in radioisotope techniques by Oak Ridge Institute of Nuclear Studies
1948, July	Beginning of production and distribution of radioisotope-labeled compounds by commercial firms
1948, Sept.	Distribution of tritium (radioactive hydrogen) and stable helium-3

1948, Sept.	Completion of a nationwide intercomparison of iodine-131 radiation analyses conducted by the National Bureau of Standards and Atomic Energy Commission
1949, Feb.	Extension of Commission's program for support of cancer research to include the availability of all normally distributed radioisotopes free of production costs for use in cancer research
1949, June	Initiation of a program for cyclotron production and distribution of certain long-lived radioisotopes not producible in the nuclear reactor
1949, July	Enactment of regulations by the Civil Aeronautics Board for shipment of radioisotopes by commercial aircraft
1950, Mar.	Completion of new large-scale radioisotope processing facilities at Oak Ridge National Laboratory
1950, Oct.	Announcement of program for returning radioactive wastes to Commission facilities for disposal
1950, Dec.	Initiation of cooperative visitation with state health departments to radioisotope users
1951, Jan.	Announcement that Brookhaven National Laboratory will provide reactor irradiation services and radioisotopes
1951, Mar.	Elimination of requirement for publication of results obtained with AEC-produced radioisotopes
1951, Apr.	Publication of first issue of the quarterly bulletin, "Isotopics--Announcements of the Isotopes Division"
	Publication in Federal Register of regulations governing radioisotope distribution, including designation of "generally licensed" quantities (requiring no license application)
1951, July	Expansion of international distribution of radioisotopes to include all reactor-produced radioisotopes (except tritium and polonium-210) and to authorize the export of these materials for use in scientific research, medical research, industrial research, medical therapy, and industrial applications
1951, Nov.	First radioisotope (iodine-131) accepted by Federal Food and Drug Administration as effective new drug; phosphorus-32 accepted June, 1952; and colloidal gold-198 accepted October, 1954.

1952, Jan.	First film of thirteen-part radioisotope training film series, "The Radioisotope," made available for distribution
1952, Apr.	Announcement of availability of neutron activation analysis service at Oak Ridge National Laboratory
1952, May	Publication of AEC-financed supplement to the Journal of the American Chemical Society on labeled-compound synthesis
1952, June	Revision of AEC Cancer Program to make most radioisotopes available at 20 percent of catalog prices
1952, July	First authorization of industrial waste disposal service
1953, Dec.	Publication in the Federal Register of criteria governing the authorization of AEC-supplied radioisotopes for research and development, for human use, and for use under a general authorization; use of license-exempt quantities of radioisotopes in humans expressly prohibited
1954, Jan.	Announcement of availability of facilities for irradiation of large items at Brookhaven National Laboratory
	Publication by USAEC of list of 492 isotope-labeled compounds available from 13 commercial suppliers, the National Bureau of Standards, and Oak Ridge National Laboratory
1954, July	Announcement of standard reactor units of radioisotopes
	Revision of Federal Radioisotope Distribution Regulations to cover radioisotopes produced in any publicly or privately owned nuclear reactors located within the United States, its territories, or possessions
	Stable isotopes made available for export
1954, Sept.	Issuance of Argonne National Laboratory booklet, "Irradiation Services," containing detailed information on services available at ANL's Research Reactor CP-5, Experimental Breeder Reactor, and its 60-inch cyclotron
1954, Oct.	Approval of Kilocurie Fission Product Separation Plant to be located at Oak Ridge National Laboratory
	Announcement of Gamma Irradiation Facilities at National Reactor Testing Station to provide gamma-ray fields of the order of 10 million roentgens per hour
1955, July	Large-scale production of high-specific-activity cobalt-60 begun at AEC's Savannah River Plant

1955, Nov.	AEC withdraws from processing and distribution of cyclotron-produced isotopes
1956, Feb.	Generally licensed quantities of radioisotopes increased, permitting wider use and interchange of small samples without an application; simplification of international distribution of radioisotopes permitting U. S. licensees to export radioisotopes of elements with atomic numbers 3 to 83
	Sale of enriched stable isotopes approved by AEC
1956, Aug.	Records relating to radioisotope licenses made publicly available in AEC's Public Document Room
1958, Mar.	Cobalt-60 encapsulation services turned over by AEC to private enterprise
1958, Apr.	Fission-product cerium-144 reduced by AEC to $1.00/curie for orders of 100 kilocuries or more
1958, Aug.	Kilocurie Fission Product Separation Plant started operation at Oak Ridge National Laboratory
1958, Oct.	Isotope Development Program initiated for "encouraging the growth and development of radioisotope and radiation uses in industry"
1959, Jan.	Division of Isotopes Development Advisory Committee on Radioisotopes and Radiation formed
1959, Aug.	Routine availability of calcium-47 announced
1960, May	AEC withdraws from sale of polonium-210 sources of less than 20 curies
1960, July	AEC discontinues providing standard reactor-irradiated units
	AEC reaffirms policy of withdrawal from neutron irradiation service
	Abbott Laboratories announces availability for first time of iodine-131 from private manufacturer
1961, Jan.	Electromagnetic separations facilities at Oak Ridge National Laboratory expanded from 4 to 28 calutrons
1961, May	Cobalt-60 metal sources become available from private industry; AEC limits supply from Commission facilities to orders of 100 kilocuries or more; large-order price reduced to $1.00/curie for specific activities less than 30 curies per gram

1961, June	Completion by Oak Ridge National Laboratory of first large fission-product source for use in thermoelectric generator--17 kilocuries of $SrTiO_3$, 110 thermal watts; later placed in generator developed by Martin Company to power an unmanned arctic weather station for the U. S. Weather Bureau
1961, July	AEC terminates its radioisotope discount program in medicine, agriculture, and biology research
1961, Aug.	Routine availability of iodine-125 announced
1961, Nov.	AEC withdraws Mound Laboratory as a supplier of plutonium-beryllium neutron sources
1961,(early)	First private production and marketing of Co^{60} (GE and Westinghouse)
1962,(early)	First private production and marketing of short-lived isotope (Isoserve, Inc.)
1962, Mar.	AEC withdraws from neutron activation analysis service in favor of industry
1962	High Intensity Radiation Development Laboratory to generate the engineering design and operational knowledge essential to expansion of the potentials of process radiation completed at Brookhaven National Laboratory
1962, Mar.	Fission-product cesium-137 reduced by AEC to $0.50/curie for orders of 100 kilocuries or more
	AEC makes up to 10 grams americium-241 and neptunium-237 available to industrial licensees
	Fission-product strontium-90 reduced by AEC to $0.75/curie for orders greater than 30,000 curies
1962, May	Completion by Oak Ridge National Laboratory of first large gamma-ray (cesium-137) source for thermoelectric generator--27 kilocuries cesium borosilicate, 125 thermal watts; later inserted in generator by Royal Research Corporation for operation of a deep-sea seismograph station for Lamont Geological Observatory
1961-1962	Fabrication of a series of SNAP sources for Navy navigational beacon, Coast Guard buoy, Navy Antarctic weather station, and unmanned Navy floating weather station; sources by Oak Ridge National Laboratory, generator assembly by Martin Company; 31-225 kilocuries $SrTiO_3$, 195-1400 thermal watts

1962, Sept.	First AEC license to industry for land disposal of by-product wastes
1962, Oct.	AEC withdraws from fabrication and sale of polonium-210 sources of less than one kilocurie for private use
1962, Dec.	Development of Cs^{137}-Li silicate glass at Oak Ridge National Laboratory for Brookhaven large-scale research gamma-irradiator
1963, Jan.	First strontium-90 fuel source processed by private industry
	Price reduction for cobalt-60 in the high-curie-supply area; aimed at encouraging greater commercial reprocessing and distribution
	AEC withdraws from providing iridium-192 (solutions and sealed sources) due to industry capability
1959-1962	AEC Division of Isotopes Development arranged and participated in 29 isotope symposia throughout the country to encourage industrial use of isotopes
1962-1963	AEC relinquishes certain of its regulatory authority to states of Kentucky, California, Mississippi, New York, Texas, and Arkansas.
1963, Aug.	AEC withdraws from by-product waste disposal for licensees

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   b. Ibid., Five-Year Summary, August 1951, 451 pp. (5-1/2 x 9 in.), 1400 references.

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   a. Isotopes: A Bibliography of United States Research and Applications 1955-57, USAEC Report TID-3076, May 1958, 261 pp., 6000 references, $2.25.

   b. Isotope Techniques in Biological Sciences, USAEC Report TID-3512, February 1958, 44 pp., 807 references, $1.25.

   c. Isotopes in Biochemistry and Biosynthesis of Labeled Compounds, USAEC Report TID-3513, March 1958, 89 pp., 2430 references, $2.25.

   d. Radioisotopes in Medicine and Human Physiology, USAEC Report TID-3514, August 1958, 124 pp., 2862 references. Out of print; $19.80 in facsimile; microfilm $6.30.

   e. Radioisotopes in Animal Physiology, USAEC Report TID-3515, December 1958, 118 pp., 2439 references. Out of print; $9.60 in facsimile; microfilm $3.74.

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5. a. W. F. Libby, Industrial Uses of Isotopes, Ann. Rev. Nucl. Sci., 11: 461-82 (1961).

b. Atomic Energy Research, U. S. Atomic Energy Commission, December 1961, 333 pp. Available from the Superintendent of Documents, Washington. This is a general review of the applications of atomic energy to life and the physical sciences. $2.25.

c. Radioisotopes in the Service of Man, Fernand Lot, United Nations Educational, Scientific, and Cultural Organization, Paris, 1959, 82 pp., $1.00.

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6. Oak Ridge National Laboratory Radioisotopes Catalog and Price List, May 1960.

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FIGURE LIST

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	Fig. I-2. Effect of Radiation on Plastics	32
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AC-1-1-1059	Fig. I-4. Determination of Hidden Liquid Level in Cans (Indus. Nuc.)	34
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BASIC PRINCIPLES OF RADIOISOTOPE UTILIZATION

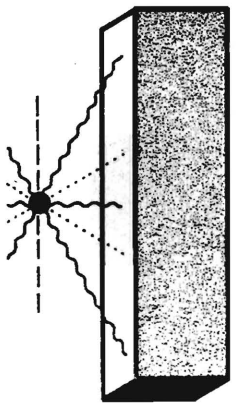
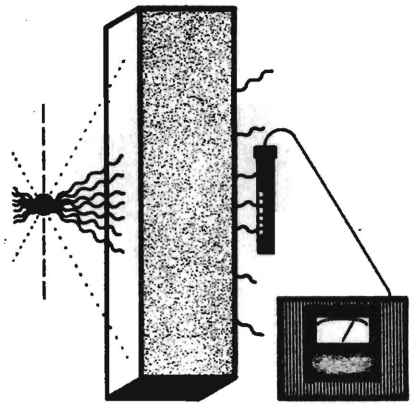
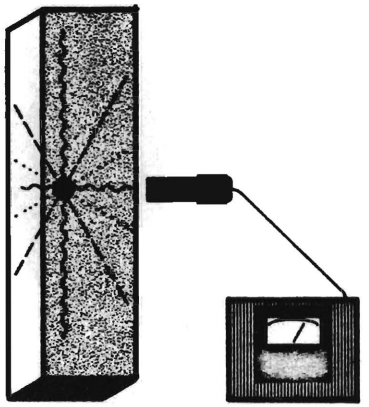
RADIATION AFFECTS MATERIALS	MATERIALS AFFECT RADIATION	RADIATION TRACES MATERIALS
		
FOOD & DRUG STERILIZATION	CLINICAL RADIOGRAPHY	METABOLIC STUDIES
STATIC ELECTRICITY ELIMINATION	THICKNESS (DENSITY) GAGING	FLUID FLOW
TELETHERAPY	INDUSTRIAL RADIOGRAPHY	ABRASION & WEAR MEASUREMENT
RADIATION GENETICS	LIQUID LEVEL & HEIGHT INDICATION	STUDY OF REACTION MECHANISMS

Fig. I-1

USAEC-ID-298A

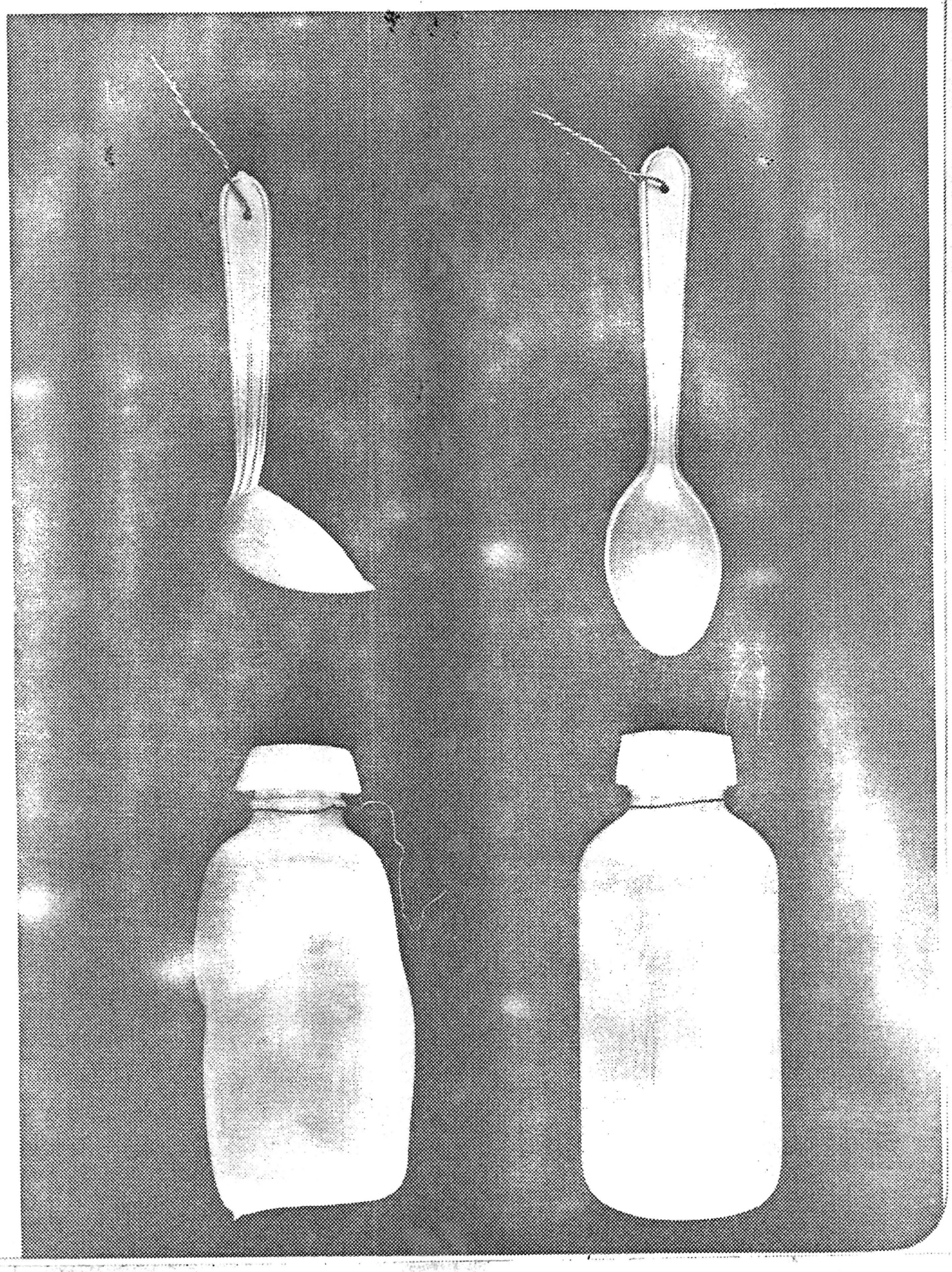


Fig. I-2

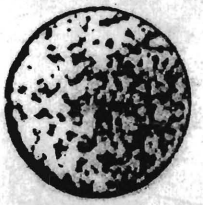
EFFECT OF IRRADIATION ON THERMAL STABILITY OF PLASTICS

Cold Sterilization

AN

INDUSTRIAL USE OF RADIOISOTOPES

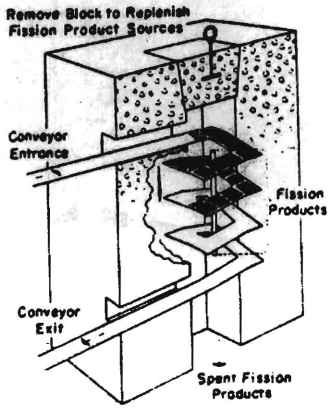
EFFECT OF RADIATION
ON MICROORGANISMS
(ASPERGILLUS NIGER)



NO RADIATION
(MOLD COUNT = 45,000,000)

260,000 REP
(MOLD COUNT = 0)

STERILIZATION UNIT



ATOMIC RADIATION HAS BEEN USED TO PRESERVE DRUGS — FOODS — KILL INSECT LARVAE IN PACKAGED PRODUCTS — CONTROL REPRODUCTION OF LIVESTOCK PESTS.

Fig. I-3

USAEC-ID-271A

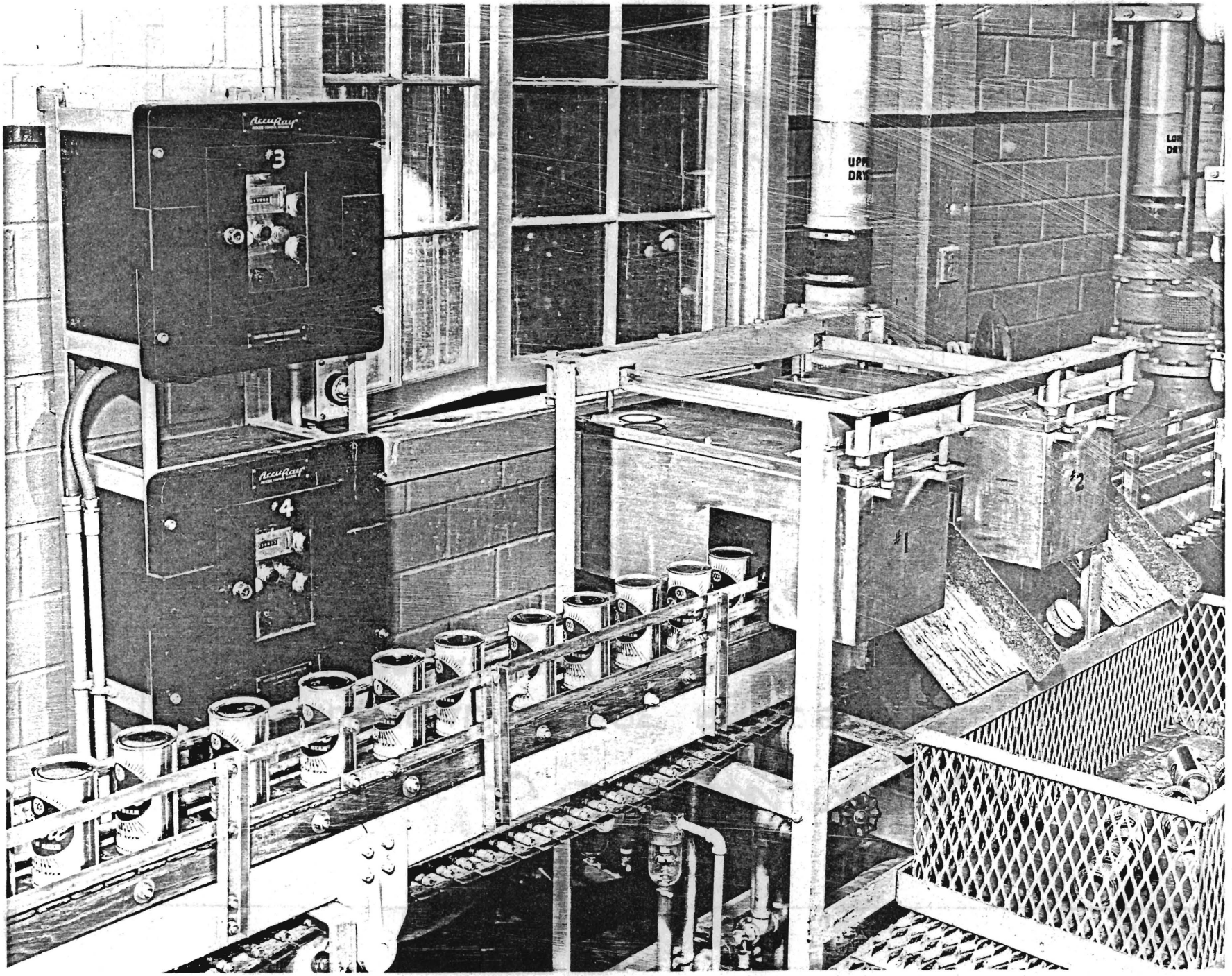
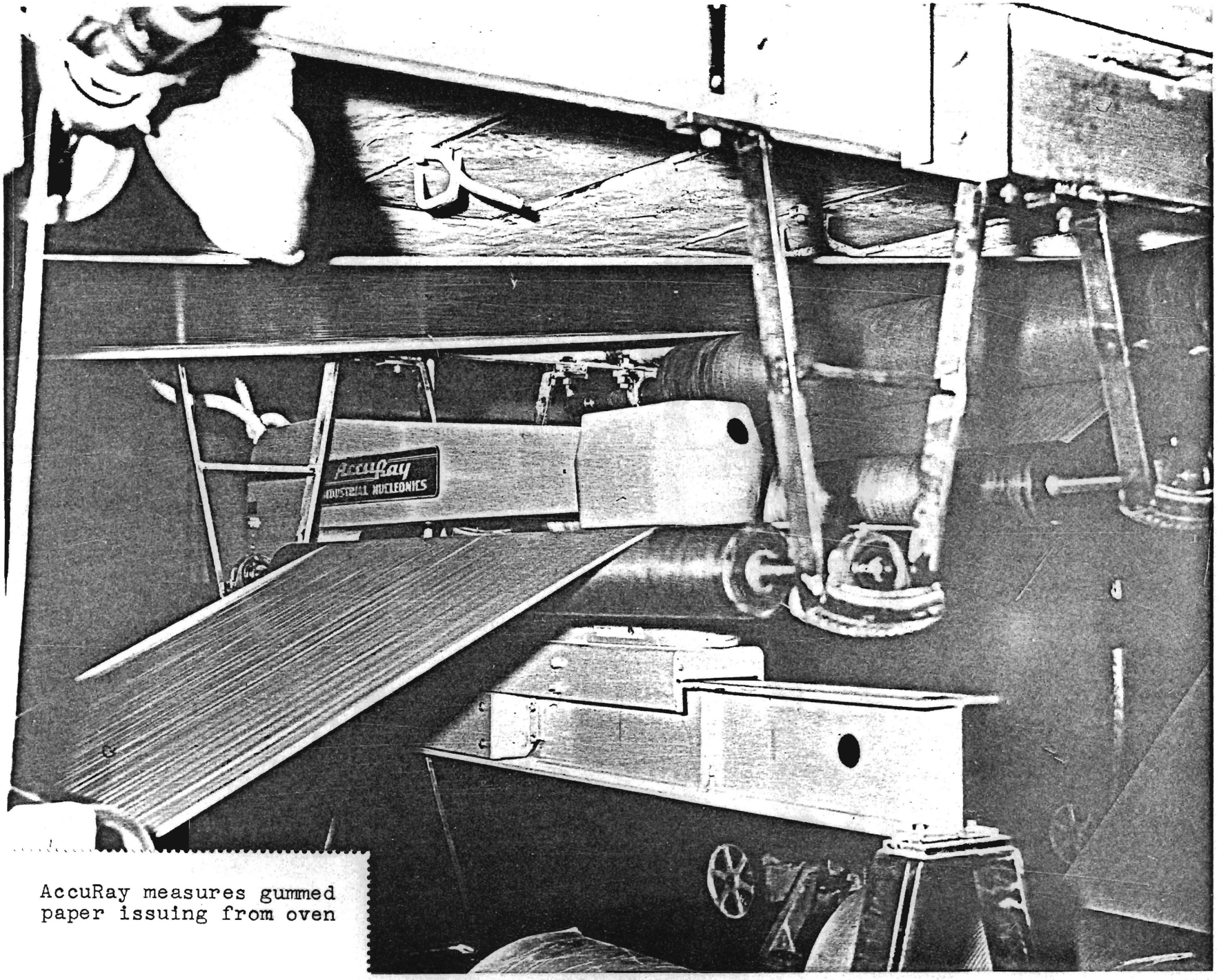
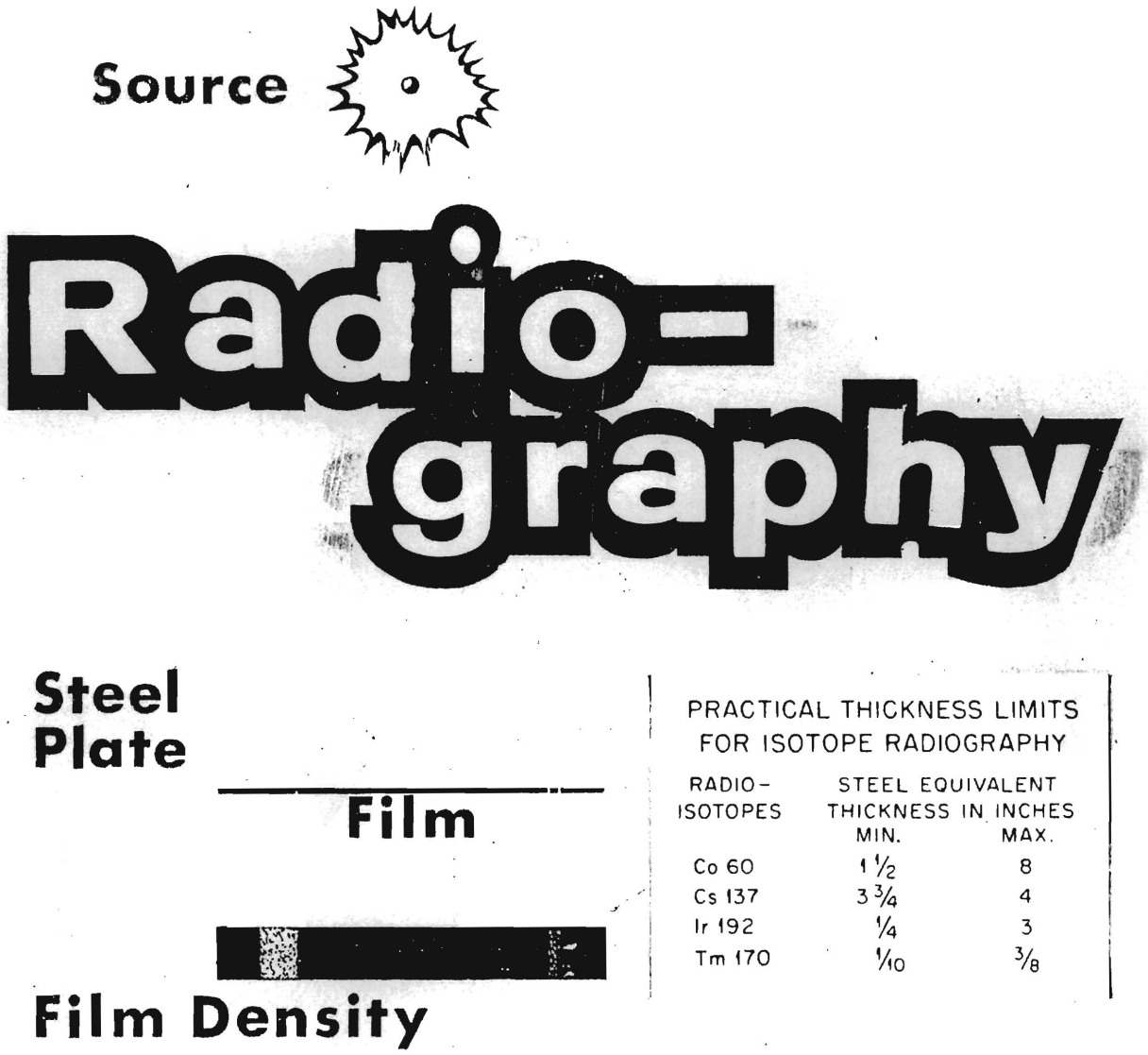


FIG. 1. 4



AccuRay measures gummed paper issuing from oven

Fig. 1-5



PRACTICAL THICKNESS LIMITS FOR ISOTOPE RADIOGRAPHY

RADIO-ISOTOPES	STEEL EQUIVALENT THICKNESS IN INCHES MIN.	MAX.
Co 60	1 1/2	8
Cs 137	3 3/4	4
Ir 192	1/4	3
Tm 170	1/10	3/8

Fig. 1-6

Tracing
STORAGE
A
B
Oil A
Oil B
Radioisotope
Oil A
Pumping Sta.

Fig. 1-7

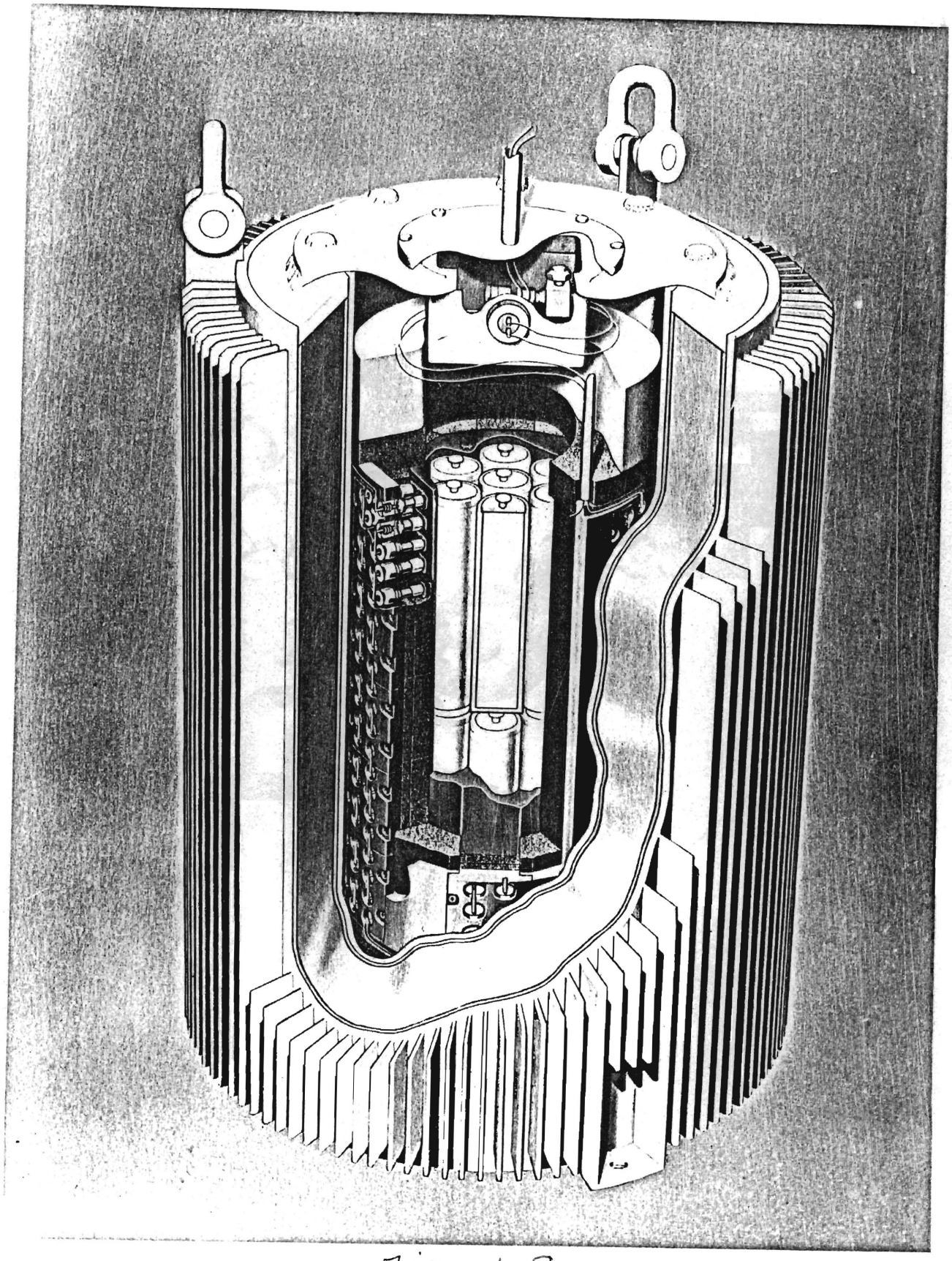


Fig. 8

The following review article, an official report of the status of U. S. Atomic Energy Commission research and development activities, is presented as written for oral presentation at the Fifth Japan Conference on Radioisotopes, Tokyo, Japan, May 21-23, 1963.

STATUS OF THE UNITED STATES PROGRAM ON ISOTOPES AND RADIATION DEVELOPMENT

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United States Atomic Energy Commission
Washington, D. C.

INTRODUCTION

Major technological advances are being made in the United States to extend and accelerate the beneficial applications of radioisotopes and radiation. The government's program, carried out by the Atomic Energy Commission, is contributing importantly to the overall progress in the United States on the development of radioisotopes and radiation technology. The AEC program is directed toward (1) advancing radioisotopes and radiation technology to encourage development of beneficial applications, particularly those to meet problems of urgent public interest, (2) accelerating realization of the potential contribution of radioisotopes and radiation applications to the national economy and welfare, and (3) providing an important contribution to world development in the peaceful uses of atomic energy.

To be presented at the Fifth Japan Conference on Radioisotopes, Tokyo, Japan, May 21-23, 1963. Note: This paper is for publication; it will be excerpted for oral delivery.

These objectives are being achieved through a multiphase research and development program including the following major elements:

1. Radioisotope and Fission Product Production and Separations Technology fulfills the Atomic Energy Commission's responsibility for ensuring the availability of radioisotopes and radiation sources to meet the changing needs of advancing science and technology in the United States.
2. Isotopic Power Fuels Development is directed to the development and production of isotopic fuels and fuel forms for a broad spectrum of thermal applications, including the SNAP auxiliary electrical power devices.
3. Process Radiation Development fosters development of a broad technology leading to productive use of megacurie quantities of fission products and other radioisotopes for process radiation purposes.
4. Radiation Pasteurization of Foods involves the development of technology for low-dose radiation processing of certain fruit and fishery products to extend their refrigerated shelf-life.
5. Radioisotope Technology Development creates a broad base of new and improved technology required for extending and speeding up the application of radioisotopes in science and technology.

The progress and scope of activities in each of these areas over the last year have been impressive.

In the production of radioisotopes, advances continue to be made through development of processes for producing new isotopes and chemical forms, as well as high radiochemically pure materials to meet the needs of science and technology in the United States. We are now routinely producing multimegacurie quantities of Cobalt 60, Strontium 90, and Cesium 137 for process radiation application developments and for use as thermal sources for devices to power automatic weather stations, light buoys, seismographic units, and underseas electronic equipment. During the past year the world's largest single Cobalt 60 source, approximately 1.4 million curies, was produced at the Atomic Energy Commission's Savannah River production site and delivered to the U. S. Army Radiation Laboratory at Natick, Massachusetts. Included in our current large-source production effort are 120,000 curies of Cobalt 60 for the Japan Atomic Energy Research Institute. The requirement for reliable, long-term auxiliary electric power in the national space program has resulted in a stepped-up development effort in the production of large quantities of previously scarce radioisotopes such as Plutonium 238, Curium 242, and Curium 244. These radioisotope power sources are planned for use in lunar scientific probes, communications, and meteorological satellites.

In the United States an estimated 50 kilowatts of installed radiation power is now being used for commercial process radiation purposes. The value of production is estimated at 20 million dollars annually. We have witnessed the emergence on the commercial market of new radiation-produced products which can either be produced better by radiation, or

which cannot be made readily by other means. The first industrial use of radiation for bulk chemical manufacture in the United States is now being carried out by the Dow Chemical Company. This involves the commercial-scale production of ethyl bromide, using Cobalt 60 as the source of radiation energy. Significant advances made in the Commission's process radiation development program range from radiation polymerization of wood impregnated with monomer materials to form "wood-plastic alloys" to the discovery of a new reaction mechanism for formation of hydrazine by irradiation of gaseous ammonia.

Significant achievements have been made in developing the technology for radiation pasteurization of foods, including processing conditions needed to insure the retention of freshness and nutritional values. In addition, preliminary marketing and economic studies are being made in anticipation of the ultimate commercialization of the process.

In the area of radioisotope technology, advances range from developments leading to an ocean-bottom sediment density meter to an activation analysis technique for analyzing the surface of the moon. A significant new development is a portable beta-excited X-ray system which uses a Promethium 147 radioisotope source and film developing unit so compact that both can be held in one hand. Neutron activation analytic techniques have been greatly expanded in range of application from identification of manufactured materials, such as plastics, to scientific crime detection.

The Atomic Energy Commission's research and development work on isotopes and radiation is managed by its Division of Isotopes Development

and carried out in the AEC's National Laboratories and with private research laboratories. Appendix 1 identifies radioisotopes and radiation research and development projects now being carried out for the Atomic Energy Commission by private research groups.

Radioisotope and Fission Product Production and Separations Technology

In certain respects, the Atomic Energy Commission's work on radioisotopes and fission product production and materials development is probably one of its most important activities. This is based on the fact that all its radioisotopes and radiation technology development effort must ultimately be based on a continuing availability of suitable radioactive materials in adequate quantities and at a reasonable price.

The changing needs of science and technology have presented a continuing challenge to our radioisotopes production program. Today 137 individual radioisotopes are routinely available from the Atomic Energy Commission. On almost a day-to-day basis efforts are directed toward development of new radioisotopes and radiation sources, as well as toward improved production processes for existing radioisotopes. Improved production is particularly important because many recent industrial and medical applications require ever increasing quantities of radioactive materials. For example, as of January 1, 1963, Oak Ridge National Laboratory had produced and distributed about 2.1 million curies of radioactive materials during the entire 16 years of the radioisotopes distribution program. However, almost half of this amount or about one-million curies was distributed during just the past two years. This substantial

growth in the demand for radioactive materials and associated services and equipment has also led to the development of a substantial private radioisotopes business in the United States. The market for modified isotope products and devices in 1962 was in the $27 - 34,000,000 range. The following is a break-down according to product areas:

-- current sales of labeled organic compounds are $3 - 4,000,000 per year. Growth rate is at a level of approximately 10 - 15% per year.

-- current sales for radiopharmaceuticals are at a $5,000,000 level. Growth rate is at a level of approximately 25% per year.

-- sales of teletherapy units have reached the $2 - 3,000,000 per year level. Seven U. S. manufacturers produced these devices.

-- the market for radiographic equipment and commercial irradiation services has reached the $15 - 20,000,000 per year level.

Several developments in the area of new radioisotopes production should be mentioned. One of the most promising is the use of enriched targets in radioisotope production. For example, Hafnium 175, a low-energy gamma emitter with a 70 day half-life was produced by the reactor irradiation of a Hafnium 174 target enriched to 7.85% Hafnium 174. Similarly, Germanium 71 was produced through the irradiation of 86% enriched Germanium 70. Tin 119m was made by the irradiation of a target enriched in Tin 118.

Another area is the development of X-ray and bremsstrahlung radiation sources, tailor-made to emit a specific energy X-ray for a specific purpose. We have prepared Promethium 147 pellets encapsulated in aluminum to form sources of low energy X-rays. Beta emission from the decay of Promethium 147 produces its characteristic 38 Kev K X-ray as secondary radiation. In the same manner, the characteristic 30 Kev X-ray of antimony can be produced by pressing a mixture of antimony metal and promethium oxide into a pellet. Promethium 147 bremsstrahlung sources have also been fabricated with platinum sponge and tungsten carbide. Three-hundred millicuries of Promethium 147 absorbed on Decalso and mixed with 1 gram of platinum gave 1.7×10^8 photons per minute of the characteristic 67 Kev platinum X-ray. The same quantity of Promethium 147 absorbed on Decalso and mixed with two grams of tungsten carbide emits 1.7×10^8 photons per minute of the characteristic 59 Kev tungsten X-ray.

Substantial progress is also being made in techniques for the preparation of isotopically enriched materials. For example,

we expect soon to be prepared to offer Krypton 85 enriched up to as much as 50%.

Another potentially valuable development we are actively following is the use of Iodine 125 sources for medical and industrial X-ray radiography.

During the past year, Oak Ridge National Laboratory has been intensively studying the preparation and encapsulation of Iodine 125 X-ray sources. At first, the compound used was silver iodide. The source was prepared by precipitating silver iodide with the minimum iodide carrier added, and then centrifuging and drying the precipitate. The precipitate containing 2 curies of Iodine 125 was encapsulated in stainless steel having a 2.5-mm inner diameter and a 1-mil-thick window. Evaluation of the use of Iodine 125 as medical radiography source has been made at the Ohio State University.

It was determined, as a result of the evaluation, that the efficiency of the emission was decreased because of the high K absorption edge of silver for the characteristic Iodine 125 gamma photon. To improve the output, sodium iodide was used as the compound for a second source which is now being evaluated.

Oak Ridge National Laboratory has also investigated plating Iodine 125 on platinum, silver, and palladium by chemical reaction. Only limited deposition of iodine was obtained. Also, the deposited activity rubbed off easily and could be released upon heating. We are therefore presently examining the absorption of Iodine 125 vapor on a tiny porous carbon plug and encapsulating it in stainless steel with an electroplated nickel end window 1 mil thick.

Proceeding apace with Iodine 125 source fabrication is the development of improved production techniques to reduce cost. Oak Ridge National Laboratory is now preparing to produce enriched

Xenon 124 target materials in order to increase yields, and is examining the installation of a loop in the Oak Ridge Research Reactor for continuous production of Iodine 125.

Isotopic Power Fuels Development

A most exciting new area of radioisotopes application is based on the liberation of thermal energy during radioisotope decay. This has led to the development of small nuclear power supplies, which derive their energy from radioisotope decay, for use in locations and under environments where more conventional sources of electric power are unsatisfactory.

Isotopic power systems, or SNAP devices as they are called, are now being used to power navigational satellites in space, remote automatic weather stations near both the North and South Poles, and navigational buoys and underseas electronic equipment. Other SNAP devices will probably soon power barge-mounted automatic weather stations, lunar scientific probes, communications and meteorological satellites, and a host of additional terrestrial, marine, and space systems.

Present indications are that by 1970 many electrical kilowatts of radioisotope power will be required annually. Consequently, the Atomic Energy Commission program has as its ultimate objective the yearly production of perhaps as much as 1,000 thermal kilowatts of radioisotope materials. Tables I and II identify present and planned production goals.

The incredibly large quantities of radioactive materials, possessing appropriate characteristics, which will be required to satisfy isotopic power needs have posed the most severe challenge to the AEC's materials development and production activity since its inception in 1946.

Two major categories of radioisotopes are being considered as heat sources: the beta-emitting fission products separated from gross radioactive wastes generated in the nuclear fission process, and certain very energetic alpha emitters.

It is important to note that several hundred individually identifiable isotopes are produced by nuclear fission but only a relatively few have characteristics suitable for thermal application. These characteristics are:

1. A reasonably long half-life.
2. A high enough yield in the fission process to make substantial quantities available at reasonable cost.
3. A capability of being formed into high-power-density heat sources.
4. A decay which ideally does not involve the emission of high-energy gamma rays.

In the familiar double-humped yield curve for the thermal fission of Uranium 235, the peak occurs at mass numbers of roughly 90 and 140 and isotopes in yields of around 5% will be found in the vicinity of these numbers. Thus, Strontium 90, Cesium 137, Cerium 144 and Promethium 147 are the only radioactive fission products with half-lives greater than six months and less than 100 years that are of interest to us.

The potential alpha emitters, which are produced by specific reactor irradiation, must be selected on the basis of the same general criteria (except fission yield) as the fission products.

~~shows that~~ nly Polonium 210, Plutonium 238, Curium 242, and Curium 244 will meet these criteria. The quantities of these materials which can be produced are, of course, limited only by the availability of target materials, reactor space, and chemical processing facilities.

In almost every instance the elemental forms of the radioisotopes are not considered the most desirable for heat sources. Rather, they are ~~combined~~ made into compounds exhibiting physical and chemical properties required for particular application. Ordinarily, it is desirable to concentrate as much radioactivity as possible in a given volume. ~~and~~ Thus an important criterion in evaluating the usefulness of an isotope compound will be its "power density" expressed in units of watts/cc. This unit in turn is the product of the specific power in watts/g and the material density in grams/cc.

The radioisotopes of interest are summarized in Table III.

The chemical form of the radioisotope must possess other desirable properties if it is to be useful for thermal source applications. Certainly, its melting point should be higher than the maximum temperature it will experience in an operational situation. It should be chemically inert with respect to the materials used to encapsulate it. It should be stable at the expected operating temperatures and not decompose or swell in an unusual fashion. Its thermal conductivity should be high enough to permit escape of the heat generated without undue temperature

differentials through the compound. It should be relatively insoluble in naturally occurring fluids, such as sea water and fresh water if it is to be employed for terrestrial and marine applications; and it should be of a form which is readily dispersible upon re-entry heating if it is employed in space missions. Lastly, it must be a compound or mixture which is readily fabricable by remote techniques in a hot cell.

This discussion of criteria used in the selection of appropriate radioactive materials for thermal applications provides some idea of the constraints placed upon the Atomic Energy Commission both in materials development and in production.

Today the Atomic Energy Commission's Hanford Atomic Products Operation is the only source for fission products in the United States. Until reprocessing plants are built specifically for handling power reactor fuel elements, Hanford will probably remain the only source. Several years ago, when the need for large quantities of separated fission products was recognized, interim facilities for the separation of the four principal fission products (Strontium 90, Cesium 137, Cerium 144, and Promethium 147), were installed at Hanford. These crude cuts are then shipped to Oak Ridge National Laboratory and the Martin Company facility at Quehanna, Pennsylvania, for further purification, processing, and encapsulation. This interim arrangement, nevertheless

gives us an annual capability for producing multimegacurie quantities of final product. At Hanford a new concept of radioactive waste management has been proposed. It would insure the continuing availability of greatly increased quantities of the four principal fission products. These isotopes would be removed from the gross fission product stream and the remaining wastes would be solidified in storage tanks by evaporation. The separated long-lived fission products could then either be purified and processed for beneficial utilization or absorbed on an inorganic ion exchange material and sealed in stainless steel cans for permanent storage. Such a plan would alleviate present difficulties resulting from self-heating wastes in storage caused by the long-lived fission products.

Hanford also now is studying the design of a fully integrated fission products processing, fabrication, and encapsulating facility. Such a facility could convert and package for utilization all the Strontium 90, Cesium 137, Cerium 144, and Promethium 147 generated in the operation of the Hanford reactors. The design study is not complete, but preliminary data indicate that we can make available, annually, well over one-hundred million curies of packaged fission products at only a fraction of today's costs.

In addition to efforts in improving and optimizing fission products production, significant emphasis is also being placed on the development of improved materials. For example, while strontium titanate is an excellent material for terrestrial and marine applications, it does not have the appropriate characteristics for space applications. Research

and development work is underway ~~in progress~~ on the preparation of a Strontium 90 fuel form for future space use. The program is expected to yield a form of strontium that will not only meet the rigid safety requirements set for space nuclear systems but will possess a significantly higher power density than strontium titanate. This latter characteristic is an absolute~~ly~~ one ~~essential~~ for those applications where maximum power to weight ratio is essential.

Efforts are continuing apace in the development of improved fuel forms for Cesium 137 and Cerium 144. The presently accepted product for Cerium 144, ceric oxide, while possessing very high power densities and temperature stability, liberates oxygen on radioactive decay. This poses problems in the design of appropriate encapsulated thermal sources. We have, therefore, a continuing program aimed at the development of appropriate oxygen-free forms of Cerium 144.

~~One of the most technically~~ A particularly challenging effort is the development of technology leading to the production of large quantities of Curium 242. Produced ~~up to the present~~ now in only ~~fractional gram~~ milligram-to-gram quantities for research purposes, Curium 242 has been selected as the isotopic fuel for auxiliary power in the Surveyor spacecraft because of its very high power density and easily shielded radiations. The Surveyor mission, designed to land instruments on the moon's surface in support of the Apollo program, will require tens of grams of the Curium 242 fuel annually. Oak Ridge National Laboratory, responsible for the effort, is now completing the installation of facilities for the production of Americium 241 targets, the solvent extraction separation of Americium 241 - Curium 242 fraction from the fission products, and ~~for~~ the fabrication of thermal sources. Americium 241 will be pelletized, encapsulated, and irradiated

in a high-flux reactor such as the Oak Ridge Research Reactor or the Materials Testing Reactor ~~located~~ at the National Reactor Testing Station in Idaho. The irradiated material will be dissolved and the fission products removed by solvent extraction, but the Americium 241 will probably not be separated from the Curium 242. The final solution will be ~~is~~ further processed into a Curium 242 oxide, which will be ~~is~~ calcined, pelletized, sintered, doubly encapsulated, welded, and leak checked. ~~It is anticipated that~~ The first substantial quantities of material will probably be produced this year with our production capability rising by 1964 to at least 80 grams annually.

The technology being developed under the Curium 242 program will be useful in ~~has applicability to~~ the production of Curium 244, a radioisotope that is ~~which is also~~ particularly interesting as an auxiliary electrical power source for aerospace applications. Again, large-scale production represents a truly pioneering effort. The objectives of the Curium 244 production program are the development of chemical processes for recovering and purifying a Cm-Am mixture from irradiated plutonium targets, the development of the optimum Curium 244 source compound or mixture for specific objectives, and the determination of the characteristics of Curium 244 thermal sources. Although chemically identical with Curium 242, Cm-244 ~~the~~ processing and utilization ~~of Curium 244~~ present unique problems because of differences in feed material composition and specific power generation. Applications of Curium 244 may require its separation from americium, which is not necessary in the case of Curium 242, in order to achieve the required power density. Source compounds and mixtures of Curium 244 will have different requirements from those of Curium 242 for thermal conductivity, radiation emissions, size, shape, and encapsulation.

~~For~~ Both Polonium 210 and Plutonium 238, ~~we~~ have been routinely producing and utilizing the metal for thermal applications. However, the trend in SNAP application is towards higher efficiency direct energy conversion systems requiring isotope sources capable of operating at greatly elevated temperatures. Consequently, our research has been directed towards ~~the~~ developing ~~of~~ high temperature-low vapor pressure compounds of polonium and plutonium and towards welding techniques and materials of encapsulation for high-temperature containment. Although our results are not ~~as~~ yet conclusive, the family of rare earth polonium compounds looks most promising, as do some of the ceramic forms of Plutonium 238.

Safety Testing

The remaining major program activity associated with the Atomic Energy Commission radioisotope production efforts relates to safety testing of sealed radioisotope sources. ~~The purpose of~~ The program ~~is to~~ will provide a fund of information which can be used by source manufacturers, users, and regulatory bodies to help maintain the present high standards of safety in radioisotope applications.

The first objective in the program is to develop a uniform source-classification system based on the mechanical and environmental integrity of the source. A uniform system will give U.S. ~~enable our~~ regulatory groups ~~in the United States to use~~ consistent methods for ~~of~~ defining source characteristics, ~~in their regulations.~~ The classes will be defined wherever possible in terms of numerical values (i.e., impact strength, tensile strength, corrosion rate, etc.) to minimize misinterpretation.

When strength and corrosion values for the various classes of sources are established, standard tests will be developed that can be used to determine the class in which a source belongs. At the same time, simple quality control tests will be developed that can be used by manufacturers to measure their product's quality against the requirements of the source class.

The third objective of the program is to develop a rating system that can be used by both manufacturers and regulatory groups to determine the classes of sources required for particular applications. The system will be based on data obtained by testing a variety of sources now in use that have demonstrated their reliability from a safety standpoint, on experience gained in developing classification and quality control tests, and on results of a survey of environmental conditions existing in various types of source applications.

The survey of environmental conditions has recently been completed by Battelle Memorial Institute under contract to the Atomic Energy Commission. The source classification system should be available by the end of this year.

In the interim, detailed testing of commercially available sources is proceeding at Oak Ridge National Laboratory. Work is expected to start in 1964 on standardizing quality control tests along lines compatible with the classification system and tests. By 1965 sufficient data should be available to begin work on the development of a sealed source rating system.

Another aspect of our safety program is the test evaluation of radioisotope shipping casks and medical teletherapy units to ensure that they adequately meet existing and planned safety criteria. Many drop tests have been conducted during the past year to test the structural integrity of a broad range of shipping casks. The casks are dropped on a pad which consists of a 12 by 12-foot piece of steel armor plate 4.5-inches thick, backed up by a 5-foot thick slab of reinforced concrete. Below this pad is a 3-foot diameter reinforced column reaching down 10 feet into bedrock.

Each cask is instrumented with acceleraometers, strain gauges, and compressometers to measure inner wall deflections. Some of the casks are covered with brittle lacquer material to determine the areas of high stress.

Casks have been dropped horizontally, on end, on a corner, and on pistons. Results indicate the casks will meet free fall safety requirements, although there is some doubt whether the outer shells of casks weighing more than six tons can resist penetration by a 6-inch diameter piston.

Similarly, we are fire testing both shipping casks and shielding devices such as medical teletherapy heads to determine if they meet the standard one hour fire requirement as established by the National Fire Protection Association.

Process Radiation Development

Contrasted to the United States' slow progress during the past decade toward industrial utilization of radiation for manufacture

of chemicals and chemical products, we are today witnessing new gains, new interests, and new effort in developing ionizing radiation for process purposes.

An estimated 50 kilowatts of installed radiation power is now being used for commercial radiation purposes. We are witnessing the emergence on the commercial market of new radiation-produced products. The first industrial use of radiation for bulk chemical manufacture in the United States is now being carried out by the Dow Chemical Company. This involves the commercial-scale production of ethyl bromide, using Cobalt 60 as the source of radiation energy. Other products include radiation-crosslinked polyethylene wrap and insulation, radiation-induced graft polymers, radiation-cured industrial coating, radiation-modified semiconductors, and radiation-sterilized medical supplies. Estimates place the commercial process radiation business at 20 million dollars a year. Listed in Tables IV and V are radiation-processed products now being manufactured in the United States or planned for production.

In the United States today, the government, through the Process Radiation Development Program of the Atomic Energy Commission, is attempting to provide a productive, balanced effort on radiation processing in concert with that of private industry. The AEC's process radiation development program is designed and directed toward:

1. Seeking original radiation reactions, thereby establishing a substantial scientific foundation leading ultimately to production of unique radiation products.

2. Establishing the technical feasibility of specific process radiation systems.
3. Learning to design, construct, and operate large kilowatt radiation processing facilities.
4. Finding ways of cutting radiation costs.
5. Developing large-scale requirements for fission products and other radioisotopes in the process radiation field.

The major areas of program work include:

1. Radiation Engineering - These are studies devoted to problems of radiation source and facility design, fabrication, and construction necessary for the support of general and specific radiation processes.
2. Exploratory Processing Systems - These are studies which are conducted on an exploratory or "scouting" basis with the most promising systems being developed to the point of demonstrating technical feasibility.
3. Advanced Development Systems - These are engineering scale-up projects designed to establish economic feasibility of those processes which show the greatest and most immediate commercial promise.
4. Dosimetry - This comprises studies necessary to provide dosimetric systems for eventual use in a radiation process. These systems can be considered primarily in terms of quality control.

During the past year the Atomic Energy Commission has completed and placed into operational use its Radiation Development Center at Brookhaven National Laboratory. This new Laboratory is one of the world's most

versatile and unique radiation facilities. It will serve as the center for government work on process radiation engineering, the science of handling and using large radiation sources for processing purposes. The laboratory is a unique canal-cell complex. It consists of a preparation cell for building large radiation sources into desired configurations and an experimental cell which will permit mock-up of process systems to investigate such problems as source geometry, radiation dose distribution, and process radiation utilization efficiencies. The Radiation Development Center will be capable of handling several million curies of Cobalt 60 or its equivalent. By this summer it is expected that 600 kilocuries of Cobalt 60 will be assembled and installed in the Center for use. We plan to increase the size of the Cobalt 60 source up to 3 million curies. We are also completing fabrication of a 200 kilocurie Cesium 137 source which will be installed in the Radiation Development Center. As work progresses, other radioisotopes, fission products, and machine radiation sources will be studied and compared.

Research in the Radiation Development Center is intended to provide data leading to a radiation engineering handbook. This handbook will be published in its first edition form by 1964. It will contain information on basic radiation physics, source design and fabrication, computational procedures, safety aspects, health physics, shielding compilations, and economic guidelines.

Our purpose is to encourage use of the Radiation Development Center to provide experience in radiation engineering problems, on a

research participation basis, to government and industry scientists as well as scientists from other countries.

The Radiation Development Center can thus serve as the major focal point for radiation development in the United States.

Within the scope of this paper it is not practical to provide a comprehensive discussion of research and development being conducted under the Atomic Energy Commission's process radiation development program.* Therefore, with the exception of brief identification of overall process work, discussion will be limited to those process radiation systems which now appear most promising for ultimate development.

The overall research and development on process radiation includes the following projects:

1. Studies in radiation technology which consist of theoretical and experimental work on basic and applied radiation mechanisms to aid in the development of large-scale radiation chemical processes.
2. Studies on radiation chemical methods for the preparation of fluorinated organic compounds either as an alternative to classical methods of synthesis, or of those compounds which are not conveniently prepared by conventional procedures.

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*Such a review is being presented at the Conference on "The Application of Large Radiation Sources in Industry," sponsored by the International Atomic Energy Agency in Salzburg, Austria, May 27-31, 1963. Copies of the review paper entitled "Technical Developments in USAEC Process Radiation Development Program," may be obtained upon request to the Division of Isotopes Development, U. S. Atomic Energy Commission, Washington 25, D. C., U.S.A.

3. Induced gelation involving the incorporation of multifunctional monomers into polymers for the purpose of (a) reducing the radiation dose required for attaining incipient gelation and high gel contents and (b) inducing the radiation crosslinking of polymers that normally are degraded upon irradiation. Cross-linking upgrades the properties of polymers by increasing the molecular weight (viscosity), melting temperature, and tensile strength, while decreasing solubility and elongation.
4. Investigation of radiation-induced reactions in swollen polymers, natural and synthetic, to produce novel polymers which apparently cannot be produced by other means.
5. Development of technology leading to the preparation of novel and useful polymers of controlled structure by radiation-induced graft polymerization. The objective is to provide an understanding of the mechanisms involved in radiation-induced graft processes and to create specific polymeric structures of interest to commerce and unique materials for aerospace applications.
6. Radiation-induced gas-phase reactions utilizing Krypton 85.
7. Development of a chemical dosimetry system wherein rupture of the organo-metallic bond occurs proportionately to gamma dosage and a solar cell dosimeter for high-level dosimetry.

Several process radiation systems currently under advanced development within the Atomic Energy Commission program are described below in detail.

Wood Plastic Combinations

Improvement of wood properties has been the objective of many diversified research programs totally apart from nuclear energy development. Among the important areas of study are resistance to decay and insects, dimensional stability, and mechanical properties. By impregnating various woods with monomers and polymerizing using radiation, a wood-plastic combination results that exhibits superior properties in these areas.

The experimental work on radiation produced wood-plastic combinations includes:

1. Determination of the properties of various wood-plastic combinations produced by radiation polymerization.
2. Chemical studies to determine in situ polymerization rates of various monomer systems, characterization of the polymer, and investigation of methods of accelerating the polymerization to permit reduction of total radiation dose requirements.
3. Development of data necessary to control the quantity of monomer absorbed by various wood species and examination of test specimens, to determine uniformity of polymer loading.

Radiation-produced wood-polymer combinations prepared from a variety of woods and monomers exhibit increased compressive strength and hardness. White pine-polymethylmethacrylate, for example, which contains 0.6 part of polymer per part of base wood has a compressive strength at the proportional limit about 40 per cent greater than that of the unmodified wood.

Although higher polymer loadings do not yield further increases in compressive strength, the hardness continues to increase with increasing polymer content. A loading of 1.55 parts of polymer yields a pine product having a hardness (measured by ball penetration) 130 per cent greater in the axial direction, 450 per cent greater in the tangential direction, and 500 per cent greater in the radial direction than unmodified white pine. A specimen containing 0.75 part of polymethylmethacrylate absorbed 40 grams of water when immersed for 20 days while a control absorbed 110 grams.

Birch and white oak combined with polymethylmethacrylate and polyvinylacetate were as much as 200 per cent harder than unmodified controls, and compressive strength at the proportional limit showed a marked increase. For example, white oak-polymethylmethacrylate containing 0.25 part polymer was 50% stronger than the unmodified control.

In general, wood-plastic combinations exhibit increased hardness and compression strength and absorb water more slowly, thereby reducing changes in dimension due to varying environmental conditions.

Incorporation of a rigid, unyielding material into the wood structure can be expected to increase brittleness. Qualitative observation indicates that this is true in many of the wood-plastic combinations prepared. The testing program is being extended to include measurement of this property.

The rates of radiation polymerization of several monomers impregnated in wood have been measured, and the time required to effect complete polymerization has been determined in some cases. For example, a dose

of 6×10^6 rads, applied at the rate of 1.3×10^5 rads/hr, was found to effect complete polymerization of methyl metharcylate in white pine. Various additives are being tested to decrease the total dose requirements. Carbon tetrachloride allows a considerable decrease in dose when used at concentrations of 10% in styrene and methyl methacrylate. Additives also receiving attention include ethylene glycol dimethacrylate, polyethylene glycol dimethacrylate, tetraisopropyl titanate, and divinyl benzene. Density measurements provide a rapid and accurate method for determining the concentration of polymer formed in the wood. The impregnation conditions necessary to obtain desired loadings of polymer in various wood species are being studied. Reasonably good control can be obtained by properly specifying the vacuum pretreatment and the length of soaking time.

Future work will include:

1. Characterization of the physical properties of additional wood-plastic combinations. The testing program will be expanded to include shear strength, static bending, toughness, and, if possible, decay and insect resistance.
2. Development of additional data on radiation polymerization rates of additives intended to decrease dose requirements.
3. Specification of conditions necessary to effect the desired extent of impregnation of selected species of wood.

Hydrogenation of Coal

Conversion of coal to gasoline and oil products by an economically competitive process would open up a vast new market for the coal

industry. Technically, such conversion can be accomplished by hydrogenation of coal. One way this is done involves coal extraction to obtain a meltable organic fraction (80-85% of coal), followed by hydrogenation over conventional contact catalysts at high hydrogen pressure and elevated temperature, to yield the oil products.

A serious expense in the present technology is replenishment and regeneration of the contact catalysts used for hydrogenation. The catalyst is inactivated by metallic "poisons" in the extract and under some conditions by carbon laydown. Since radiation cannot be diminished by the above factors, radiation catalysis of the hydrogenation step provides sufficient merit to warrant specific investigation. Accordingly, a full research program has been established.

In this research a 90-curie Sr^{90} source is used inside a 10,000-psi 500°C heavy steel-walled reaction vessel. Pure beta-emitting radioisotopes are preferred over gamma emitters to obtain good absorption efficiency in the reaction vessel. The radiation source was specially designed to withstand the conditions of elevated temperatures and pressures in the reactor, to permit beta irradiation of the reactants, and to avoid radiocontamination of products. This radiation processing unit is operated on batches,with the feed (usually melted extract-tetralin solutions) pumped into the reaction vessel, held at 5000 psi hydrogen pressure at 380-430°C, followed by drainage of the liquid products from the vessel. Appropriate safety precautions such as the contaminant cell, barricades, and remote radiation monitoring are used.

Ten radiation processing test runs have been completed so far, as have comparative nonradiative runs. The residence time for the reactions was 1 hr, with a dose rate of 0.98 Mrad/hr. Results on the coal extract from two runs appear in Table VI.

The first row of data shows the conversion yields and hydrogenation (7.39% H in feed, raised to 10.37% in products) obtainable with a fresh contact catalyst. The last column shows that hydrogen addition is less in runs with radiation than without any catalyst at all. This may be due in part to inefficient mixing with the radiation source in the vessel. In any case, hydrogen addition is not accelerated by radiation in all the runs made. Despite this, the yield of desirable oils in the boiling range 100-400°C, is seen to be somewhat higher for the last radiation run shown, than for the noncatalyzed, thermal reaction run. The yield of 250-400°C oil seems definitely higher. Tentative explanations are that radiation may be promoting hydrogen transfer from the tetralin solvent, or radiation is promoting selective "cracking" of the extract to the 250-400°C oil without much hydrogenation. The third set of data in Table VI shows that hydrogen gas saturation diminished the yield of 250-400°C oil, so that the yield in the last run may also be partially due to less inhibition of the cracking reaction by less hydrogen in solution. Results of radiation runs under the same conditions on the model aromatic compounds biphenyl and methylnaphthalene show no radiation-induced hydrogenation in the liquid phase in excess of that obtained by thermal reaction at 429°C. No tetralin was present in these runs.

Liquid phase hydrogenation of pure aromatic model compounds is not measurably promoted in excess of the thermal reaction at elevated temperatures. Likewise, hydrogen addition to coal extract is not promoted by radiation alone. However, some increase in production of desirable oils from coal extract was observed when radiation was used and when the reactants were not saturated with H_2 gas. Since the lack of dissolved H_2 may also have affected this "cracking" oil yield favorably, additional experiments are needed to isolate the effect of radiation alone. Then a more exact conclusion can be made about the specific benefit of radiation.

Radiation runs on coal extract-tetralin solution, without hydrogen gas pressure, will be conducted to measure the effect of radiation alone on the H transfer or cracking production of 250 – 400°C oil from the extract. This can be a very useful selective reaction which could be extended by increased dose if proved. Test runs with both radiation and disposable homogeneous or contact catalysts will also be made.

Transmutation Doping of Semi-Conductor Devices

All semi-conductor devices, e.g., transistors, diodes, microelectronic circuits, consist of electronically active impurities (denoted N or P type, depending on whether they contribute Negative electrons or Positive holes) deposited within a semi-conductor crystal in patterns of various degrees of spatial complexity. The conventional fabrication processes diffuse these impurities into the crystal from the outside.

In neutron transmutation doping these electronically active impurities are inserted into the semi-conductor crystal in a spatial pattern by means of neutron irradiation. A nuclear transmutation reaction occurs in

which a semi-conductor nucleus (Germanium 70, for example) absorbs a neutron and is transmuted to a new nucleus (e.g., Gallium 71, a P type dopant in germanium) which is electronically active in the semi-conductor. The spatial pattern of this impurity deposition is controlled by completely enclosing the semi-conductor within a "radiation die." This "radiation die" is a thin box-like structure composed of thermal neutron absorbing material, e.g., boron or cadmium, with a pattern of slits to permit neutrons to enter into the semi-conductor in a geometry determined by the slit arrangement. The work on this project consists of the detailed development of the technology of the neutron transmutation doping process, an experimental proof of the principle, and the application of this technology in semi-conductor device fabrication.

The analyses performed to date indicate that a variety of simple and complex devices, including many with unusual performance characteristics, can be fabricated at reasonable cost by this process. These include diodes, transistors, microelectronic circuits, solar cells, and thermoelectric elements. Doped configurations of unusual and useful shapes can be produced, and wide barrier regions can be made.

The "radiation die" can be made at low cost and with considerable flexibility when cadmium is used. Silicon carbide devices, which cannot be doped by conventional methods, can be prepared by neutron transmutation doping.

Semi-conductor specimens are enclosed in "radiation dies" with appropriate slit patterns. These assemblies are then "canned" and irradiated. The resistivities and other appropriate electrical characteristics are measured before irradiation, and suitable surface treatment

and passivation performed. After irradiation, the specimens decay (for about two months) until their activation is negligible. They are then annealed to remove gamma radiation dosage effects. Dislocation density is measured by microscopic examination of surface etching effects, both before and after annealing. The specimens are then tested for resistivity and for their device performance characteristics, to determine if the predicted device performance can be obtained. Devices to be fabricated include diodes, transistors, solar cell elements, and microelectronic circuits of germanium and silicon.

Analyses and calculations have been performed to determine the effects of "radiation die" parameters, slit geometry, neutron flux levels, and reactor location parameters on the characteristics of the device produced. Neutron irradiation will be carried out in the Oak Ridge Research Reactor. Work is now in progress on the fabrication of the "radiation dies," the irradiation assembly, and specimen preparation.

The principal results to date are

have

1. Design data ~~has~~ been developed for predicting the device characteristics from the process parameters, e.g., "radiation die" and slit characteristics and neutron flux levels.
2. Data on fabrication methods for the "radiation die" ~~has~~ have been obtained. Both boron and cadmium can be suitably fabricated into "radiation dies" with the required characteristics. Cadmium is particularly convenient since it is easy to fabricate, and its poorer epithermal shielding characteristics can be circumvented by appropriate design.

3. Heat transfer experiments have shown that the semi-conductor temperature can be held below 250°C during the irradiation process if the die holder and die are fabricated to close tolerances. Impurity diffusion is thereby avoided.

The first specimens should be ready for irradiation soon. A number of experiments are planned for proof-of-principle, and to fabricate and test a number of progressively more complex devices (including test of their performance characteristics) in order to demonstrate this process's utility.

Polymerization of Ethylene. During the past decade several studies have been made on the Cobalt 60 gamma-radiation-induced polymerization of ethylene. Because of the potential importance of a Cobalt 60 based process, additional work has been initiated to increase the knowledge of this system's behavior in a radiation field.

The present research program is concerned with measurement of the reaction yield and rate of ethylene polymerization in a Cobalt 60 gamma field. These data were obtained by utilizing a continuous pressure recording technique. The conditions under which the rate was measured varied in pressure from less than 68 to 680 atm, and in temperature from -78°C to 40°C. Polymerization takes place in the liquid phase at -78°C at a very low rate. In the gas phase, at 20° to 40°C, a strong inhibition and retardation effect due to oxygen impurity was noted. The induction period as a function of oxygen concentration was determined. The experiments indicate that the effect of the presence of polymer on the polymerization rate is smaller than the effect of oxygen and ethylene pressure.

The differential G values, based on monomers alone, were determined, and attempts were made at expressing these values as a function of ethylene pressure and radiation intensity. In the range of 80 to 350 atm and 21,000 to 132,000 rad/hr, the rate is proportional to the 0.93 to 1.13 power of the pressure, and to the 0.33 to 0.46 power of the intensity in the falling pressure period. At 133,000 rad/hr a minimum differential G value of 15,000 was obtained. At 21,000 rad/hr a maximum value of 47,000 was reached. Additions of solid substrates of alumina and molecular sieves increase the G value by a factor of 2.2 to 2.5 and the rate by a factor of 1.6 to 1.9, indicating a surface effect.

All the polymer produced in this work by gas phase radiolysis was a fine white powder In the homogeneous experiments the entire vessel was filled with polymer. The polymer could be removed from the vessel in a coherent mass and had various bulk densities depending on the percentage conversion. Under a microscope the polymer powder showed agglomerates of submicron particles.

Because of the varying pressures during the formation of the mass, it is not possible to obtain a quantitative evaluation of the polymer as a function of pressure. However, qualitative effects can be obtained.

1. The density measurements, falling in the range 0.93-0.94 g/cc, indicate a medium density polymer. Annealed measurements show values of about 0.95 g/cc. Amorphous polymer has a density of 0.86 and fully crystalline material, 0.99 g/cc.

2. The reduced viscosity measurements indicate that the molecular weight increases with decreasing total dose. The average molecular weight for polymers formed up to 250 atm falls in the range 80,000 to 300,000. For the liquid phase polymer, the intrinsic viscosity measurements indicate that the material is of low molecular weight.
3. The addition of argon decreases the molecular weight.
4. The degree of branching, indicated by the number of methyl groups per thousand carbon atoms, decreases with increasing pressure, decreasing dose rate, and decreasing total dose. The range of values for initial pressure of 650 atm is indicative of medium-density high-pressure conventional material.
5. The polymers which were fused before removal from the vessel were crosslinked with the exception of those produced at low total doses, which tend to be non-crosslinked. Most of the polymers removed from the vessel without melting showed a high degree of solubility in decalin, and indicate less than one crosslink per molecule. It appears that the solid polymer contains residual radical species, which result in crosslinking on heating. Exposure of the unreacted radicals to the atmosphere, on the other hand, can cause termination of radicals due to oxidation.
6. The melt flow and melt index indicate the processibility of the polymer. The radiation-produced polymer has a lower melt flow index and a higher melt index than the low-density polymer. The melt index for conventional high-pressure polyethylene is of

the order of 1.7 and the melt flow for high molding is about 50 to 60.

7. The material has a high crystalline melting point.
8. The decalin soluble non-crosslinked polymer could be made into a pliable film by hot pressing.
9. Low melt flow index, low branching, and non-crosslinked material are indicative of a higher molecular weight linear polymer. The type of polymer produced is dependent on combined conditions of temperature, pressure, intensity, and total dose.

Further characterization and evaluation of the polymer await experiments at constant pressure, both in static and in flowing systems. Work is continuing to determine the quantitative effect of the various parameters on the rate of polymerization.

Textile Modification

Under this program, textile processes in two general areas have been investigated. These included the use of various nuclear radiations to modify fiber properties and the use of graft copolymerization to modify the chemical structure of fibers. The results of three and one-half years of research have shown that the most promising field for commercial application is in the area of fiber modification by radiation-induced graft copolymerization. Means of overcoming fiber deficiencies and thus producing materials of more functional or aesthetic value by this procedure are being investigated.

New uses for old fibers will be found and

fibers will be upgraded in their performance characteristics. In the work to date emphasis has been placed on procedures for getting satisfactory grafting at levels of radiation where fiber deterioration due to exposure to the radiation itself is minimized. The approach was to expose a number of different textile fibers to vinyl monomers in the vapor phase, using a third component in the system to accelerate the grafting process. The vapor phase technique gave grafted products of good uniformity, and for this reason work was largely confined to this system.

The fibrous materials that are the least stable to radiation are the ones that were grafted most readily. Thus, the best results were obtained with cellulosics, cellulose esters, polyamides, and polypropylene. Polyesters and acrylics showed little tendency to graft. The best vinyl monomers investigated were acrylonitrile, methyl acrylate, ethyl acrylate, and butadiene. The presence of water in the system was necessary for the grafting of all these monomers to the cellulosics and the polyamides except the cellulose esters and polypropylene. Other factors that affected the rate of grafting were the concentration of the monomers in the vapor phase, the presence of oxygen, and the temperature of the system. Generally, the amount of grafting increased with increased monomer content; oxygen had an inhibiting effect, presumably by acting as a scavenger for free radicals; and the effect of temperature was to decrease the amount of graft at any given radiation dose.

Fibers of the different chemical types were partially evaluated after grafting with four different levels of the four vinyl monomers mentioned. Stress-strain properties, moisture regain, solubility, softening point,

and weather resistance of a few selected materials were determined. The cellulose ester grafted with acrylonitrile behaved somewhat differently than the other fiber types. Its breaking strength was unchanged as a result of grafting, but the elongation of the fiber decreased. This resulted in an increase in modulus. Moreover, the yield point was increased as the amount of graft copolymerization increased. The overall data indicated that cellulose esters were crosslinked as well as grafted by acrylonitrile. Generally speaking, the other fiber types showed a slight deterioration in physical properties when grafted with the different monomers, although in no case was the deterioration severe. The moisture regain of all fiber types with all monomers was decreased. The solubility of the grafted materials depended on the monomer used. The addition of acrylonitrile tended to insolubilize all fiber types in their common solvents. Greater solubilities were generally obtained when ethyl acrylate was used as the monomer. There was a level of grafting for each monomer on each fiber type that resulted in insolubility. The addition of the monomers to the thermoplastic fibers failed to increase their softening or melting point except in the case of acrylonitrile. The addition of this monomer to a cellulose ester and to polypropylene gave a material that charred without melting. Cotton grafted with as little as 3.25% acrylonitrile was highly resistant to cellulose-degrading microorganisms. The addition of styrene to nylon appeared to improve weather resistance.

Concurrently with the above work, experiments were initiated to correlate both the kinetics and diffusion for the vapor-phase technique on grafting acrylonitrile to cellulose acetate, nylon, polypropylene,and

polyester fibers. The kinetics of polymerization and the rate of diffusion of monomer both play a very significant part in this process, and accurate data are necessary for the optimum design of graft-polymerization processes. Reaction velocity constants were determined for propagation, termination, and reaction with oxygen. The diffusion of acrylonitrile monomer did not obey a simple mechanism.

Future work will involve continued studies of the kinetics and of the role of diffusion of monomers into the fiber substrates using computer techniques. The evaluation of important textile fiber properties will be continued. Some preliminary work has indicated the possibility that cotton cellulose might be made more dimensionally stable by the use of a radiation process.

Radiation Pasteurization of Foods

Research on radiation preservation of food is being carried out in many countries including the United States and Japan as well as a number of countries throughout Europe. Major developments have been achieved during the past year contributing to the steady advance on radiation preservation of foods technology. Potatoes irradiated to 10,000 rads for control of sprouting have been approved for human consumption in Canada. Last fall more than a million pounds of potatoes were irradiated in Canada as part of a field "test" program to establish better potato response to radiation treatment, process and storage conditions, and economic and consumer acceptance. The Russian Ministry of Public Health has reportedly authorized the use of irradiated potatoes by consumers, and an experimental production facility to irradiate up to 25 tons of potatoes per year is planned.

In the United States radiation-sterilized bacon was approved for human consumption by the Food and Drug Administration in February, 1963.

A formal petition for clearance of wheat and wheat products treated with radiation for disinfestation purposes has been submitted to the Food and Drug Administration

The U. S. Army Radiation Laboratory, Natick, Massachusetts, was completed in 1962 and used for studies on radiation sterilization of food products important to the military establishment. The facility was designed and constructed by the Atomic Energy Commission for the Department of Army. The Laboratory contains the world's largest Cobalt 60 radiation sources of approximately 1.3 million curies, a high-energy 24 Mev 18-kw linear accelerator, and a complete food preparation area.

The Atomic Energy Commission research program on radiation pasteurization of foods aims at developing the technology for providing fresh-like fish and fruits far from harvest areas, thereby opening up new markets for these products. Relatively low radiation doses (less than 500 kilorads) are being used to inhibit bacterial growth for periods ranging from days up to several weeks under normal refrigerated distribution and storage conditions for perishable foods. The low-dose radiation processing of foods program of the AEC is complementary to the radiation sterilization of foods program of the Department of Army where high doses of radiation are used to completely destroy bacteria and permit food storage without refrigeration for months.

The AEC research and development program is limited to selected fruit and fisheries products and emphasizes factors such as storage and shelf-life extension, treatment variables including radiation dosage, quality including organoleptic effects, wholesomeness, and packaging materials studies. In addition, continuing consideration is being given to marketing and consumer acceptance studies. The selected food items under study are:

Fruit: strawberries, citrus, peaches, grapes, pears, cherries, and tomatoes. (Preliminary experiments have been carried out on figs, pineapples, papayas, asparagus, and nectarines.)

Fish: haddock fillets, soft-shelled clam meats, Pacific crab meat, Petrale sole fillets, and shrimp. Recent work has been initiated on cod, pollack, and ocean perch; however, results are not available for reporting at this time.

Radiation for control of sprout inhibition, disinfestation of grain, and alleviation of quarantine problems is also being considered.

Research results achieved to date with both fruits and fisheries products show continuing promise for successful development of radiation pasteurization technology and for ultimate commercialization of the process.

Progress on specific fruit products:

1. Strawberries: The amount of decayed fruit after a simulated transit and marketing period is considerably less in fruits irradiated at doses of 200 kilorads than in the control lots. The texture of the irradiated fruit is altered less than that of other

fruits. This derives in part from the fact that strawberries, both irradiated and control, become progressively firmer in cold storage. Taste panel studies reveal that irradiated strawberries are not significantly different in flavor to non-irradiated fruits. Color, aroma, and texture are significantly different in irradiated fruits, but are not objectionable to the taste. Ascorbic acid levels in irradiated strawberries are not altered by a nutritionally significant amount.

2. Lemons: Severe injury symptoms were found in irradiated lemons when the fruit was stored for periods of a month or more. Air pockets develop in the fruits adjacent to the lobe membranes. Ascorbic acid content declines by 50 per cent in fruits subjected to 200 kilorads and the "buttons" are destroyed, making the fruit very susceptible to *Alternaria citri*.

3. Oranges: This species does not show the adverse effects observed with lemons. Air pockets do not develop, the ascorbic acid level in irradiated fruit is not significantly different from that of the controls, and the "buttons" are not adversely affected. There is a change in flavor at 200 kilorads with increasing storage time. With two months' storage, this changed flavor is [illegible] acceptable.

4. Peaches and Nectarines: The capacity of these fruits to ripen is not altered by irradiation. They are subject to dramatic effects by radiation on the texture of the flesh. Some varieties of these species have shown a drop in flesh firmness

of 70 to 85 per cent at 200 kilorads.

5. Grapes: White varieties of grapes are more susceptible to adverse radiation effects than are red varieties. Maturity of fruit has a profound effect on its response to radiation. As the fruit becomes riper, the adverse effects of radiation are less. The Thompson Seedless variety shows moderate to severe textural changes at doses which may prove effective against *Botrytis* and *Rhizopus*.

6. Bartlett Pears: Ripening of this fruit is clearly inhibited at doses of 250 kilorads and higher. There is an immediate softening of the flesh at doses as low as 100 kilorads, and at 300 kilorads the firmness of the flesh is only 50 per cent of that of the un-irradiated fruit. Subsequent to the initial decrease in firmness, fruits irradiated at 250 kilorads and higher soften more slowly than the control lots. Fruits subjected to ripening inhibiting levels of radiation in air, are "mealy" when ripe and do not develop full yellow color. Irradiated fruits respond more slowly to ethylene treatment than do control fruits.

When pears were irradiated in a nitrogen atmosphere, their color development, texture, and flavor were more nearly normal, indicating that the adverse effects noted above may be an indirect effect of high ozone concentration in the fruits rather than an effect of radiation *per se*.

There is a spurt of ethylene production by irradiated pear fruits immediately after treatment. Thereafter, ethylene production by fruits subjected to 300 and 400 kilorads is much less than that of un-irradiated fruits. Fruits subjected to 100 and 200 kilorads produce considerably more ethylene than do the control fruits, suggesting that with this species there may be a very narrow range of doses between adverse and non-harmful effects on ripening by irradiation.

7. Sweet Cherries: Cherries have been studied in radiation physiology for the past three years. During this time casual observation and tasting of the fruit have given promise that these fruits can well tolerate radiation doses up to 250 kilorads. It is also reasonable to assume that such a dose would result in an important extension of the post-harvest life.

8. Tomatoes: No retardation of ripening has been found at doses below 200 kilorads. Doses between 200 and 400 kilorads retarded ripening and decay development, but this range of dosage gave an undesirable effect on flavor.

9. Figs: Mission figs seem to respond favorably to radiation. While no inoculations were made, irradiated fruit showed much less decay than did control lots. There were no apparent ill effects of doses as high as 300 kilorads.

10. Pineapples: These fruits respond very favorably to radiation. At 200 and 300 kilorads there is a slight bronzing of the exterior color of the fruits. Flavor of the irradiated fruits seems the same or slightly improved over that of control fruits.

11. Papayas: Fruits were shipped by air from Hawaii and subjected to doses of 200 and 300 kilorads. The fruits seemed to ripen more slowly than unirradiated fruits. There was no evidence of adverse effect on flavor.

12. Asparagus: The post-harvest elongation of asparagus spears was reduced by 66 per cent and tip curvature by 80 per cent at doses as low as 5 kilorads. There were no adverse effects on quality of the product at doses as high as 150 kilorads.

Progress on specific fisheries products:

1. Haddock Fillets and Soft-shelled Clam Meat: Research results show conclusively that at the optimum radiation dose levels of 250,000 and 450,000 rads, respectively, haddock fillets and soft-shelled clam meats can be kept satisfactorily for 30 days at 33°F. At these optimum dosage levels no noticeable "irradiation" odor is detectable in the products. An increase in storage temperatures from 33°F to 42°F decreased shelf life by about 50%. Results have not yet demonstrated a difference in product quality due to use of vacuum or air packaging.

2. Pacific Crabmeat: Radiation pasteurization of Pacific crabmeat packed in cans increases the storage life at ice temperatures by 5 times, or from 7 days to 35 days. Radiation dose levels of 200 kilorads to 350 kilorads appear to be the optimum. Quality of the product remains excellent.

3. Petrale Sole Fillets: 400 kilorads appears to be the optimum radiation dose. A 7-fold extension of shelf life, 5 days to 35 days, was attained at 33°F in the vacuum packed samples. The storage

life of this product was half as great at 42°F as compared to 33°F. In air-pack samples, processed under the same conditions, an off-flavor, described as rancid, developed.

4. Shrimp: The shelf life of radiation-processed shrimp held at 33°F, and using a radiation dose of 100 kilorads, was doubled from 20 days to 40 days. At this dose level, organoleptic and chemical results were consistently better than those of the unirradiated samples.

Cooked irradiated shrimp appear to have excellent keeping qualities as shown by bacteriological, chemical, and organoleptic tests.

A valuable finding is that "black spot" (a melanosis resulting from the action of phenol oxidase in shrimp) is virtually eliminated in radiation-processed products.

9 Design and Fabrication of Irradiation Facilities.

Three research irradiators have been completed. Each utilizes approximately 30,000 curies of Cobalt 60 with a specific activity ranging from 7 curies per gram to 18 curies per gram. These irradiators are designed with a built-in flexibility to permit the performance of irradiations under various types of atmospheres, as well as under a temperature variation from 10 - 150°F. The gamma flux for each has been measured to be about 2.5×10^5 rad/hour, and the irradiation chamber is capable of handling a package measuring 14" x 18" x 6".

Transportable irradiators are designed for use in semiproduction-scale acceptability, storage, and shipping tests. The transportable irradiator provides a radiation processing capability close to the harvest areas as well as data on process parameters and economics.

Design concepts for two transportable irradiators have been completed. One will use a 100-kilocurie Cobalt 60 radiation source and the other, a 2-Mev 3-kw electron machine radiation source. These irradiators have a designed minimum production rate of 500 pounds per hour at a radiation dose of 250 kilorads.

The Marine Products Development Irradiator is designed to demonstrate the technical and economic feasibility of radiation pasteurization of fishery products on a near-commercial scale. It will use a 235,000-kilocurie Cobalt 60 source and will have a production rate of 1,000 lb per hour at 500 kilorads. The facility will be located at Gloucester, Massachusetts, which is the center of the east coast fish-packing industry in the United States. The facility will be operated for the Atomic Energy Commission by the Department of the Interior, Bureau of Commercial Fisheries. Construction on the Marine Products Development Irradiator will begin this summer and it is expected to be completed in July 1964.

Radioisotope Technology Developments

This phase of the Isotopes Development Program is concerned with creating new and improved technology leading to beneficial applications in areas such as water exploration and management, atmospheric pollution, meterology, oceanography, and crime detection. Research and development are being carried out on problems involving neutron activation analysis, radiometric analysis, low-level process control, radiation absorptiometry, ionization technology, non-destructive testing, and isotope measurement systems. Emphasis in the research and development work is directed primarily to filling critical gaps in technology which are acting as major deterrents to advancement of the use of radioisotopes.

While important contributions have been made in each of the above-mentioned areas, discussion will be limited to work of most timely interest and significance.

Neutron Activation Analysis. Neutron activation has moved out of the research laboratory into broad application throughout science and industry in the United States. Some 25 private organizations now offer neutron activation as a commercial service. The method is beginning to find wide use in the chemical and petroleum industries.

The development of new instrumentation, particularly in neutron generators, multichannel analyzers, and detectors, continues to accelerate. The rapid developments in instrumentation have opened up new areas for the investigator, especially in the utilization of fast neutron reactions. This has revealed gaps in the basic

knowledge which must be filled.

The Atomic Energy Commission is continuing its strong interest in activation analysis by supporting a broad range of research and development projects. Fundamental studies are being carried out on gamma ray spectra of neutron-deficient nuclides as well as on solid-state detector systems which will provide superior beta and low energy photon resolution as compared to crystal detectors. Development of instrumentation systems to provide rapid, automatic analyses of very large numbers of samples is an important area of work, as is development of improved neutron sources. Much effort is being placed on applying neutron activation analysis to problems of major national importance, including analysis of environments in space and in oceanography.

1. Analysis of the Lunar Surface - The Atomic Energy Commission and the National Aeronautics and Space Administration are jointly sponsoring a program to develop a workable, remote, activation analysis technique for elemental analysis of the moon's surface. It is planned that a neutron source and detector will be included as part of the instrument package of a scientific lunar landing mission. A small area of the moon's surfact will be neutron-irradiated, the induced gamma radiations will be detected and the data will be telemetered to earth. The data will then be analyzed by an automatic computer-coupled

system which will identify the major chemical elements in the moon surface sample.

A working system has been operated successfully under simulated remote conditions. The activator is a neutron tube which produces a flux of 5×10^8 neutrons/cm^2/sec at the target located at a distance of about 0.1 in. from the end of the accelerator unit. This flux is barely adequate and a more powerful source is being considered. The detector unit is a 3" x 3" NaI (Tl) scintillation crystal, photomultiplier tube, a pre-amp, and a lead collimator. The control unit powers the accelerator, controls pressure automatically, and programs the analysis sequence. This involves activation by the neutron source for 5 minutes, rotation of the assembly to place the detector over the activated sample for 10 minutes, and transmission of the data signals to the earth-stationed multichannel analyzer.

Tests have been conducted with simulated moon matrices, using a mixture of aluminum oxide (Al_2O_3), ferric oxide (Fe_2O_3O), magnesium oxide (M_gO), and silicon dioxide (SiO_2). The gamma radiation was analyzed with a 400-channel transistorized analyzer, and photopeak counts for the various components were taken from a plot of channel number versus activity. Spectrum-stripping techniques were used to

resolve peaks containing activity from more than one radioactive source.

Data from an actual moon shot are to be processed with the Mark II automatic gamma analyzer, on command from the moon probe. This analyzer is a development of the Mark I, previously reported at this Conference in 1961. Advances in Mark II include faster operation by direct coupling of computer tape, elimination of some interfering elements by a "dynamic subtract" unit, and improved resolution of gamma peaks.

2. Shipboard Neutron Activation Analyzer - The shipboard analyzer performs rapid elemental analysis on core samples of the ocean floor on the vessel as they are collected. This aids effective exploration by making it possible for the ship to modify its survey pattern in accordance with the trends indicated by the data.

The unit was developed in conjunction with Lamont Geological Laboratory of Columbia University. The special neutron source is a small, rugged (d,T) neutron generator which emits 10^8 neutrons/second. It is contained in a lead and paraffin shield designed to permit insertion of an entire ocean-bottom core sample so that intervals along the core length may be activated without cutting the core. A shielded 3" x 3" NaI (Tl) scintillation crystal is used to observe the activity induced in the core and the gamma spectrum is analyzed in a 400-channel analyzer. The analysis of cores for their content of silicon, aluminum, magnesium, sodium, and iron is

made by neutron activation and for their contents of potassium, uranium, and thorium, by the spectra of their natural radioactivities. The activation conditions are: 10 min for core irradiation, 1 min for transfer of the core to the detector, and 10 min for measurement of the induced activity. Analyses of standard samples have indicated the following sensitivities, in mg/cm^3: Al and Si, 0.3; Mg, 1.6; Na, 2.2; Fe, 3.8.

3. Coal Analysis and Grading - In cooperation with research groups of the coal industry, the Atomic Energy Commission is sponsoring investigations into the feasibility of using activation analysis to continuously measure the Btu and ash content of coal. By the use of activation analysis using a neutron generator emitting 14.5-Mev neutrons combined with the related techniques of inelastic scatter and capture gamma-ray analysis, it appears to be possible to analyze for the elements that determine the Btu value of coal, i.e. C, N, O, and H; the water content; the sulfur content; and ash. The data for the several quantities are fed into a computer for automatic calculation of the Btu. It is anticipated that such units can be installed at conveyor belt lines carrying the finished coal. These can be coupled to automatic sorting equipment.

   Carbon content is determined to an estimated accuracy of ±1% using inelastic scattering of fast neutrons. Oxygen content is determined to ±2.5% using neutron activation analysis. The ash constituents, Si, Al, and Fe, are determined by activation to within about ±5%. Methods for S, N, and H are under development.

4. Automated Activation Analysis in Metabolic and Environmental Studies -

Determination of the role of trace elements in metabolism and its correlation with the incidence of disease and existent chronic deficiency conditions is of major interest to medical research in the United States. The analytical problem in this research is one of tremendous scope inasmuch as a serious study should include analysis for several elements in sufficient samples to investigate statistically the major environments across the nation and to establish the trace element content of all the life-sustaining portions of these individual environments. This includes the atmosphere, the drinking water, and the food supply. Ideally, seasonal changes should also be investigated. Further, any specific effect in a particular area such as coal dust in a coal-mining region, for example, must be covered intensively. Clearly, such a program poses a monumental analytical problem, involving hundreds of thousands of samples.

Previous studies of trace element effect on life processes have been limited to specific elements and usually to specific geographic areas where the effect of a particular deficiency or excess was conspicuously apparent. Studies of fluoride effect on teeth and toxic selenium effects are examples.

Neutron activation has provided the first general analytical approach that appears to be economically capable of coping with the enormous task. Automated computer-coupled analysis in environmental health studies is now underway under joint sponsorship of the National Institutes of Health and the Atomic Energy Commission. Elements selected for initial studies are silica, selenium, and tellurium. The neutron activation technique is particularly advantageous for

determination of selenium. This is a difficult analysis by classical methods, involving considerable risk of losing selenium in the ashing process. The selenium activation technique is nondestructive, rapid, and sufficiently sensitive.

5. Activation Analysis in Law Enforcement - Activation analysis is particularly suited to the chemical analysis of material evidence taken in law enforcement investigations because it is nondestructive and the sensitivity is adequate. The Atomic Energy Commission has been conducting extensive studies in this area, both at its Oak Ridge National Laboratory and at private laboratories. Much of the work at Oak Ridge National Laboratory has been conducted in close consultation with the U.S. Dept. of the Treasury. It has included such diverse problems as identification of automobile paint, soot, drugs, soil samples, hair, and fingernails.

6. Compendium of Activation Analysis - Under the AEC program, continuing attention has been given to providing a standard reference compilation of information on neutron activation analysis. The publication of standard reference atlases of gamma spectra has been completed. A compendium on neutron activation analysis, which is a companion to the standard gamma atlases, is nearing completion at the Oak Ridge National Laboratory. The compendium provides the analytical chemist with needed information for determining the pertinence of activation to his particular problem (with the standard spectra atlases), and most of the information needed to begin work in activation analysis.

While the procedures detailed in such a reference are quickly

outdated, the modification or amplification of the basic material is much slower. Supplements will report significant developments as they achieve the status appropriate for inclusion in a reference work.

High-altitude Atmospheric Density Sonde. A direct-measuring balloon sonde to telemeter density information during flight has been developed in cooperation with the U.S. Weather Bureau and the U.S. Air Force. This sonde is intended to replace the indirect and frequently inaccurate calculation of density based on thermistor data.

The device is a forward-scatter gas-density gage using a 10-mc gaseous Kr^{85} source. It has been flight-tested under Air Force supervision. Use of the forward-scatter geometry, which gives a better signal-to-noise ratio than backscatter geometry, and a 14-in. gap between source and detector, provides sensitivity for measurement in the extremely rarified atmosphere up to 140,000 ft. The average error in the interval from 45,000 to 140,000 ft appears to be $\pm 0.05\%$. The instrument contains two scintillation detectors (one to measure and subtract background), a battery high-voltage supply, two amplifiers, a difference circuit, and radio transmitter.

Deep-water Isotopic Current Analyzer ("DWICA"). One of the many problems confronting both the hydrologist and the oceanographer has been the lack of a suitable method to measure precisely and to record accurately direction and flow of slow-moving currents in the range 0.01-1 knot. Now, for the first time, such a method has been developed, based on an isotopic principle.

The principle of the isotopic technique depends on injection of a radioisotope into a moving water environment and measuring drift as the

radioactivity passes a detector. In this system a series of 16 detectors is arranged in a wagon-wheel configuration with a diameter of 2 ft. The detectors completely encompass a radioisotope injection mechanism located in the center of the configuration so that any release of activity will be seen by the detector system. After the radioisotope is injected, the direction and rate are determined by time of arrival of the radioactivity at a particular detector in the configuration.

The isotope method is far superior to previously available techniques for determining current direction and flow rates.

The greatest advantage of this new system is that there are no moving parts to impose friction drag on the system as occurs in the case of a rotor mounted on a sapphire bearing. No matter how well a rotative cup is designed, there will always be a certain amount of friction, while with the isotope method the actual drift of the current is the mechanism by which the radioactive material is transported and recorded.

The first field tests of this instrument were made on Dec. 14 and 15, 1961, in Round Bay, Md., in 19 ft of water. The instrument was placed 13 in. off the bottom, and measurements were made with I^{131}. The tests began at 5:00 p.m. on Dec. 14 and ran until 3:00 p.m. on the 15th. During these 22 hr, measurements were made at intervals of from 6 to 20 min.

A second series of field tests was performed in the same location on Dec. 19 and 20. Measurements were made from 5:30 p.m. on Dec. 19 and ran until 3:00 p.m. on the 20th, at intervals of 1 to 20 min.

The results of the first series of measurements in Round Bay are shown in Fig. 1. In this figure direction is given by the upper curve; note that more than one full turn of the compass is plotted.

Current speed is indicated by the lower curve. Current directions are indicated on the left-hand axis and speeds on the right-hand axis. Between 5:00 p.m. and 10:00 p.m. on the 14th, the current direction rotated in a clockwise fashion through 360°. For the next 10 hr, the current direction remained upstream, about 340°. However, starting about 8:00 a.m. on the 15th, the current direction again made a complete clockwise turn about the compass in approximately 3 hr. Current direction was downstream for the final 3 hr of the measurements.

The results show that current movement upstream has a longer duration than current movement downstream. Further, the data indicate a correlation of the movement with flood and ebb tide, which was predicted by experts of the Chesapeake Bay Institute. However, until these tests were made, there was no method of verification.

This test represented the first successful attempt to log currents with velocities as low as 0.01 knot.

The instrument has been modernized with computer-logic circuits to make it completely automatic. The instrument consists of the following units: (1) an electronic servo-injector control system which automatically adjusts the amount of radioactivity injected at the start of current velocity measurements and makes the necessary corrections, in situ, when the velocity increases or decreases; (2) an automatic digital converter system that prints out the following information: date and time of measurement, drift time of injected activity, and the number of the detectors that observed the activity (i.e., direction); and (3) an automatic program control system which determines the intervals of measurement. This, of course, depends on the velocity of the current being

measured. The present design of the DWICA can measure velocities in the range 0.001-10 knots, and has been used by the Chesapeake Bay Institute to collect data on summer and winter cycles in Chesapeake Bay. The lower limit of the instrument is determined by molecular diffusion of the radioactive material in still water.

The instrument is being redesigned and pressurized to measure ocean currents at 6000-ft depths. The new instrument will include many of the above-mentioned features, as well as several additional ones, i.e.: (1) a compass for proper orientation at great depths; (2) a unit that can generate its own radioisotopes in situ. The generator will produce indium-116 by activation of an indium solution with ten 1-curie Am^{241}-Be neutron sources with a flux of 1×10^6 n/cm^2.sec.

The first field test of the redesigned instrument is scheduled for August 1963, and will be conducted in cooperation with the U.S. Navy Oceanographic Office.

New Improved Tritiation Method for High-molecular-weight Organic Compounds. High-molecular-weight hydrocarbons are now successfully labeled by a new tritium reagent. This new technique produces labeled compounds of a higher specific activity than produced by the radiation-induced self-labeling method of Wilzbach. It is faster and cleaner and requires less initial tritium to produce a product of higher specific activity. The principle of this new technique is based on the hydrogen-tritium exchange brought about in the presence of a tritiated acid catalyst.

The tritiating reagent is a complex of tritiated phosphoric acid and boron trifluoride. It normally has the formula $TH_2PO_4.BF_3$, i.e. the tritium/hydrogen ratio is arbitrary as the complex is easily prepared

by stoichiometric admixture of tritiated water and phosphorus pentoxide followed by saturation with boron trifluoride.

Its applicability is best illustrated in Table VII, which compares this new technique and the standard radiation-induced self-labeling one.

Many advantages are seen from the comparison. Experimental studies of radiochemical purity and tritium activity in the tracers show that the $TH_2PO_4.BF_3$ reagent produces no highly tagged side products, thus obviating extensive purification as required in the radiation-induced method. The method is rapid, and pure, usable tracers are often prepared in a few hours as compared to days by the Wilzbach method. High-specific-activity compounds, up to curies per gram, can be produced when desired by starting with a large tritium content in the reagent. Table VIII shows the theoretical limit reached in a 6-hr test at 23°C.

As seen in Table VIII, labeling takes place on contact under ambient conditions and causes tritium to exchange readily with hydrogen atoms on all aromatic positions and on tertiary carbon atoms; there is no labeling on nonbranched aliphatic carbon atoms.

Kryptonates as Universal Radioactive Indicators

Development of a general, or "universal," radioactive indicator technique, called "kryptonate," which was first reported at this Conference in 1961, has been completely successful. Krypton gas containing 5% of the radioactive isotope is incorporated into the substance by either bombardment with ionized krypton or diffusion under high temperature and pressure. The ionization procedure, which was originally favored, is now used only for selected substances that cannot withstand the pressures or temperatures required of the more efficient diffusion process.

Typical conditions for the diffusion saturation of a substance with krypton are temperatures around 650°C and pressures to 5,000 psi. Several hours to three days are required. The saturation activity that can be attained is a function of the structure of the compound. Substances with numerous interstices of the proper dimensions to contain the krypton atom assume the highest saturation activity. Pyrolytic graphite has been saturated to an activity of one curie per gram, whereas with less favorable structures the activity is frequently limited to millicuries or microcuries per gram. The diffusion method has successfully induced some measurable, though often small, permanent krypton activity in all substances tested so far. Therefore, the title "universal" indicator is still justified.

Uniform incorporation of krypton into a solid is possible if the substance is "kryptonated" in the form of a fine powder which can be subsequently compressed into a solid. The temperature stability of the "kryptonates" varies. Some lose most of their activity at atmospheric pressure when the temperature is elevated only 75 or 100 degrees, while "kryptonated" copper metal retains 75% of its activity to 900°C. Generally, the stability at room temperature is excellent.

The possible applications of the "kryptonates" in science and technology range from pure chemistry to applied mechanical engineering. One of the most important applications is the accurate measurement of atmospheric

moisture, especially at high altitudes. "Kryptonates" appear useful here, as Krypton 85 incorporated into desiccant material is released when these substances absorb water vapor. A research study based on the formation of hydrates to release Krypton 85 and another based on chemical reaction to form the hydroxide, thus releasing Krypton 85, are in progress.

APPENDIX 1

LIST OF RESEARCH AND DEVELOPMENT CONTRACTS
DIVISION OF ISOTOPES DEVELOPMENT
UNITED STATES ATOMIC ENERGY COMMISSION

Process Radiation Development

AIR REDUCTION COMPANY, INC. Murray Hill, New Jersey	Investigation of the initiation of chemical reactions using krypton-85 beta radiation.
ARMOUR RESEARCH FOUNDATION Chicago, Illinois	Study of radiation reaction mechanism producing high value halogenated aromatic compounds.
ARMOUR RESEARCH FOUNDATION Chicago, Illinois	Study of ultra high intensity radiation effects.
ATLANTIC RESEARCH CORP. Alexandria, Virginia	Study of the use of organo-metallic bond in radiochemical dosimetry.
BATTELLE MEMORIAL INSTITUTE Columbus, Ohio	Study of structural and compositional factors influencing radiation induced polymerization reactions.
CONSOLIDATION COAL CO. Pittsburgh, Pa.	Evaluation of beta radiation as a hydrogenation catalyst.
FUNDAMENTAL METHODS ASSOC. New York, New York	Research on neutron transmutation doping.
GEORGIA TECHNOLOGICAL RESEARCH INSTITUTE Atlanta, Georgia	Effects of high intensity ionizing radiation on colloidal systems and suspensions.
GEORGIA TECHNOLOGICAL RESEARCH INSTITUTE Atlanta, Georgia	Physical properties and structural characteristics of polymers resulting from post effect polymerization.
RADIATION APPLICATIONS, INC. Long Island City 1, New York	Study of the mechanism of radiation induced gelation in monomer-polymer mixtures.
STANFORD RESEARCH INSTITUTE Menlo Park, California	Radiation-induced reactions in swollen polymers.
TEXTILE RESEARCH CENTER Raleigh, North Carolina	Applications of nuclear radiation and radioisotope techniques to textile materials and processes.
WESTINGHOUSE ELECTRIC CO. Pittsburgh, Pa.	Investigation of stimulated emission of gamma radiation.

WEST VIRGINIA UNIVERSITY Morgantown, West Virginia	Development of wood-plastic combinations using gamma radiation to induce polymerization.
WM. H. JOHNSTON LABS Baltimore, Maryland	Mass spectrometry of fast primary ion reactions.

Radiation Pasteurization of Foods

AGRICULTURAL MARKETING SERVICE U. S. Department of Agriculture Washington, D. C.	Studies on gamma radiation treatments for the control of post-harvest diseases of fresh fruits and vegetables.
BUR. OF COMMERCIAL FISHERIES U. S. Department of Interior Gloucester, Massachusetts	Research to conduct quality studies on several East Coast seafoods, to include determination of process parameters, flavor and odor studies, effect of pre-irradiation quality on post-irradiation storage, and development of appropriate packaging containers.
BUR. OF COMMERCIAL FISHERIES U. S. Department of Interior Seattle, Washington	Research on the radiation pasteurization of dungeness crab meat and flounder.
ECONOMIC RESEARCH SERVICE U. S. Dept. of Agriculture Washington, D. C.	Economic feasibility of radiation pasteurization of selected fruits and vegetables.
HAZELTON LABORATORIES, INC. Falls Church, Virginia	Extractive studies on packaging material to be used with irradiated foods.
LOUISIANA STATE UNIVERSITY Baton Rouge, Louisiana	Radiation pasteurization of shrimp.
MASSACHUSETTS INSTITUTE OF TECHNOLOGY Dept. of Food Technology Cambridge, Massachusetts	Study of the effects of radiation pasteurization on the storage life extension of soft-shelled clams and haddock fillets.
UNIVERSITY OF CALIFORNIA Department of Pomology Davis, California	Research on radiation pasteurization of fruits and vegetables.
UNIVERSITY OF WASHINGTON Seattle 5, Washington	Study of radiation pasteurization of marine products, installation and operation of Cobalt-60 gamma research irradiator.

Isotopes Technology Development

ARMOUR RESEARCH FOUNDATION Chicago, Illinois	Scintillation spectrometer measurements of capture gamma rays from natural elements.
BATTELLE MEMORIAL INSTITUTE Columbus, Ohio	Development and evaluation of safety performance criteria for sealed radiation sources.
BUREAU OF MINES U. S. Department of Interior Washington 25, D. C.	Research on radioactive tracers to evaluate heterogeneities in repressured petroleum reservoirs.
CATHOLIC UNIVERSITY OF AMERICA Washington 17, D. C.	Investigations regarding limiting factors in rapid identification of selected materials by nuclear techniques.
CLEGG RESEARCH LABORATORIES Santa Fe, New Mexico	Absorptiometry with beta-excited x-rays.
COLORADO SCHOOL OF MINES RESEARCH FOUNDATION, INC. Golden, Colorado	Evaluation of large volume detectors and their application to isotope process control.
DRESSER RESEARCH Tulsa, Oklahoma	Development of an ocean bottom sediment density meter and a shipboard neutron activation analyzer for ocean bottom samples.
EDSEL B. FORD INSTITUTE Detroit, Michigan	Compilation of beta-excited x-ray source spectra.
FOREST SERVICE U. S. Dept. of Agriculture Berkeley, California	Use of radioactive isotopes in measurement of snow density profiles.
GENERAL ATOMIC Div. of General Dynamics Corp. San Diego, California	Neutron activation analysis in scientific crime detection.
GENERAL ATOMIC Div. of General Dynamics Corp. San Diego, California	Development of techniques for utilizing reactor fast neutrons for activation analysis.
GEOLOGICAL SURVEY U. S. Dept. of Interior Washington 25, D. C.	Development of radioisotope techniques for water resource studies.
INTERNAL REVENUE SERVICE Alcohol & Tobacco Tax Div. U. S. Treasury Department Washington 25, D. C.	Evaluation of the effectiveness of neutron radioactivation analysis in criminalistics.

MARTIN-MARIETTA CORP. Baltimore, Maryland	Development of radioisotope energized thermally actuated underwater acoustic device.
MB ASSOCIATES Walnut Creek, California	Alpha detector.
MULLER-MONROE DESIGNS Santa Fe, New Mexico	Investigation of methods for producing and measuring beta-excited x-ray diffraction effects.
NATIONAL CANNERS ASSOCIATION Berkeley 10, California	Investigations of radioisotope tracer techniques to detect and measure detergent residues on washed food products.
NATIONAL CANNERS ASSOCIATION Berkeley 10, California	Development of radioisotope techniques to evaluate and improve routine practices for removal of pesticides from food.
PARAMETRICS, INCORPORATED Saxonville, Massachusetts	Development of krypton-85 as a universal tracer.
PARAMETRICS, INCORPORATED Saxonville, Massachusetts	Feasibility study of radiological mechanisms for use in geophysics research.
RESEARCH TRIANGLE INSTITUTE OF NORTH CAROLINA Durham, North Carolina	Low-level tracer studies, fission product uses, soil moisture studies and quality control techniques.
TEXAS NUCLEAR CORPORATION Austin, Texas	Investigation of the feasibility of using nuclear techniques for coal analysis.
TRACERLAB, DIVISION OF LABORATORY FOR ELECTRONICS Waltham 54, Massachusetts	Radioisotope application to printed circuit board technology.
TRACERLAB, DIVISION OF LABORATORY FOR ELECTRONICS Waltham 54, Massachusetts	Development of narrow band beta-excited x-ray detector for analysis of chemical elements in complex matrices.
UNDERWRITERS' LABORATORIES INCORPORATED Chicago 11, Illinois	Fire resistance tests of teletherapy unit and shipping shields.
UNION CARBIDE NUCLEAR CO. Research Center Tuxedo, New York	Investigation of radiochemical separations and metastable isomers for use in activation analysis.
UNIVERSITY OF CHICAGO Chicago 37, Illinois	Radioisotope research, development and related activities designed to lead to new or improved techniques or applications of radioisotopes to scientific, industrial and public benefit problems.

WESTINGHOUSE ELECTRIC CORP. Pittsburgh, Pa.	Study of applications of the Mossbauer effects to explore its potential to public benefit.
W. H. JOHNSTON LABORATORIES INCORPORATED Baltimore 15, Maryland	Development and testing of a deep-water isotopic current analyzer.

Production and Materials

MARTIN-MARIETTA CORP. Baltimore, Maryland	Isotopic power fuels research and development.

TABLE I

RADIOISOTOPE PRODUCTION CAPABILITY FOR THERMAL APPLICATIONS

Isotope	1963	1964	1965	1966	1967	1968	1969	1970	1971	1972
Sr-90 (Mc)	3	5	5	5	10	10	10	10	10	10
Cs-137 (Mc)	1	3.5	3.5	3.5	10	10	10	10	10	10
Ce-144 (Mc)	3.5	3.5	3.5	3.5	100	100	100	100	100	100
Pm-147 (Mc)	0.02	0.5	0.5	0.5	30	30	30	30	30	30
Pu-238 (Kg)	3	6	13	18	24	32	36	42	47	51
Cm-244 (Kg)	-	0.05	0.1	6	18	40	56	56	58	58
Cm-242 (Gm)	-	12	80	1000	1000	1000	1000	1000	1000	1000
Po-210 (Gm)	20	50	100	100	1000	1000	1000	1000	1000	1000

TABLE II

THERMAL POWER AVAILABILITY FROM RADIOISOTOPES*

(Kilowatts/year at Fuel Fabrication Time)

Isotope	1963	1964	1965	1966	1967	1968	1969	1970	1971	1972
Sr-90	19	32	32	32	63	63	63	63	63	63
Cs-137	5	17	17	17	48	48	48	48	48	48
Ce-144	25	25	25	25	700	700	700	700	700	700
Pm-147	0.01	0.2	0.2	0.2	14	14	14	14	14	14
Pu-238	1.5	3.0	6.0	0.5	11.5	15	17	20	23	25
Cm-244	-	0.1	0.2	14	41	92	129	129	134	134
Cm-242	-	1.5	9.5	120	120	120	120	120	120	120
Po-210	3.0	7.0	14	14	140	140	140	140	140	140

*Based on quantities of material in Table I on Radioisotope Production Capability for Isotopic Power Applications.

TABLE III

PROPERTIES OF RADIOISOTOPES FOR ISOTOPIC POWER APPLICATIONS

Isotope	Type of Decay	Half-life	Chemical Form	Specific Thermal Power			Material Requirement for Mission	
				Watts/kc	Watts/gm	Watts/cc	Mission Life (yrs)	Curies/Electrical Watt
Sr-90	Beta	28 yrs	Titanate	6.5	0.2	0.7	10	4,000
Cs-137	Beta Gamma	27 yrs	Glass	4.8	0.072	0.22	10	5,500
Ce-144	Beta Gamma	285 days	Oxide	7.9	2.3	13.8	1	7,000
Pm-147	Beta	2.6 yrs	Oxide	0.37	0.18	1.0	2.6	108,000
Pu-238	Alpha	89.6 yrs	Metal	34.5	0.48	9.3	10	625
Cm-244	Alpha	18.4 yrs	Oxide	35.0	2.3	22.4	10	840
Cm-242	Alpha	163 days	Oxide	36.2	120	1170	0.5	1,190
Po-210	Alpha	138 days	Metal	31.7	140	1320	0.5	1,550

TABLE IV

EXISTING COMMERCIAL RADIATION PROCESSES

Process	Source	Company	Volume
Production of Ethyl Bromide	Cobalt-60	Dow Chemical Co.	400 tons per year
Polyethylene Film	Machine	W. R. Grace	1500 tons per year
Crosslinked Polyethylene Wire Insulation, Circuit Boards, etc.	Machine	Radiation Application, Inc. General Electric Raychem (Redwood City, Calif.)	$15 million per year
Medical Supply Sterilization	Machine Cobalt-60	Ethicon, Inc. Hospital Supply Co. Becton, Dickinson Co.	80% of USA Market (Sutures)
Goat Hair Sterilization	Cobalt-60	Westminster Carpet Co. (Australia)	2,250 tons per year
Sprout Inhibition on Potatoes	Cobalt-60	Atomic Energy of Canada, Ltd.	500 tons per year
Production of Polyvinyl Acetal Resin	Machine	Toyo Rayon (Japan)	no estimate
Semiconductors	Machine	At least six of the larger semiconductor producers	15-20 million units a year

TABLE V

NEAR-FUTURE COMMERCIAL RADIATION PROCESSES

Process	Probable Source	Potential Use
Radiation curing of coatings	Machine	Plastic coated plywood panels, paint primer curing
Textiles	Machine	Dyeability, mildew resistance, fire retardant, water resistance
Wood-plastic alloys	Machine Cobalt-60	Wood construction, furniture, piling, sporting equipment
Sterilization of Medical Supplies	Cobalt-60	Bulk packaged sutures, catheter kits, etc.
Food pasteurization	Cobalt-60	Extension of shelf life (fish and fruit)
Semiconductor devices	n- γ flux*	Transistors diodes multifunctional devices
Food sterilization	Cobalt-60	Military rations (chicken, beef, pork, and ham)
Enzyme sterilization	Machine	----------

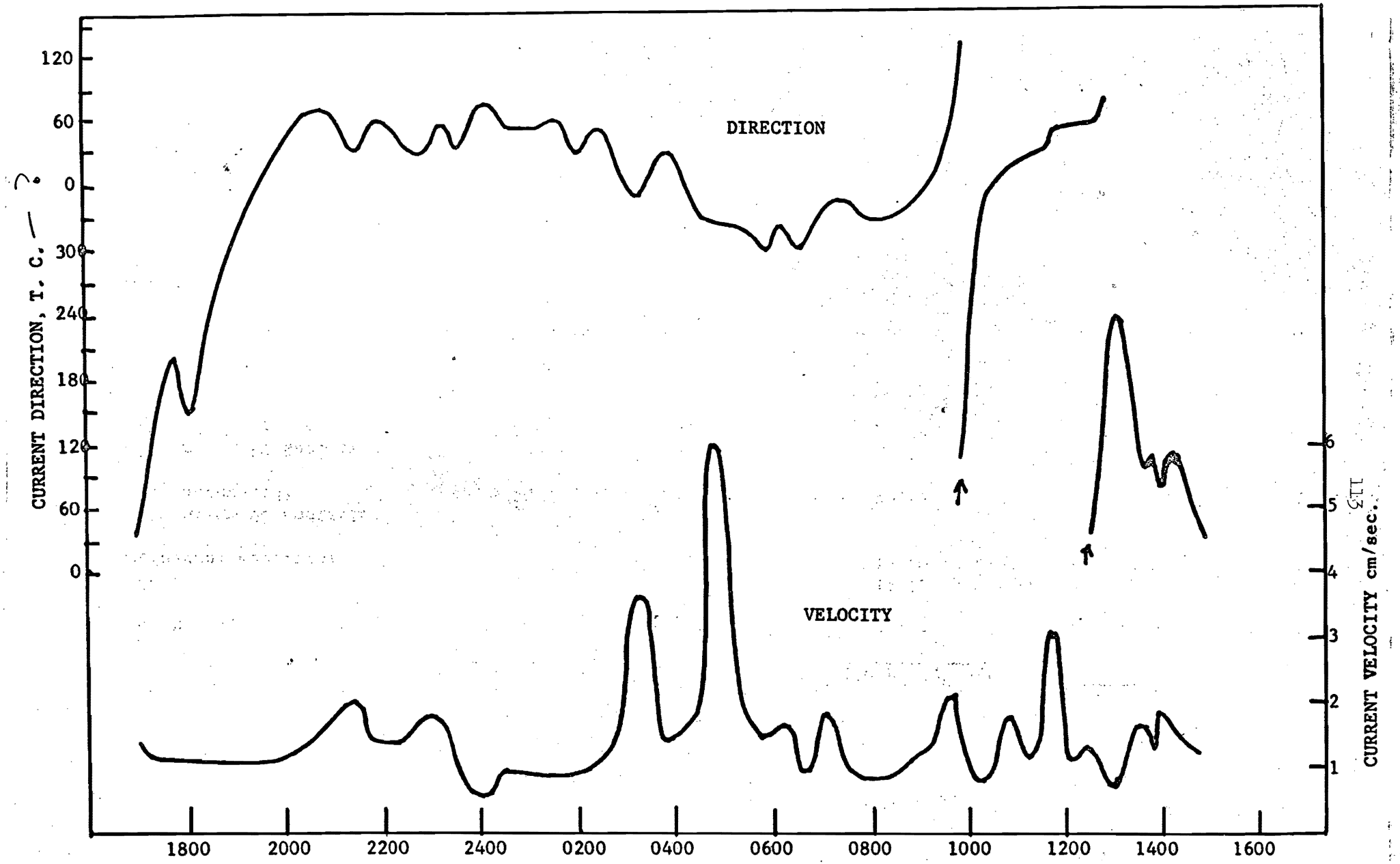
* Nuclear Reactor

TABLE VI

TYPICAL RESULTS, COAL EXTRACT TO OIL CONVERSION

T, 429°C ; P, 500 psi; Feed, Extract:Tetralin in a 1:1 Ratio with 7.3% Hydrogen

Catalyst	Yield From Coal Extract, %			% H in Overall Product
	100 - 250°C Oil	250 - 400°C Oil	100 - 400°C Oil	
Co,Mo,Ni,S	33.47	17.69	51.16	10.37
None,	5.12	9.65	14.77	8.24
(Thermal Reaction)	8.55	9.65	18.20	8.24
Strontium 90 Radiation (H_2 Saturated)	8.73	6.79	15.52	7.76
Strontium 90 Radiation	4.82	15.80	20.72	8.00
(Not H_2 Saturated)	5.32	16.30	21.62	8.00



SUMMARY OF DWICA FIELD DATA -C.B.I VESSEL "MAURY" ROUND BAY, MD., DEC. 14-15, 1961 (TIME E.S.T)

FIGURE 1

TABLE VIII

Degree of Labeling in Six Hours, Compared to Ultimate Theoretical Limit
(T = 23°C)

Compound	Theoretical Limit of Tagging (μc/gm)	Tagging in Six Hours (μc/gm)	Percent of Ultimate Tagging Achieved In Six Hours (%)	Experimentally Observed Limit of Tagging (μc/gm)
Benzene	630	108.2	17.2	~630
Decalin	435	31.4	7.2	-
Tetralin	545	542	99.4	~550
Toluene	603	554	92.0	~560
Methylcyclohexane	381	9.36	4.1	-
Naphthalene	2830	1290	45.6	-
Cyclohexane	No exchange tagging.			

II. ISOTOPE PRODUCTION AND DEVELOPMENT

Radioisotopes become available in a number of ways. Megacuries of activity result from the fissioning of reactor fuels; kilocuries of such isotopes as Co^{60} and Ir^{192} and curies or millicuries of many others can be made by activating targets in reactors; and small amounts of a wide variety of nuclides result from the bombardment of suitable targets in accelerators such as cyclotrons. Stable isotopes are concentrated by a number of methods, the most important of which are electromagnetic separation, thermal diffusion, and gaseous diffusion.

In this first issue of "Isotopes and Radiation Technology," fission product separation and stable isotope research and development are discussed briefly. Cyclotron-produced isotopes will be discussed in future issues, along with additional details of these processes and material on reactor-produced isotopes.

FISSION PRODUCTS

When gross amounts of U^{235} undergo fission, the fission products comprise, either as intermediates or as isolatable nuclides, a rather sizeable percentage of the known isotopic species. The familiar curve in Fig. II-1 shows the distribution of the isotopic masses and Table II-1 lists some of the more important recoverable radioactive products. Table II-2 is of potential interest, since it shows abundances of inactive (stable) fission-product isotopes; some of these, as a result of their abnormal isotope ratios, are worthy of consideration for calutron charge to prepare enriched stable isotopes.

Inherent in the operation of all nuclear reactors are the problems of recovery of the fertile fuel and handling of the fission-product wastes. In many instances the wastes are being stored or buried.

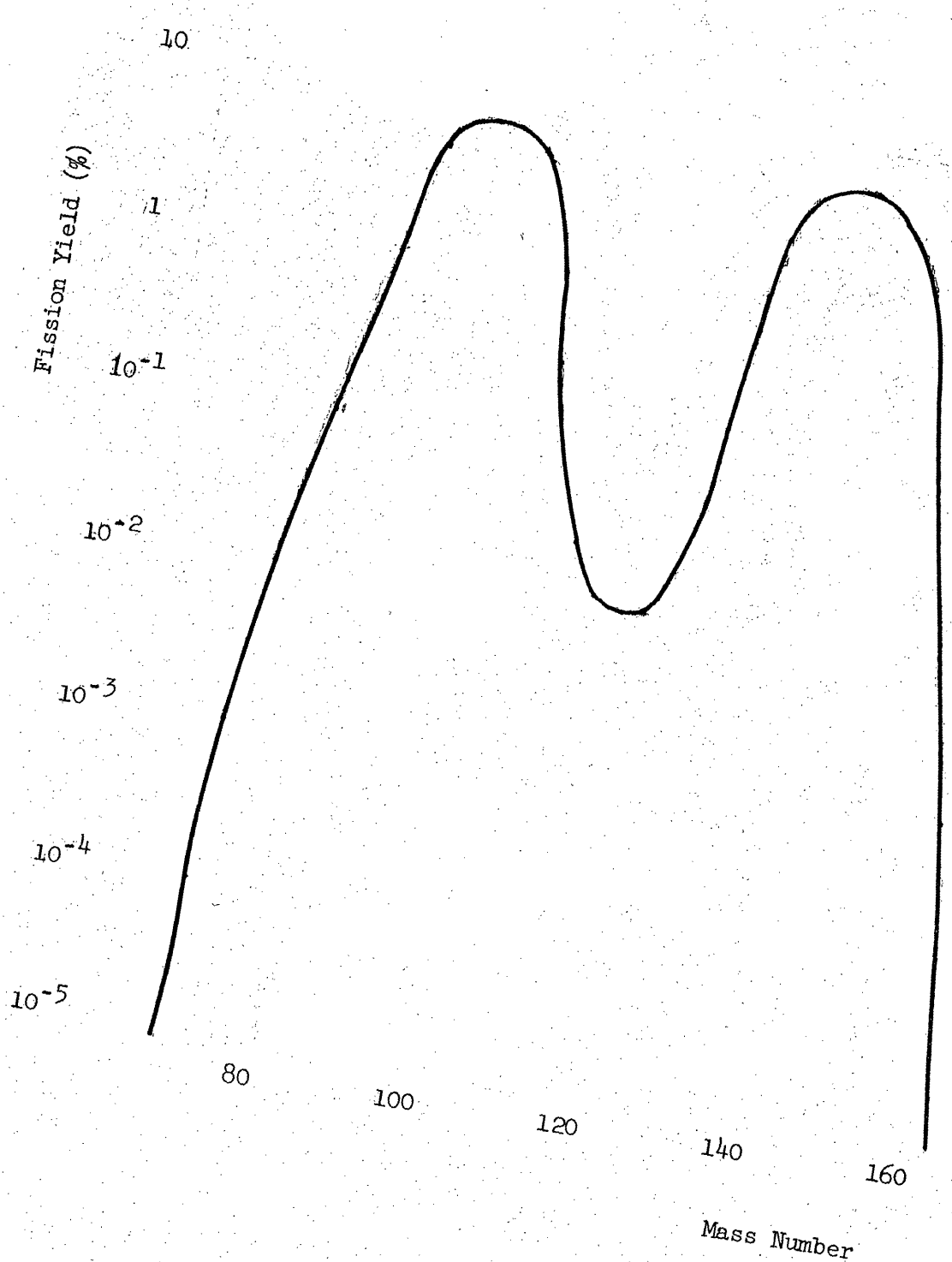


Fig.II-1.Fission Product Distribution.

TABLE II-1. ACTIVE FISSION PRODUCT ISOTOPES OF HALF-LIFE GREATER THAN 57 DAYS

Isotope Z	A	Fission Yield, %	Half-Life	Amount in Nature, %	Thermal Neutron Cross Section barns
34	Se^{79}	0.04	6.5×10^4 y	0	-
36	Kr^{85}	0.3	10.27 y	0	< 15
37	Rb^{87}	2.7	6.2×10^{10} y	27.85	0.14
38	Sr^{90}	5.9	28 y	0	~1.0
39	Y^{91}	5.9	58 d	0	-
40	Zr^{95}	6.4	63 d	0	-
	Zr^{93}	6.5	1.1×10^5 y	0	< 4
41	Nb^{95m}	2.1	4.2 y	100	-
43	Tc^{99}	6.1	2.12×10^5 y	0	-
44	Ru^{106}	0.38	1.0 y	0	-
46	Pd^{107}	0.2	7.5×10^6	0	-
47	Ag^{110m}	2×10^{-7}	270 d	0	-
48	Cd^{113m}	?	5.1 y	12.26	-
49	In^{115}	0.0099	6×10^{14} y	95.77	145 + 52
50	Sn^{110m}	0.01	275 d	8.58	-
	Sn^{123}	0.0012	131 d	0	-
51	Sb^{125}	0.023	2.7 y	0	-
52	Te^{130}	2.0	$>2.0 \times 10^{15}$ y	34.49	< 0.01 + 0.3
	Te^{127m}	0.056	90 d	0	-
53	I^{129}	1.0	1.67×10^7 y	0	11
55	Cs^{135}	6.2	3.0×10^6 y	0	15
	Cs^{137}	5.9	33 y	0	< 2

Table II-1 (continued)

Isotope Z	A	Fission Yield, %	Half-Life	Amount in Nature, %	Thermal Neutron Cross Section barns
58	Ce^{144}	6.1	290 d	0	-
60	Nd^{144}	6.1	1.5×10^{15}	6.1	4.8
	Nd^{150}	0.74	2.0×10^{15}	5.6	2.9
61	Pm^{147}	2.6	2.6 y	0	60 + ?
62	Sm^{147}	2.6	1.4×10^{11} y	15.07	-
	Sm^{151}	0.5	73 y	0	7000
63	Eu^{155}	0.031	1.7	0	14,000

TABLE II-2. A COMPARISON OF AMOUNTS OF INACTIVE FISSION PRODUCT ISOTOPES AND NATURALLY OCCURRING ISOTOPES

Isotope Z	Isotope A	Fission Yield	Amount, % In Fission	Amount, % In Nature	Cs^{137} per 50 Kcuries, g	Thermal Neutron Cross Section, barns
32	Ge^{70}	0	0	20.55		
	Ge^{72}	1.5×10^{-5}	0.6	27.37	-	0.94
	Ge^{73}	1.0×10^{-4}	4.1	7.67	-	13.7
	Ge^{74}	3.0×10^{-4}	12.4	36.74	-	0.6
	Ge^{76}	2.0×10^{-3}	82.9	7.67	-	0.15 + 0.30
33	As^{75}	8.0×10^{-4}	100	100	-	
34	Se^{74}	0	0	0.87	-	-
	Se^{76}	0	0	9.02	-	-
	Se^{77}	9.1×10^{-3}	2.5	7.58	-	40
	Se^{78}	2.0×10^{-2}	5.6	23.52	3.0	0.4
	Se^{80}	8.0×10^{-2}	22.3	49.82	12.1	0.03 + 0.5
	Se^{82}	0.25	69.6	9.19	37.9	2.0
35	Br^{79}	3.7×10^{-2}	21.7	50.52	5.6	2.6 + 7.6
	Br^{81}	0.133	78.3	49.48	20.0	2.6
36	Kr^{78}	0	-	0.354	-	
	Kr^{80}	0	-	2.27	-	
	Kr^{82}	3.5×10^{-5}	0.09	11.56	-	45
	Kr^{83}	0.48	13.0	0	72.8	205
	Kr^{84}	1.1	29.9	56.90	166.9	0.1 + 0.06
	Kr^{86}	2.1	57.1	17.37	318.8	0.06
37	Rb^{85}	1.1	-	72.15	227.7	0.05 + 85
	Rb^{87}	-	-	27.85	-	0.14

Table II-2 (continued)

Isotope Z	Isotope A	Fission Yield	Amount, % In Fission	Amount, % In Nature	Cs^{137} per 50 Kcuries, g	Thermal Neutron Cross Section, barns
38	Sr^{84}	0	-	0.56	-	
	Sr^{86}	2.8×10^{-5}	0.04	9.86	-	1.3
	Sr^{87}	2.7	42.2	7.02	409.8	
	Sr^{88}	3.7	57.8	82.56	561.6	0.005
39	Y^{89}	4.8	100	100	728.6	1.4
40	Zr^{90}	5.9	19.1	51.46	895.6	0.1
	Zr^{91}	5.9	19.1	11.23	895.6	2.0
	Zr^{92}	6.1	19.8	17.11	925.9	0.2
	Zr^{94}	6.5	21.1	17.40	986.7	0.07
	Zr^{96}	6.4	20.8	2.80	971.5	0.05
41	Nb^{93}	6.5	100	100	986.7	1.1
42	Mo^{92}	0	-	15.86	-	-
	Mo^{94}	0	-	9.12	-	-
	Mo^{95}	6.4	27.1	15.70	971.5	13.4
	Mo^{96}	5.7×10^{-4}	0.02	16.50	-	1.0
	Mo^{97}	6.2	26.27	9.45	941.5	2.0
	Mo^{98}	5.9	25.0	23.75	895.6	0.13
	Mo^{100}	6.5	27.5	9.62	987.7	0.2
44	Ru^{96}	0	0	5.7	-	
	Ru^{98}	0	0	2.2	-	
	Ru^{99}	6.1	35.6	12.8	925.9	
	Ru^{100}	0	0	17.0	-	
	Ru^{101}	5.0	29.2	12.7	759.0	2.46

Table II-2 (continued)

Isotope Z	A	Fission Yield	Amount, % In Fission	Amount, % In Nature	Cs^{137} per 50 Kcuries, g	Thermal Neutron Cross Section, barns
	Ru^{102}	4.2	24.6	31.3	637.6	1.2
	Ru^{104}	1.8	10.5	18.3	273.2	0.7
45	Rh^{103}	2.9	100	100	440.2	138.0
46	Pd^{102}	0	0.8	-		
	Pd^{104}	0	9.3	-		
	Pd^{105}	0.9	65.3	22.6	136.6	
	Pd^{106}	0.38	27.5	27.2	57.7	
	Pd^{108}	0.08	0.58	26.8	12.1	0.07 + 11
	Pd^{110}	0.02	0.15	13.5	3.0	? + 0.04
47	Ag^{107}	0.2	87.7	51.35	30.0	30
	Ag^{109}	0.028	12.3	48.65	4.2	2 + 82
48	Cd^{106}	0	1.215	-		
	Cd^{108}	0	0.875	-		
	Cd^{110}	2×10^{-7}	0.033	12.39	-	
	Cd^{111}	0.018	30.5	12.75	2.7	
	Cd^{112}	0.011	18.6	24.07	1.6	0.03 + ?
	Cd^{113}	0.01	16.9	12.26	1.5	25,000
	Cd^{114}	0.01	16.9	28.86	1.5	0.14 + 1.1
	Cd^{116}	0.01	16.9	7.58	1.5	1.4
50	Sn^{112}	?	0	0.95	-	
	Sn^{114}	0		0.65	-	
	Sn^{115}	0.01	12.04	0.34	1.5	
	Sn^{116}	0	0	14.24	-	

Table II-2 (continued)

Isotope Z	Isotope A	Fission Yield	Amount, % In Fission	Amount, % In Nature	Cs^{137} per 50 Kcuries, g	Thermal Neutron Cross Section, barns
	Sn^{117}	0.010	12.04	7.57	1.5	
	Sn-118	0.01	12.04	24.04		
	Sn^{119}	0.01	12.04	8.58	1.5	
	Sn^{120}	0.01	12.04	32.97	1.5	0.001 + 0.03
	Sn^{122}	0.013	15.6	4.71	1.9	0.001 + 0.1
	Sn^{124}	0.02	24.0	5.98	3.0	0.75
51	Sb^{121}	0.014	48.2	57.25	2.1	5.7
	Sb^{123}	0.015	51.7	42.75	2.3	2.5+0.03+0.03
52	Te^{120}	0	0	0.089	-	
	Te^{122}	0	0	2.46	-	
	Te^{123}	0	0	0.87	-	
	Te^{124}	0	0	4.61	-	
	Te^{125}	0.023	3.69	6.99	3.5	1.5
	Te^{126}	0.10	16.05	18.71	15.2	0.07 + 0.7
	Te^{128}	0.5	80.2	31.79	75.9	0.016 + 0.14
	Te^{130}	0	0	34.49	-	
53	I^{127}	0.25	100	100	37.9	6.1
54	Xe^{124}	0	0	0.096	-	
	Xe^{126}	0	0	0.090	-	
	Xe^{128}	0	0	1.92	-	
	Xe^{129}	1.0	4.5	26.44	151.8	45
	Xe^{130}	0	0	4.08	-	
	Xe^{131}	2.9	13.0	21.18	440.2	120
	Xe^{132}	4.4	19.8	26.89	667.9	? + 2

Table II-2 (continued)

Isotope Z	Isotope A	Fission Yield	Amount, % In Fission	Amount, % In Nature	Cs^{137} per 50 Kcuries, g	Thermal Neutron Cross Section, barns
	Xe^{134}	7.6	34.2	10.44	1153.7	? + 2
	Xe^{136}	6.3	28.3	8.87	956.3	0.15
55	Cs^{133}	6.5	100	100	986.7	0.016 + 26
56	Ba^{130}	0	0	0.101	-	
	Ba^{132}	0	0	0.097	-	
	Ba^{134}	0	0	2.42	-	
	Ba^{135}	6.2	34.6	6.59	941.1	5.6
	Ba^{136}	6×10^{-3}	0.33	7.81	45.5	0.4
	Ba^{137}	5.9	32.9	11.82	895.6	
	Ba^{138}	5.8	32.4	71.66	880.4	0.68
57	La^{138}	0	0	0.089	-	
	La^{139}	6.0	100	99.9	910.8	8.8
58	Ce^{136}	0	0	0.193	-	
	Ce^{138}	0	0	0.250	-	
	Ce^{140}	6.3	51.6	88.5	956.3	0.6
	Ce^{142}	5.9	48.6	11.07	895.6	1.8
59	Pr^{141}	6.0	100	100	910.8	11
60	Nd^{142}	0	0	27.13	-	
	Nd^{143}	6.2	39.9	12.20	1004.9	290
	Nd^{144}	0	0	23.87	-	4.8
	Nd^{145}	4.2	37.1	8.30	637.6	52
	Nd^{146}	3.3	21.2	17.18	500.9	9.8
	Nd^{148}	1.8	11.6	5.72	273.2	3.3

Table II-2 (continued)

Isotope Z	Isotope A	Fission Yield	Amount, % In Fission	Amount, % In Nature	Cs^{137} per 50 Kcuries, g	Thermal Neutron Cross Section, barns
	Nd^{150}	0	0	5.60	-	
62	Sm^{144}	0	0	3.16		
	Sm^{147}	0	0	15.07		
	Sm^{148}	0	0	11.27		
	Sm^{149}	1.3	76.9	13.84	197.3	50,000
	Sm^{150}	0	0	7.47	-	
	Sm^{152}	0.3	17.8	26.63	45.5	150
	Sm^{154}	0.09	5.3	22.53	13.6	5.5
63	Eu^{155}	0.5	76.9	47.77	75.9	1400-7000
	Eu^{153}	0.15	23.1	52.23	22.7	480
64	Gd^{152}	0	0	0.20	-	
	Gd^{154}	0	0	2.15	-	
	Gd^{155}	0.031	70.9	14.73	4.7	70,000
	Gd^{156}	0.013	29.7	20.47	1.9	
	Gd^{157}	0.0074	0.17	15.68	1.1	160,000
	Gd^{158}	0.002	0.04	24.87	0.3	4
	Gd^{160}	3×10^{-4}	0.006	21.90	-	1.5
66	Dy^{156}	0	0	0.0524	-	
	Dy^{158}	0	0	0.0902	-	
	Dy^{160}	0	0	2.294	-	
	Dy^{161}	8×10^{-5}	0	18.88	-	
	Dy^{162}	0	0	25.53	-	
	Dy^{163}	0	0	24.97	-	
	Dy^{164}	0	0	28.18	-	

However, the economics of waste management are clearly tied in with fission-product separation and recovery.[1] An important experiment involves the Fission Products Development Laboratory (FPDL) at ORNL, which was designed specifically to study the feasibility of recovery of multikilocuries of potentially useful fission products and to make them available for a variety of uses. At first, wastes from ORNL and ICCP were used. More recently, Hanford has been supplying the bulk of the processed wastes for ORNL to handle. At present the operations involve primarily separation and purification of Cs^{137}, Ce^{144}, and Pm^{147}.[2] Strontium-90 is received as a purified feed for conversion to appropriate source compounds.

Fission-Product Processing at Hanford

The strontium is recovered from the acid waste raffinate from the Purex plant by precipitation followed by solvent extraction. Strontium and the rare earths are first separated by a lead-carrier sulfate step at pH 1.0, the strontium being carried with the lead sulfate as strontium sulfate. Tartaric acid is added to complex the iron. After centrifugation, the precipitated strontium (including lead) and rare earth sulfates are converted to carbonates by washing with sodium carbonate, and the carbonates are dissolved in dilute nitric acid. Figure II-2 indicates the essential steps of this phase of the process.

The nitric acid solution is now treated with oxalic acid to precipitate lead and the rare earths, leaving the strontium in solution. The strontium is again precipitated as carbonate to decrease the volume of feed to the solvent extraction process.

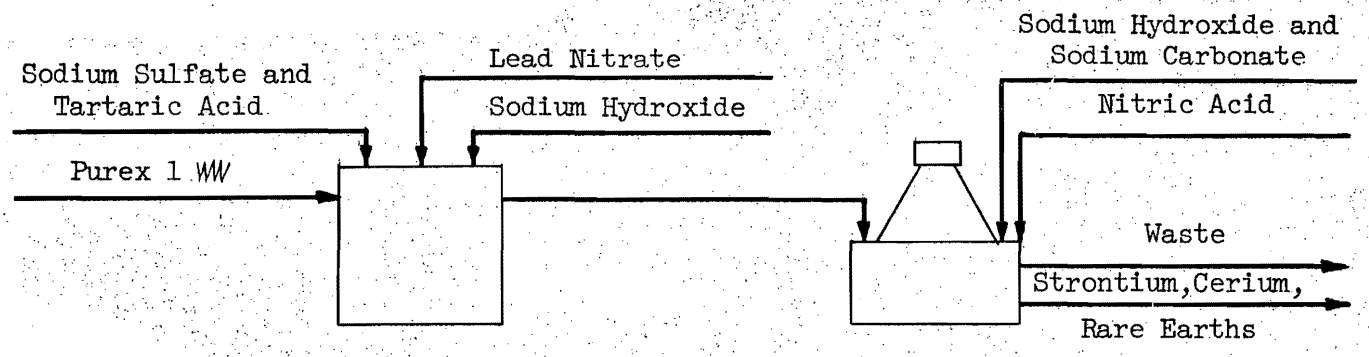


Fig. II-2. Lead Carrier Sulfate Precipitation.

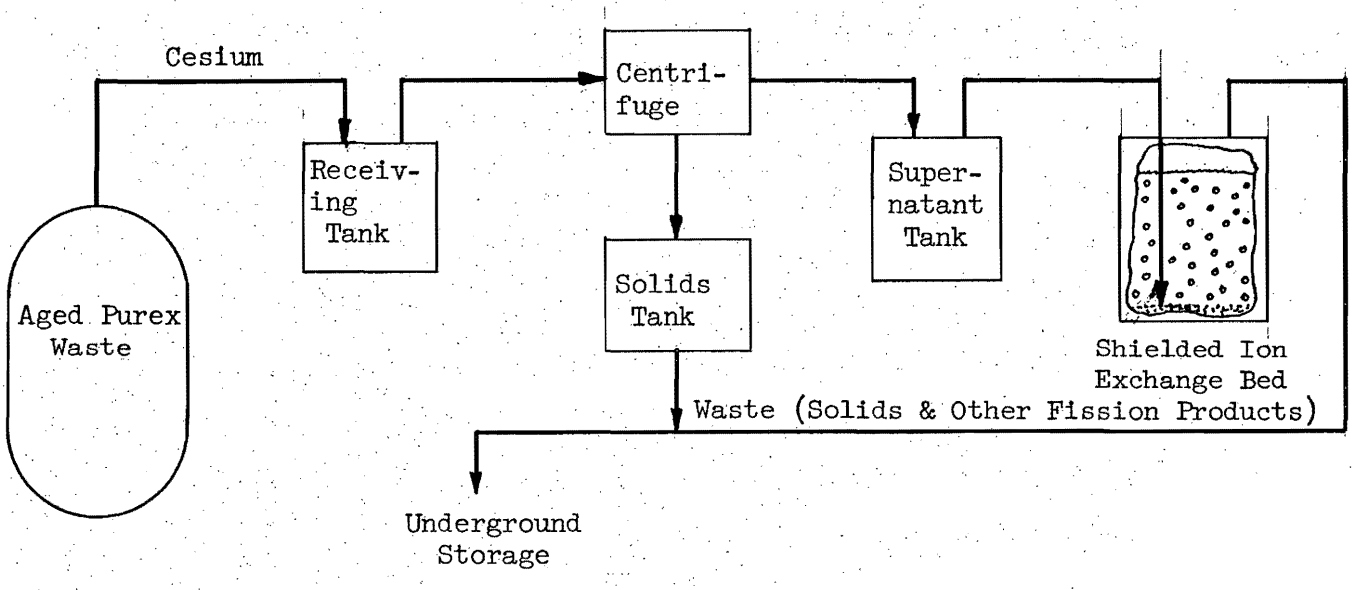


Fig. II-3. Cesium-137 Recovery.

Strontium purification is by solvent-extraction with di(2-ethylhexyl)phosphoric acid as the organic extractant. The final product is reported to be "virtually free of all radiochemical impurities, and cation chemical purities greater than 95% are typical."[2a]

The Cs^{137} recovery is outlined in Fig. II-3. The process consists primarily in passing the centrifuged supernatant liquid in aged Purex stored wastes through a shielded tank filled with an aluminosilicate ion-exchange resin (Decalso). The cesium is sorbed on the resin in a form suitable for shipment.

Transportation

Figure II-4 shows the "portable pipeline" used for transferring feed from Hanford to ORNL. Each gondola carries two 500-gal shielded transfer tanks (STT's) containing 50 kcuries of Cs^{137} per cask sorbed on Decalso resin. The flat bed on the extreme left contains a HAPO filter slurry cask with 140 kcuries of $Sr^{90}CO_3$ powder. A HAPO carrier can ship up to 250 kcuries of $Sr^{90}CO_3$ as a dried powder on a metal filter.

A cutaway of an STT carrier and its strengthened containment features is shown in Fig. II-5. The cask weighs 18 tons and is loaded with 400 gal of Decalso resin. The limiting factor on Cs^{137} shipments is the 4 in. of lead shielding, which holds the loading to 50 kcuries of Cs^{137} per cask. Upon arrival at ORNL, the fission products are handled in the FPDL.

Fission Products Development Laboratory

The fission product research and development work leading to construction of the FPDL began in 1948 with investigation of a number of

Fig. II-4. Photo 58885

Fig. II-5. ORNL LR DWG 63529

methods for removing long-lived fission products from waste solutions. Fission products were first separated in curie quantities in laboratory glassware by ion-exchange processes. Later, hundred-curie quantities were separated in small stainless steel vessels. Finally, in 1958, the FPDL facility went on stream to separate kilocurie amounts of Cs^{137}, Sr^{90}, Ce^{144}, and Pm^{147} from Redox- and Purex-type waste streams. This program was successfully demonstrated during the first 2 years of operations. In order to meet the requirements for preparation of large-scale heat sources during the past 2 years, concentrated feed materials have been shipped from Hanford on a megacurie-per-year basis.

Figure II-6 shows a cutaway of the FPDL with its manipulator cells, wet chemical process cells, analytical cell, tank farm, and associated equipment. The plant is designed for complete processing -- from a liquid waste stream to an encapsulated solid product that can be shipped directly to the user. Seven high-level manipulator cells used for product purification and encapsulation contain equipment for precipitation, filtration, calcination, blending, weighing, pelletization, sintering, and welding. Two low-activity-level manipulator cells are available for sampling fission product solutions and process control techniques. Eight wet chemical process cells are equipped with stainless steel piping and vessels to permit a variety of unit operations such as precipitation, centrifugation, filtration, crystallization, decantation, evaporation, and solvent extraction.

Recently, a megacurie storage system was installed in the FPDL facility by separating the plant into a three-product system and adding an additional powder storage well. All HEW feed material is now

Fig. II-6. ORNL LR DWG 10845

unloaded here.

Production. During the past two years FPDL operations have been concentrated on a two-product stream of Cs^{137} and Sr^{90}. The feed material was obtained from Hanford and then purified to product form. Cesium-137 chloride production exceeded 250 kcuries, and a 27-kcurie cesium glass source was produced from this material. Table II-3 indicates the quantities of $SrTiO_3$ produced for the SNAP-7 program. The Weather Bureau 17-kcurie source was produced as a prototype in FY-1961, and the program proceeded rapidly to the production of the 224-kcurie SNAP-7D source for the Navy.[3] Scaleup from 17 to 224 kcuries required additional man-days of labor, but no changes in the operating procedure. A stepup to 1-Mcurie sources can be accomplished at the FPDL with no changes to the facility or to its handling procedures. Flowsheets for Sr^{90}, Cs^{137}, and Ce^{144} are presented in Figs. II-7, II-8, and II-9.

Strontium-90. The product of the Sr^{90} process is $SrTiO_3$ of specific activity from 30 to 35 curies/g. It is stored in 10-kcurie lots in stainless steel storage cans inside a cooled storage well. When needed, the dry $Sr^{90}TiO_3$ is formed into pellets and sintered at 1400°C to yield pellets 0.25-3.5 in. dia. The final pellets, with densities of 3.5-4.0 g/cm^3 and activity concentrations well in excess of 100 $curies/cm^3$, can be encapsulated in welded stainless steel containers.

The current feed material for the process is highly purified $Sr^{90}CO_3$ obtained from the HAPO casks by water slurrying followed by dilute nitric acid rinsing. This material is radiochemically pure and contains a few percent Ca and Ba. The slurry is treated with HNO_3 to yield a $Sr^{90}(NO_3)_2$ feed solution. At the FPDL there is storage capacity

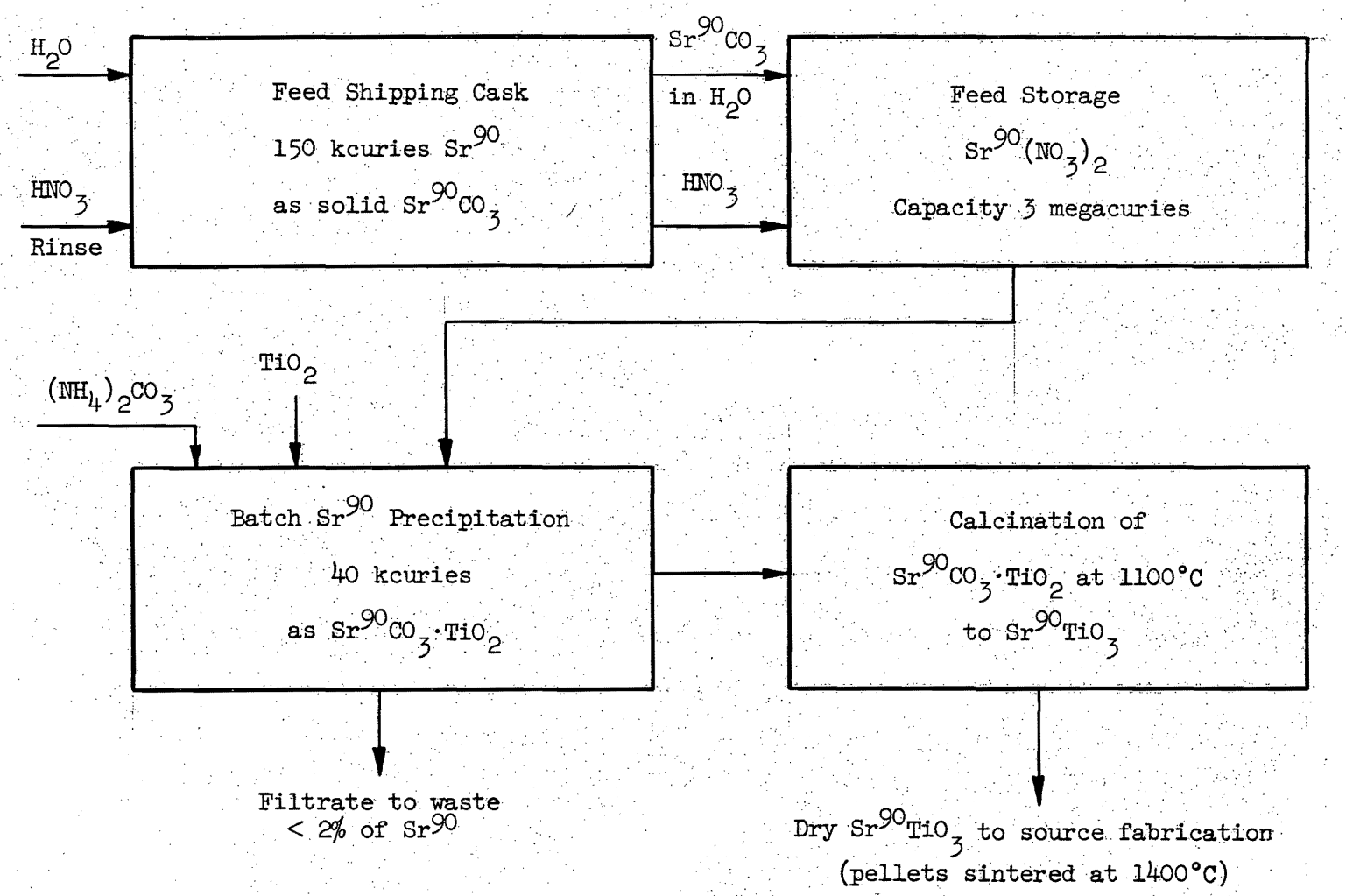


Fig. II-7. FPDL Sr^{90} Processing.

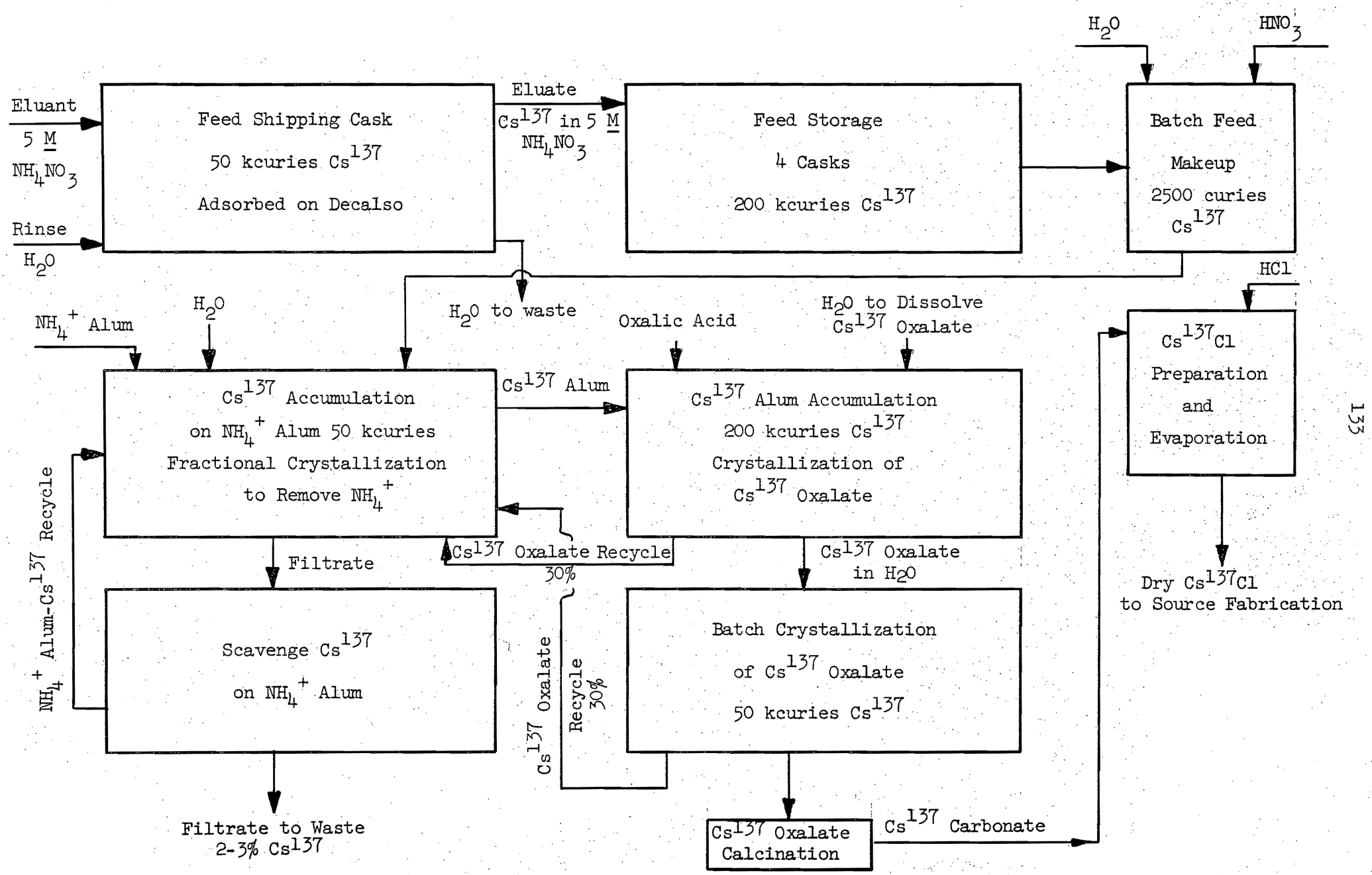


Fig. II-8. FPDL Cs^{137} Processing.

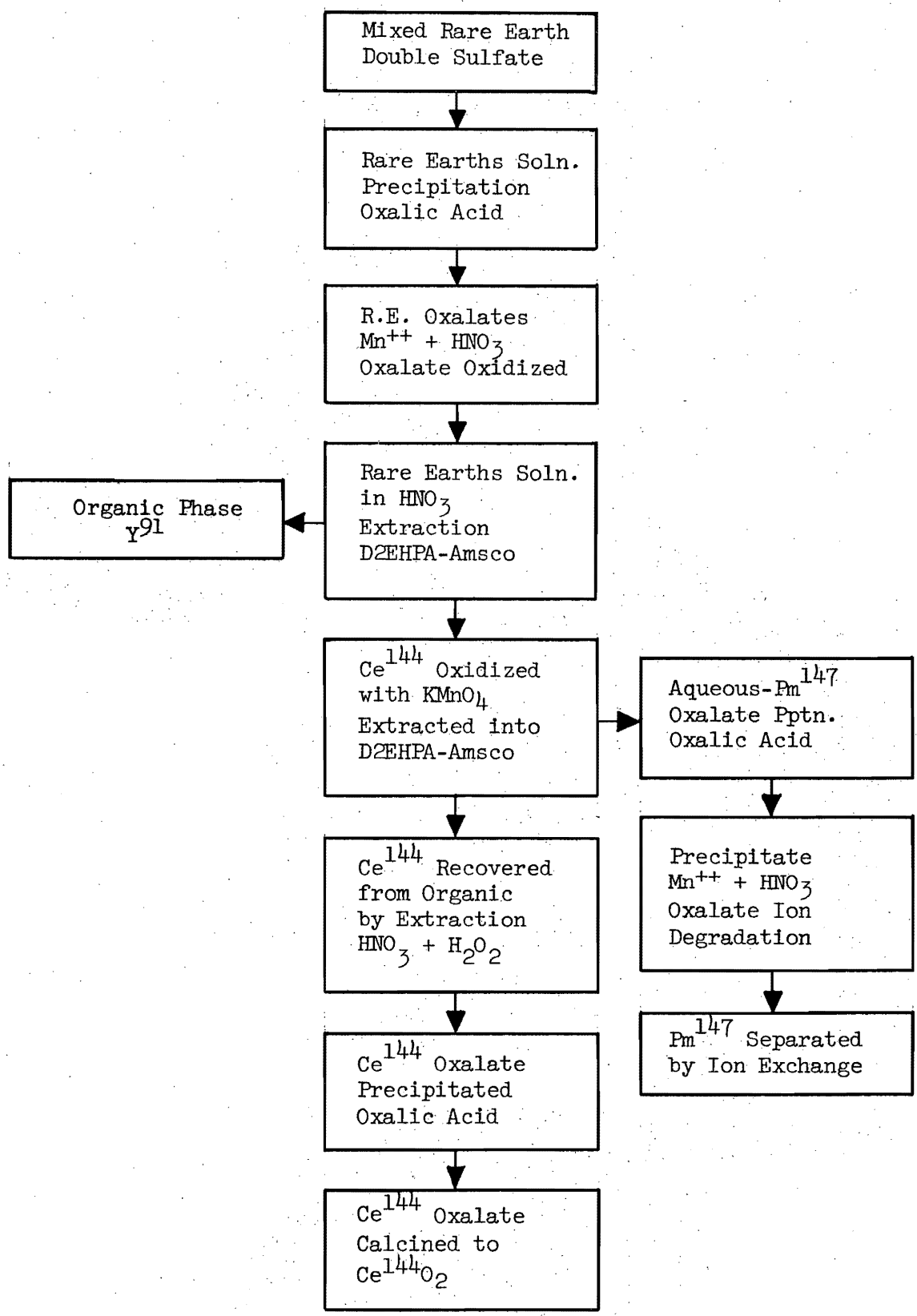


Fig. II-9. Ce^{144} Purification Chemistry.

for 3 Mcuries of such solution.

From 40-kcurie batches, strontium carbonate is precipitated in the manipulator cells by ammonium carbonate in the presence of 5-10% excess solid TiO_2. The excess TiO_2 provides a powder that can be easily pelletized. The precipitate is filtered off and the $SrCO_3$-TiO_2 mixture is calcined at 1100°C to yield $SrTiO_3$. The specific activity of the product depends on the amount of excess TiO_2 and the concentration of other alkaline earths in the feed.

Strontium-90 can be converted to $Sr^{90}TiO_3$ at a rate of 100 kcuries per week, the annual production being dependent on the availability of feed material. The preparation rate for pellets varies with the pellet size, encapsulation method, and number of pellets of a given type.

Cesium-137. Assay of the usual $Cs^{137}Cl$ product from the FPDL shows approximately 26 curies/g, compared to a theoretical specific activity of 26.4 curies/g. The mass distribution of cesium isotopes in this material is 45.5 atom % Cs^{133}, 16.8 atom % Cs^{135}, and 37.7 atom % Cs^{137}. Traces of aluminum, iron, and nickel are present, as well as small amounts of rubidium. The dry powder can be stored for extended periods in stainless steel containers with no attack on the storage cans*; it can be weighed and handled by standard manipulator procedures. No significant tendency toward water adsorption has been found. The Cs^{137} may also be incorporated in a glass with specific activity of about 14 curies/g. The glass can be cast into pellets or other shapes and then encapsulated in welded stainless steel.

Feed for the Cs^{137} process is obtained by eluting cesium from the STT Decalso beds with 5 M NH_4NO_3 at 80°C. This solution is stored in

*See report on cesium sources, p.

underground tanks which have storage capacity for as much as 500 kcuries. The STT's are rinsed and returned to HAPO with the exchanger in the NH_4^+ form.

The feed is diluted to reduce the NH_4^+ concentration, and the Cs^{137} is accumulated by a series of co-crystallizations on large ammonium alum beds. When about 50 kcuries of Cs^{137} has been collected, about 1 week's operation, the ammonium alum is removed by fractional crystallization and the cesium alum is transferred to an accumulation vessel. In addition to concentrating the Cs^{137}, the crystallization removes traces of the other radioisotopes present.

Up to 200 kcuries of Cs^{137} as alum solution is collected in the accumulation tanks, and cesium tetraoxalate is crystallized from it in ~70% yield by addition of oxalic acid. Since the filtrate is recycled to the mainstream operation, losses are negligible. The tetraoxalate is dissolved in water and the solution transferred in batches to the final purification cell where oxalate is crystallized again, and the filtrate is again recycled to the mainstream. The crystals are calcined in air at 300°C to yield dry powdered cesium carbonate. This particular form is ideally suited for conversion to any other product form, since it is readily soluble in acids.

Dry, powdered $Cs^{137}Cl$ is now prepared in 50-kcurie batches by reacting the $Cs_2^{137}CO_3$ in a water slurry with a slight excess of hydrochloric acid. This operation is done in glassware in a manipulator cell. The $Cs^{137}Cl$ solution is filtered, and the filtrate is evaporated in tantalum equipment to yield the dry salt. The powder is baked with constant stirring at 300°C for 4 hr to remove the last traces of HCl;

it is then stored in lots of 10 kcuries each in stainless steel cans, which are stored in water-cooled storage wells. In an earlier method, cesium chloroplatinate was precipitated from the cesium alum solution, filtered, washed, and treated with hydrazine to reduce the platinum to the metal. The mixture was filtered, the $Cs^{137}Cl$ was fumed with aqua regia and repeatedly digested with HCl, and the solution was finally evaporated to dryness to yield $Cs^{137}Cl$. Because of the necessity of handling relatively large quantities of HCl and HNO_3 and the time required for evaporating the acids and recovering the platinum, this method has been abandoned.

The Cs^{137} glass is prepared from cesium carbonate by dry-mixing it with silica and other substances and fusing at 1200°C.

The processing rate for the FPDL Cs^{137} operation is about one megacurie per year and is dependent primarily on the shipping schedule for the feed. Four shipping casks are presently in use; each will transport 50 kcuries per shipment. One shipping cycle requires eight weeks.

Cerium-144. The cerium product is sintered pellets of $Ce^{144}O_2$, with a density of 5.5 g/cm^3 and an activity concentration up to 300 curies/g.

The feed to the Ce^{144} process is a double sulfate of cerium and other rare earths, which is shipped as a dry solid in shielded filter casks from HAPO to the FPDL. Single shipments of 500 kcuries of Ce^{144} can be made.

The solid cake is removed from the casks by slurrying with water followed by rinsing with HNO_3. The slurry is dissolved in HNO_3, and

Ce^{144} and other rare earths are concentrated by precipitation as the oxalates. A 1 M HNO_3 solution of these oxalates is fed to the first solvent extraction cycle, in which Y^{91} and stable Y isotopes are extracted into 0.5 M di-3-ethylhexyl phosphoric acid and discarded. Cerium in the aqueous phase is oxidized to Ce(IV) by $KMnO_4$, extracted into 0.5 M D2EHPA in Amsco*, and then reduced to Ce(III) with H_2O_2 and stripped with 2 M HNO_3. From this solution cerium oxalate is precipitated in 20-kcurie batches in the manipulator cells and calcined to CeO_2. The oxide is pressed into pellets.

Promethium-147. The aqueous phase from the second solvent extraction cycle, in which cerium is recovered, contains Pm^{147} and stable rare earths. These are precipitated as the oxalates and dissolved in HNO_3 to yield a feed solution for the ion-exchange separation of Pm^{147} whenever the latter is needed.

Waste Processing. The FPDL does not currently process raw acid wastes; however, the facility retains the ability to handle such feed streams. Both Redox and Purex types of waste can be processed. The Cs^{137} is removed from the feed stream by co-crystallization on ammonium alum, and the other fission products are then concentrated by a series of precipitations at high pH with iron as a carrier (magnetite precipitation). The magnetite precipitates are dissolved in HNO_3 and the fission products are separated by a series of controlled-pH precipitations with NH_3. Fractions containing Ru^{106}-Fe, Ce^{144}-rare earths, and Sr^{90} are obtained. After further purification and concentration, the final products are processed in the manipulator cells. A processing

*Amsco-125, boiling point range 342-404°F.

rate of 500 gal of acid waste per day can be maintained.

Maintenance and Containment. The FPDL facility was designed to permit direct maintenance, and all high-activity-level cells are entered through top plug openings. The various techniques that have evolved for removing major equipment such as agitators allow a semiremote maintenance system and limit the cell downtime to a maximum of 5 days. Major piping revisions are made by personnel in plastic airline suits supplied with chilled air to increase the working time to the maximum allowable from the radiation standpoint. Contamination control on personnel leaving the FPDL cells has been greatly enhanced by washdown and removal of the suit at the cell entrance. Since standard monitoring instrumentation can detect only large quantities of beta emitters such as Sr^{90} or Ce^{144}, maximum protection is given to personnel in all suspect areas.

The containment philosophy used in the FPDL facility is shown in Fig. II-10. The 4-ft concrete cell walls form the primary containment layer and are held at 1.5 in. negative water pressure under normal conditions. The building wall forms the secondary containment shell and is under a slight negative pressure during normal operations. All air entering the building is absolute-filtered, and air leaving the cells is absolute-filtered before entering the ORNL cell-ventilation system. In an emergency, high-velocity air monitors signal an alarm which actuates the automatic containment system. The fan shuts down, inlet air dampers close, an additional cell ventilation damper opens, and the building pressure is reduced to 0.5 in. negative water pressure in less than 10 sec. Under these conditions the cells are held at 0.8 in. negative water pressure with respect to the building.

Fig. II-10

STABLE ISOTOPES

Although popular conception of the atomic energy program associates the isotopes business with the glamorous "twinkling atoms," the increasing importance of separated stable isotopes should not be overlooked; these silent partners play a rather important role in the overall applications program. Figure C1 shows the growth of the program as indicated by the number of annual shipments since 1946.

Most stable isotopes are separated in production-type electromagnetic devices called "calutrons,"* or to a limited extent by other methods, including thermal diffusion, centrifugation, and countercurrent fused salt migration. By far the most versatile of these is the calutron.

History

Since the replacement of the electromagnetic process for separating U^{235} from U^{238} by the gaseous diffusion method in 1946, a small part of the original electromagnetic facilities at Oak Ridge has been kept in operation specifically for the separation of enriched stable isotopes. Associated with the separations has been a development program for improving the efficiency of the separation process itself, as well as continuous efforts to improve the chemical procedures involved both in preparing charge materials and in refining the separated isotopes. In 1958 an expansion program was started to reactivate a number of beta calutrons which had been used for U^{235} production during

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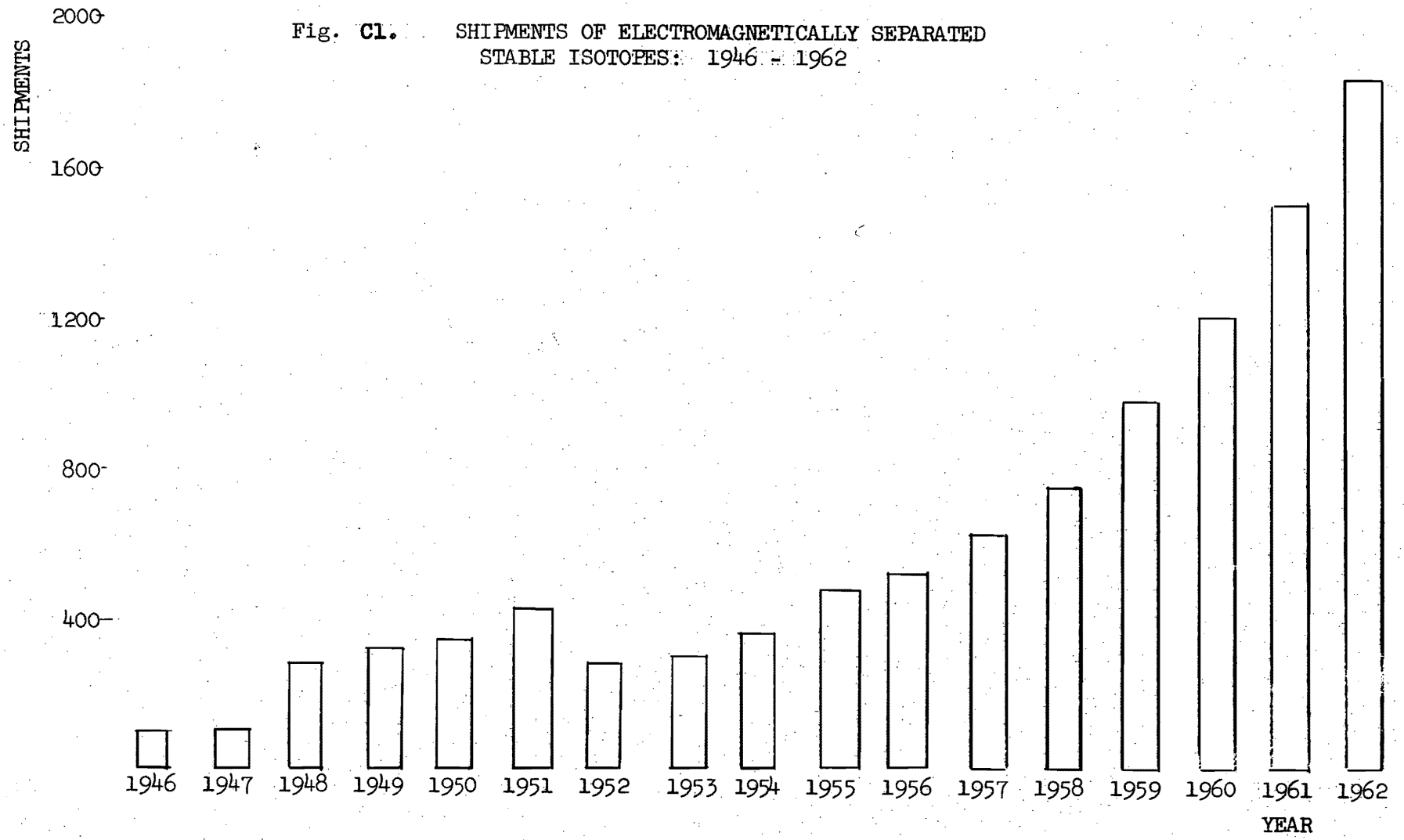
*The term "calutron" designates the production-type mass spectrometer, the term having been coined at the time of its development by E. O. Lawrence at the University of California.

Fig. C1. SHIPMENTS OF ELECTROMAGNETICALLY SEPARATED STABLE ISOTOPES: 1946 - 1962



World War II. Figure C2 shows the "track" being modified. At present, 28 units are in operation, including the original four which have been in continuous use since 1946. The separation capacity has been increased markedly; as a result of the larger scale of operation, the separated isotopes are, in general, being produced more cheaply.

The basic principle behind the electromagnetic separation of isotopes is identical with that of an analytical mass spectrometer. Both depend on high potentials for acceleration, magnetic fields for separating isotopic ion species, ion sources for ionizing the charge materials, and receivers for retaining the separated ions, all operating in a vacuum. The notable difference is that the calutron is designed to collect usable amounts of separated isotopes, while the laboratory spectrometer excels as an instrument for measurement. This does not rule out the use of the latter for collecting very small amounts of virtually any isotope, although the collection rate would be very low.

Figure C3 is a schematic diagram illustrating the electromagnetic separation process. Charged ions are accelerated by voltages of up to 40 kv. As these rapidly moving, singly charged ions move into the magnetic field surrounding the source and receiver, they are deflected or "bent," the amount of deflection for a particular field strength and potential depending only on the masses of the ions. Thus, isotopes of a particular element traverse slightly different paths and can be intercepted in such a way as to accomplish a separation.

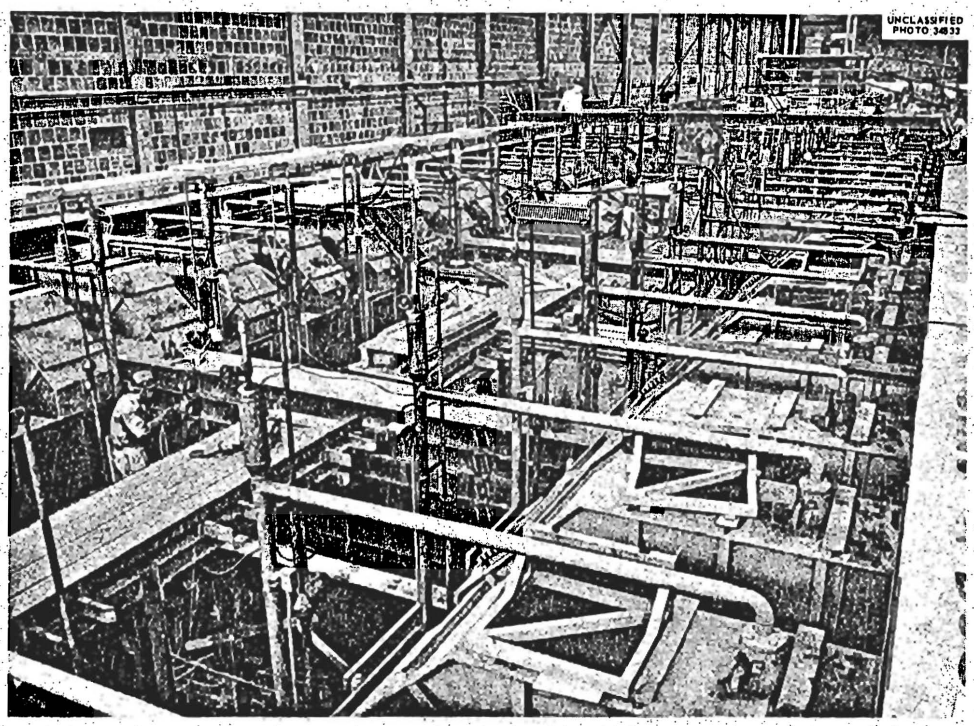


Fig. C2. A "Track" of 36 Calutron Units Being Modified for Use in the Separation of Stable Isotopes. Seventy-ton steel yokes are being installed to improve the versatility of the facility.

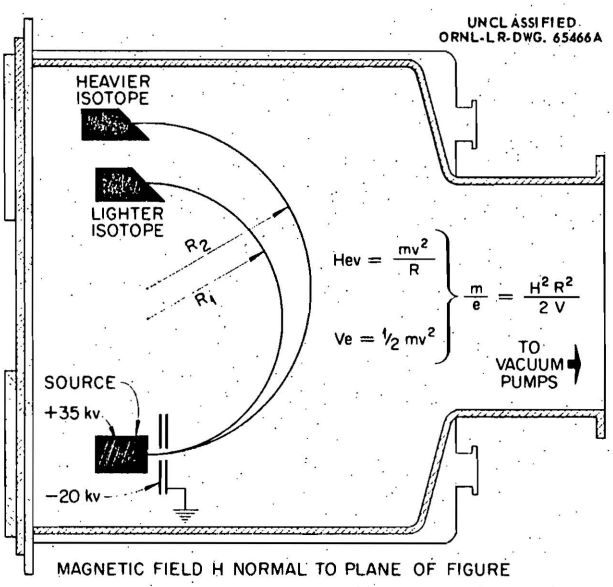


Fig. C3. Illustration of the Process of the Electromagnetic Separation of Isotopes. A beam of positively charged ions is produced in the ionization chamber and passes through slits in the accelerating electrodes, where the ions are accelerated by an electric potential applied between these electrodes. The ion beam then enters a magnetic field, acting in a direction perpendicular to the plane of the diagram, so that the beam is bent through a semicircle. In a fixed magnetic field the paths of the particles depend upon the masses of the particles; the heavier the mass, the less the deflection.

Fig. C4. A "Charge Bottle" Being Placed in "Source," Where the Ions Are Formed Prior to Being Accelerated into the Magnetic Field.

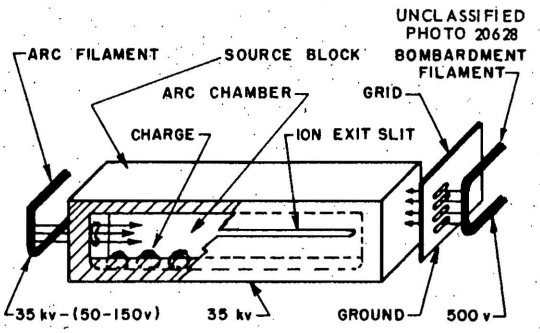


Fig. C5. Bombardment-Heated Source Used for Charge Materials Requiring Extremely High Temperatures for Evaporation.

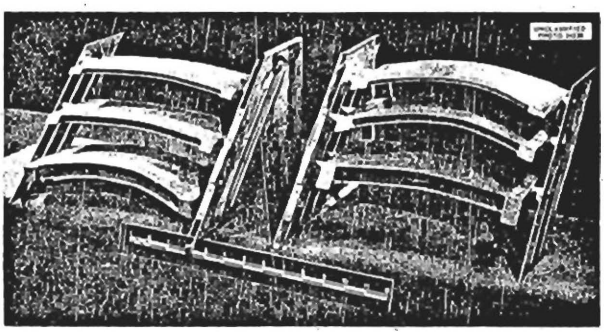


Fig. C6. Typical Receiver Pockets for the Collection of Isotopes. The slotted pieces in each assembly were machined from solid graphite. The spacing between the slots depend upon the isotopes being collected.

Fig. C7. Receiver Pocket Being Assembled. This particular pocket was used for the separation of ruthenium isotopes.

Shown in Fig. C4 is a "source" in which the ions are formed prior to being accelerated into the magnetic field. The element to be processed (or a suitable compound) is usually evaporated by resistance heating of a "charge bottle," and the vapor is subjected to an ionizing beam of electrons, which dislodges electrons from the atoms, leaving them positively charged. These ions are now ready for acceleration into the magnetic field. Typical charge materials are listed in Table C1.

Occasionally, as in the case of the platinum metals, extremely high temperatures are required for evaporation, and the heat is supplied by electron bombardment of the element. A drawing of such a bombardment-heated source is shown in Fig. C5.

Once the individual isotope beams have been separated by the magnetic field, they enter the receiver "pockets" (Figs. C6 and C7). Figure C8 shows the source and the receiver in their relative positions, and Fig. C9 shows the units about to be positioned in the vacuum chamber and the magnetic field of a calutron.

Various receiver-pocket materials may be used, depending on the element being processed and the ultimate chemistry involved in recovering the separated isotopes. For example, silver receiver pockets are used to collect mercury, which amalgamates with the silver, improving the retention. Graphite is used most frequently since the carbon can be burned in oxygen, leaving the residual oxide of the collected isotope. Copper is used in cases where simple leaching with dilute acid is feasible, as with calcium.

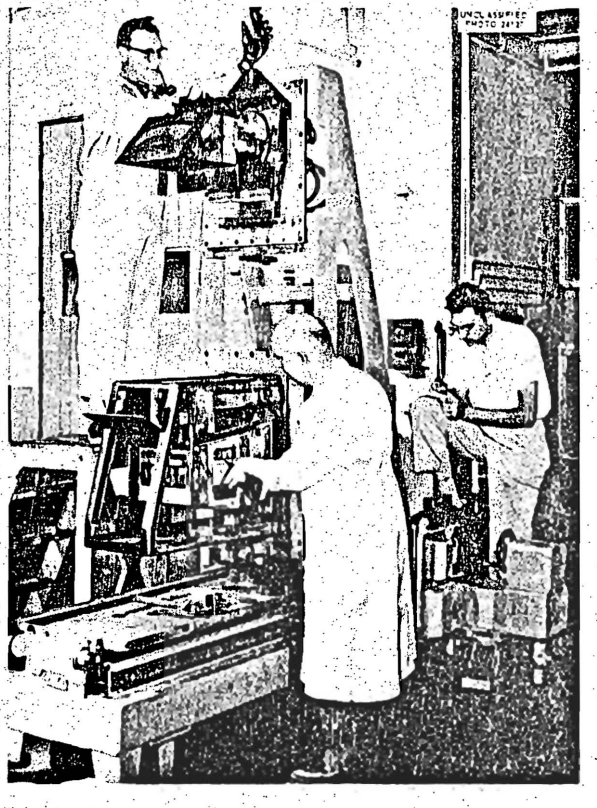


Fig. C8. Source (Lower Unit) and Receiver (Upper Unit) Being Assembled on Jig Prior to Insertion into Vacuum Chamber.

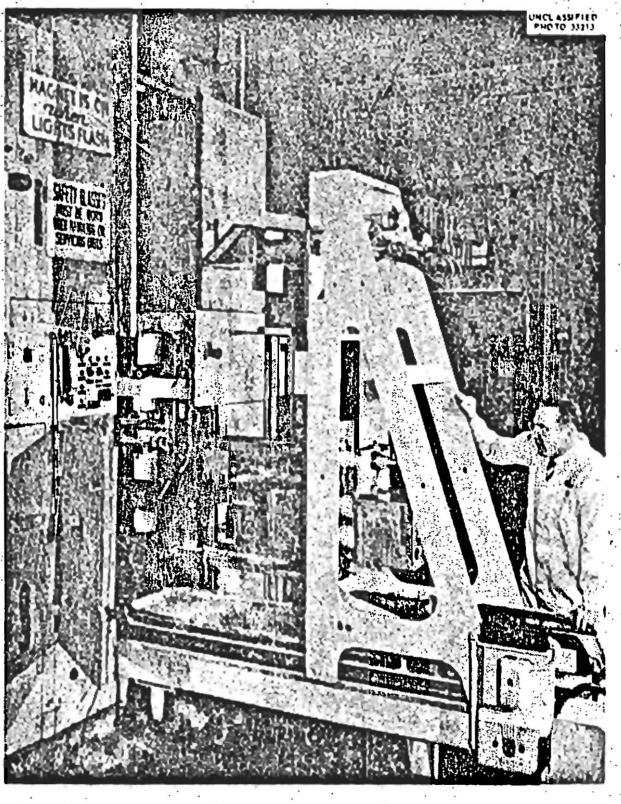


Fig. C9. Source and Receiver Being Loaded into the Vacuum Chamber of Production-Type Mass Spectrometer (Calutron).

Table 1. Typical Charge Materials

Element	Charge Material	Element	Charge Material
Antimony	Sb; Sb_2O_3; Sb_2S_3; SbI_3	Nickel	$NiCl_2$; $Ni(CO)_4$
Barium	Ba; $BaCl_2$; $BaBr_2$	Platinum	Pt[b]
Bromine	$SrBr_2$[a]	Potassium	KI; K; KBr; KCl
Cadmium	$CdCl_2$; $CdBr_2$; CdI_2; Cd	Ruthenium	Ru[b]
Calcium	Ca; $CaCl_2$; CaI_2	Silicon	$SiCl_4$
Cerium	$CeCl_3$	Silver	$AgCl$; Ag; $AgBr$; AgI
Chromium	$CrCl_3$	Strontium	Sr; $SrBr_2$
Copper	$CuCl_2$; Cu_2Cl_2	Tellurium	TeO_2; Te; $TeBr_4$
Europium	$EuCl_3$	Tin	$SnCl_2$; $SnCl_4$
Iron	$FeCl_2$; $FeCl_3$	Titanium	$TiCl_4$
Lead	$PbCl_2$; PbI_2; $(C_2H_5)_4Pb$	Tungsten	WCl_6; WBr_6; WF_6; WO_3
Magnesium	Mg; $MgCl_2$; $MgBr_2$; MgI_2	Zinc	Zn
Mercury	$HgCl_2$; HgO	Zirconium	$ZrCl_4$

[a]Strontium is also collected.
[b]Electron bombardment source.

Unfortunately, all the ions that enter a receiver pocket are not retained. Some of them bounce out again before "sitting down." Others, by physical erosion or by evaporation, dislodge some of the material already collected. As a result, the amount of material actually retained in the receiver pocket may vary from 10% for some antimony and magnesium runs to >99% for some platinum runs. As a matter of fact, from 1946 to 1956, only 56% of the ions entering receiver pockets were retained as separated isotopes in the ORNL separators.

Table C2 lists the isotopes of several elements and includes typical data to show operating parameters and results of the separa-

Table C2. Typical Operating Parameters and Results of Isotope Separations

Isotope	Average Beam Current (ma)	Receiver Pocket	Natural Abundance (%)	Enriched Abundance (%)	Product Recovered (mg/hr)
Copper	100	Graphite			30
Cu^{63}			69.0	99.9	
Cu^{65}			31.0	99.6	
Iron	100	Graphite or copper			100
Fe^{54}			5.9	93.8	
Fe^{56}			91.6	99.8	
Fe^{57}			2.2	85.0	
Fe^{58}			0.33	75.0	
Platinum	9	Graphite			25
Pt^{190}			0.012	0.8	
Pt^{192}			0.78	13.8	
Pt^{194}			32.8	65.0	
Pt^{195}			33.7	60.1	
Pt^{196}			25.4	65.9	
Pt^{198}			7.2	60.9	
Potassium	30	Copper			10
K^{39}			93.2	99.9	
K^{40}			0.012	7.7	
K^{41}			6.7	99.0	
Silver	45	Copper			50
Ag^{107}			51.4	99.0	
Ag^{109}			48.6	99.2	
Zinc	40	Copper			40
Zn^{64}			48.9	99.8	
Zn^{66}			27.8	98.8	
Zn^{67}			4.1	92.4	
Zn^{68}			18.6	98.5	
Zn^{70}			0.63	78.3	

Table C3 shows the results of modifications in operating techniques as exemplified by higher grade products. Some of these improvements are:

1. Better feed materials: higher purities, e.g., removal of the 1% Sr in ordinary Ba feed to decrease sparking; and better evaporation characteristics, e.g., the use of completely anhydrous $FeCl_2$, prepared by fusing and outgassing commercial-grade $FeCl_2$.
2. Refrigeration of vacuum-chamber walls to lower the vapor pressure of unresolved charge material that has been evaporated into the system.
3. Use of dual filaments in the source. The operating time between shutdowns is increased by having a second filament ready to operate when the first one burns out.
4. Introduction of an electrom bombardment source to increase the vaporization (and hence beam current) of low-vapor-pressure charge materials such as the platinum metals.
5. More precise monitoring methods. Continuous and careful manual control of operating parameters is essential to good separation. This control is directly dependent on the sensitivity of the monitoring system and the ability of the operators to discern fluctuations.
6. Improved receiver-pocket design for better retention; improved geometry and increased cooling; and sputtering receiver materials onto cold surfaces, thereby improving retention and reducing contamination.

Table C3. **Improvements in Isotopic Purities**

Isotope	Natural Abundance (%)	Enriched Abundance (%)	
		Original	Improved
Ag^{107}	51.4	88.9 (1946)	98.8 (1955)
Ag^{109}	48.6	95.8 (1946)	99.9 (1955)
Eu^{151}	47.8	78.7 (1955)	97.8 (1956)
Eu^{153}	52.2	86.4 (1955)	99.9 (1956)
Fe^{54}	5.9	42.8 (1946)	92.7 (1954)
Fe^{56}	91.6	98.7 (1946)	99.7 (1954)
Fe^{57}	2.2	56.1 (1946)	72.6 (1954)
Fe^{58}	0.33	22.0 (1946)	81.5 (1954)
Cu^{63}	69.0	97.0 (1945)	99.9 (1960)
Cu^{65}	31.0	93.1 (1945)	99.6 (1960)

Table C4. **Second-Pass Separations**

Isotope	Natural Abundance (%)	First-Pass Enrichment (%)	Second-Pass Enrichment (%)	Abundance of Undesirable Isotopes (ppm)	
				After 1st Pass	After 2d Pass
Fe^{56}	91.6	99.7	99.987	3×10^3	130
Cr^{52}	83.7	99.5	99.97	5×10^3	300
Li^6	7.5	95.3	99.999	4.7×10^4	10

As knowledge applicable to recovery problems has accumulated through experimentation in various separation groups, the retention of isotopic material has gradually increased. Over the past two-year period, retention has averaged 80%. The net gain in retention cannot be attributed to any one single factor but represents the combined effects of engineering features incorporated to offset the losses associated with the sputtering of receiver pocket walls and the low retention of isotopes on surfaces struck by ions.

In some investigations into retention problems ion currents associated with the more abundant isotopes and relatively high current densities have been used. Several studies have recently been made with a less abundant isotope, Ca^{46} (natural abundance 0.0033%). Investigations have used the Ca^{46} ion beam at a current density of only 0.22 $\mu a/cm^2$ on graphite, copper, and crystalline copper having the 110 and the 112 crystal faces set perpendicular to the incident ion beam. The amount of Ca^{46} striking the target surface was monitored, and quantities and purities of Ca^{46} recovered from the target area and from the pocket area were determined. In Fig. C10 the quantity and purity of Ca^{46} recovered from the surfaces of targets made of normal copper, purified graphite, and a 110 copper crystal are compared. The purity of samples recovered from each surface is essentially constant, but there appears to be more Ca^{46} retained per unit area on graphite than on either copper or a 110 copper crystal. In Fig. C-11 the isotopic purity and the ratio of wanted to unwanted isotope are compared to show equality of target materials from this standpoint.

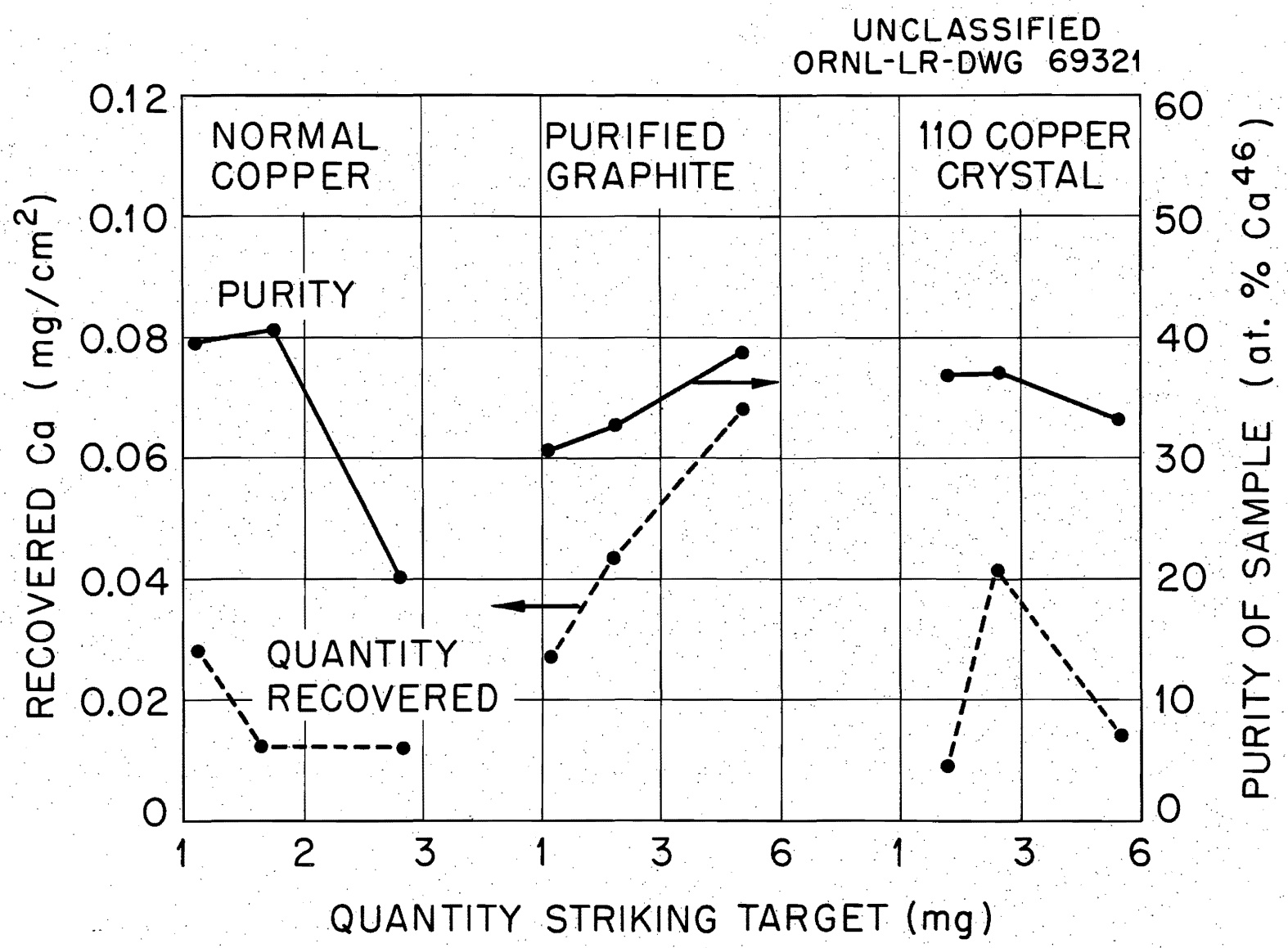


Fig. C10 Comparison of Purity and Quantity of Ca^{46} Recovered From Various Target Surfaces.

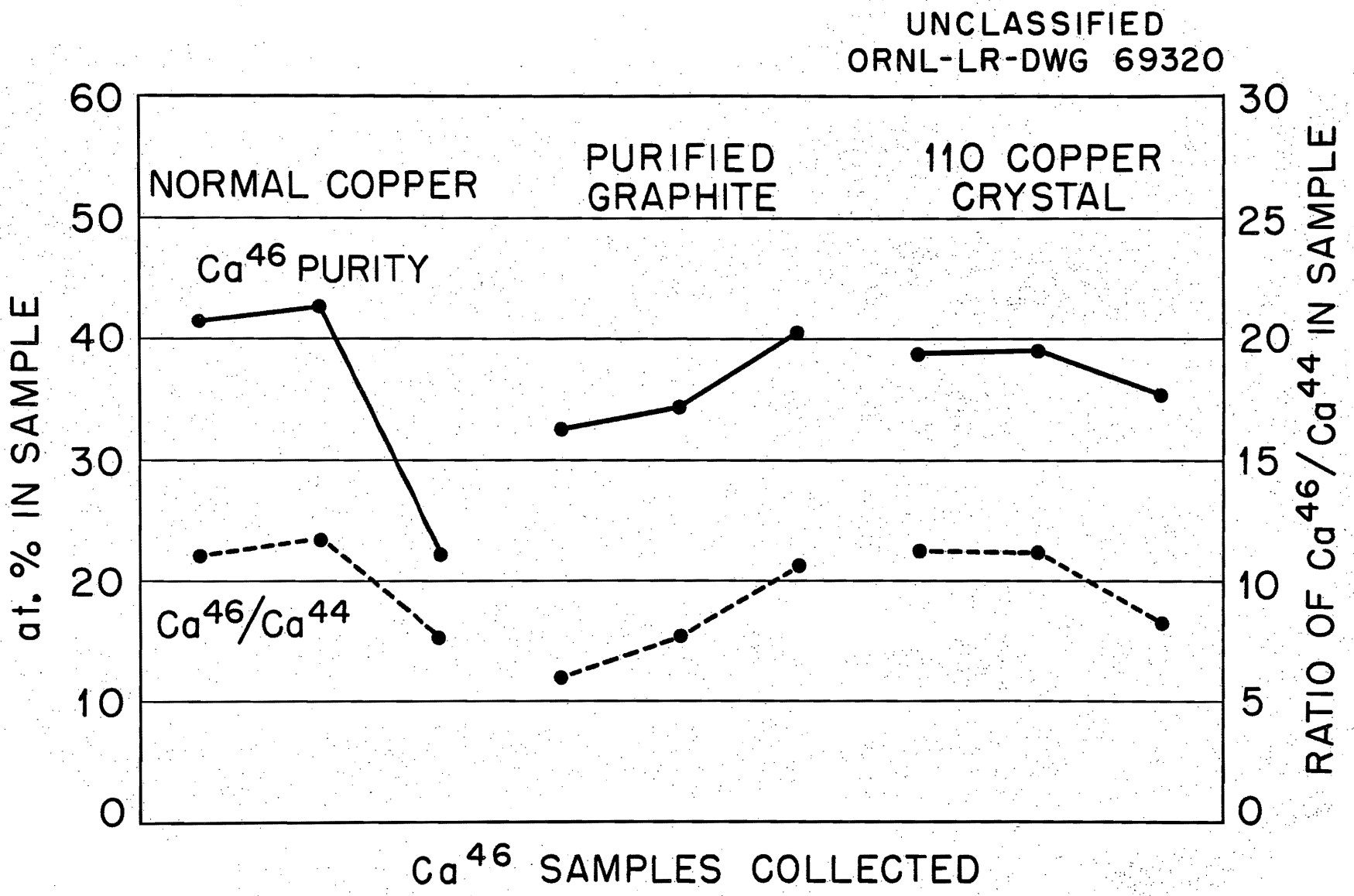


Fig. C11 Comparison of Purity and Ca^{46}/Ca^{44} Ratio Obtained on Various Target Materials.

In Fig. C-12 the isotopic purity of Ca^{46} recovered from the target area is compared with that recovered from the pocket area. Extreme differences in purity are noted, and in every determination the Ca^{46} recovered from the area struck by the beam is of higher purity than that recovered from the pocket area. In Fig. C-13, where the ratios of wanted to unwanted isotope in the recovered samples are compared, the trend noted in the previous data is again supported; namely, a better quality sample is obtained by selectively recovering the material from the area struck by the ion beam.

Although there are insufficient data to prove that oriented copper crystals have an advantage over other collector materials, there is scattered evidence that they offer some, even if slight, improvement. The actual processes producing such improvement are not yet understood, but they are sufficiently important to warrant further efforts to establish definite conclusions.

A special collector has been designed and is currently being used in the calcium separation to determine saturation values for Ca^{46} on various copper crystals. The receiver is constructed so that four different targets can be mounted on a rotating holder. The holder rotates 90 deg, holds for a few minutes, and then repeats. Each target is thus in position so that the beam strikes it normal to its surface for 6 min of each rotation. Since operating conditions vary from run to run, this technique provides comparative data that are independent of operating conditions for different receiver materials.

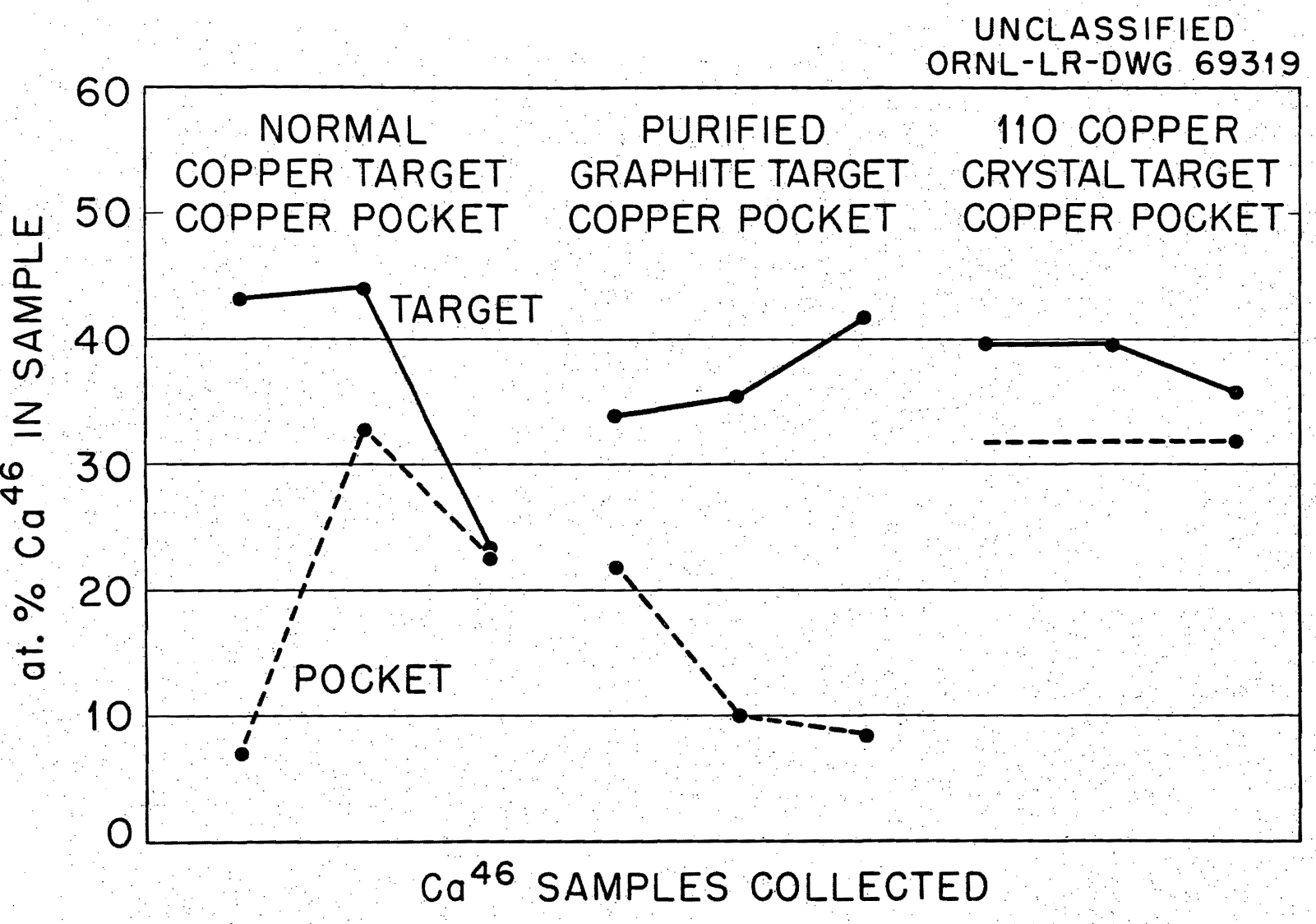


Fig. C12 Comparison of Isotopic Purity of Ca^{46} Recovered From Target and From Pocket Surface.

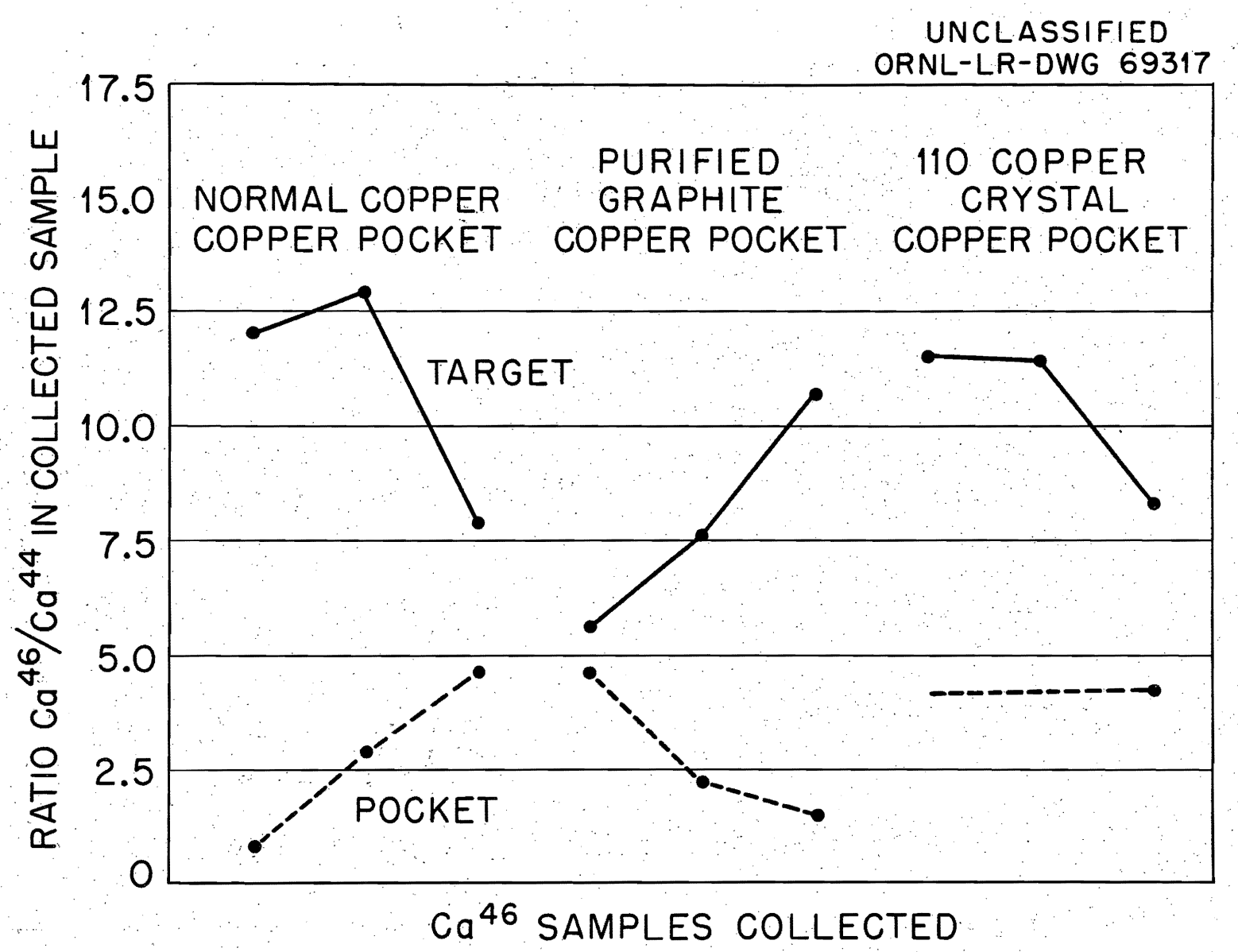


Fig. C13 Comparison of Ca^{46}/Ca^{44} Ratio in Samples Recovered From Pocket Surface.

These investigations are to be extended to include copper crystals of various orientations and other materials suitable for use in receiver construction or target formation. As background knowledge of sputtering and saturation values increases, a continued upward trend in retention is anticipated along with the increased purity attainable through selective sample recovery.

Table C4 lists isotopes that have undergone "second-pass" separations and shows original, imtermediate, and final enrichments. The receiver design is usually modified slightly to favor the wanted isotope.

The procedures for chemical recovery usually involve rather straightforward inorganic techniques. Lead, for example, is recovered by leaching copper receiver pockets with HNO_3, electrodepositing the lead at the anode as PbO_2, dissolving the PbO_2 in hot HNO_3 and H_2O_2, and finally crystallizing high-purity $Pb(NO_3)_2$ from 85% HNO_3. Copper is recovered by igniting the graphite receiver pockets in oxygen, dissolving the ash in HNO_3, and electrolyzing the copper. This copper is then redissolved in HNO_3, precipitated with cupferron, and ignited to CuO.

Silicon is collected in graphite. The resulting silicon carbide, along with the pocket, is burned in oxygen, and the SiO_2 residue is fused with Na_2CO_3. The melt is then dissolved in water, and SiO_2 is precipitated by acidification and digestion with $HClO_4$.

The chemical recovery of rare earths often involves ion exchange purifications to remove adjacent rare-earth impurities. Even then,

high-purity products are difficult to obtain.

Table C5 lists typical product forms for 26 elements. Table C6 shows the elements separated in the years 1946-60.

One of the most significant contributions of this phase of the isotopes program is the use of enriched stable isotope targets for the production of radioisotopes. Shorter irradiation times, higher specific activities, and freedom from undesirable extraneous activities are characteristic of radioisotopes prepared by bombardment of separated stable nuclides. The decreased extraneous activity usually results from both increased concentration of desired target nuclide and decreased concentration of undesired target nuclide. Table C7 lists a few of the enriched nuclides used for radioisotope production, along with information to show the advantages of using these targets rather than normal material. The same advantages apply with respect to cyclotron-produced radioisotopes.

These advantages are illustrated nicely by Ca^{47}. When prepared from the 0.003% Ca^{46} in normal calcium, the 4.5-day isotope is completely overshadowed (by a ratio of about 25:1) by the 164-day Ca^{45} produced by irradiation of the 2.1% Ca^{44} also present. However, if the Ca^{46} is enriched to, say, 30%, the Ca^{44}/Ca^{46} ratio is reduced to about 1:5, and the resulting Ca^{45}/Ca^{47} ratio becomes about 1:125; thus a useless product which is 96% undesirable Ca^{45} can become a very useful product which is 99% Ca^{47}.

Table C8 shows some typical uses for stable nuclides.[5] A recent bibliography covering these and other applications is included in ORNL-3266.[6]

Table C5. Typical Product Forms

Element	Product Form	Element	Product Form
Antimony	Sb	Nickel	NiO; Ni
Barium	$Ba(NO_3)_2$; $BaCO_3$	Platinum	Pt
Bromine	AgBr; NH_4Br	Potassium	KCl; K_2SO_4
Cadmium	CdO; Cd	Ruthenium	Ru
Calcium	$CaCO_3$	Silicon	SiO_2
Cerium	CeO_2	Silver	Ag; AgCl
Chromium	Cr_2O_3	Strontium	$SrCO_3$; $Sr(NO_3)_2$
Copper	CuO; Cu	Tellurium	Te
Europium	Eu_2O_3	Tin	SnO_2; Sn
Iron	Fe_2O_3; Fe	Titanium	TiO_2
Lead	$PbCO_3$; PbO; Pb	Tungsten	WO_3
Magnesium	MgO	Zinc	ZnO
Mercury	$Hg(NO_3)_2$; Hg	Zirconium	ZrO_2

Table C6. Elements Separated During the Years 1946–62

1946	Br[a], Cd[a], Ca[a], C[a], Cr[a], Cu[a], In[a], Fe[a], Pb[a], Li[a], Mg[a], Mo[a], Ni[a], K[a], Se[a], Si[a], Ag[a], Sr[a], Sn[a], Zn[a], Zr[a]
1947	Sb[a], B[a], Cd, Ca, Cu, Ge[a], In, Fe, Pb, Li, Mg, Hg[a], Mo, Ni, K, Se, Si, Sr, Te[a], Tl[a], Sn, W[a], Zn, Zr
1948	Ba[a], Ca, Ce, Cl[a], Cu, In, Fe, Pb, Li, Hg, Ni, O, K, Re[a], Se, S[a], Sn, W, Zr
1949	B, Ce, Cu, Fe, La[a], Li, Mg, Hg, K, Se, Si, Ag, Te, Sn, Zr
1950	Br, Ca, C, Cr, Ga[a], Hf[a], In, Fe, La, Pb, Li, Nd[a], K, Rb[a], Sm[a], Sr, S, Ti, W, V[a], Zn
1951	Ca, C, Ce, Cu, Fe, Li, Ni, Sm, Se, Sr, Te, V
1952	Ba, B, Cl, Ge, Pb, Li, Mo, Nd, Si, S, Sn, Ti
1953	B, Gd[a], Hf, Ir[a], La, Li, Nd, Pd[a], Pt[a], K, Ru[a], Sm
1954	Ca, Cr, Cu, Fe, Ni, Rb, Ta[a], Zr
1955	Ba, Br, Cr, Li, Mg, Mo, N[a], Sm, Ag, Sr, S, Te, Ti, W
1956	Sb, B, Cd, Ca, Cl, Cu, Dy[a], Eu[a], Ge, Pb, Li, Pd, Pt, Re, Rb, Ru, Si, Tl, Sn, Zn
1957	Fe, Ir, Ti, Cr, C, Er[a], Se
1958	V, Yb[a], Lu[a], Ca, K, Ag, Cu, Hf, C, Se, Mg, Gd
1959	Rb, Bi, Tl, Hg, Sr, Nd, Ni, Cu, Ge, Sm, Zn, In, Ba
1960	Pb, Cr, Os[a], Ta, Cu, Fe, Ga, Ni, Cd, Zn, K, Cl, Si, Br
1961	Yb, Zr, Cl, Mg, Sn, Sm, Ni, W, Gd, Lu, Ti, Ta, Hf, Ca, Mo, La, Re
1962	Nd, Si, Ca, Sr, Er, Fe, Te, Cu, Ta, Yb, Cd

[a]Separated for the first time.

NOTE: In a number of instances, an element was separated more than once in the same year.

Table C7. Enriched Stable Isotopes Used in Radioisotope Production

Radio-Isotope	Target Nuclide	Percent Abundance		Advantages	
		Normal	Enriched	A[a]	B[b]
Cd-109	Cd-108	0.88	70	80	Cd-113: 400 Cd-115: 550
Ca-45	Ca-44	2.1	98	47	--------
Ca-47	Ca-46	0.003	30	10,000	Ca-45: 3,000
Cr-51	Cr-50	4.3	95	23	---------
Fe-55	Fe-54	5.8	98	17	Fe-59: 18,000
Fe-59	Fe-58	0.31	76	250	Fe-55: 2,500
Ni-63	Ni-62	3.7	98	25	Ni-59: 2,500
Sr-85	Sr-84	0.55	54	100	Sr-89: 300

[a]A = The approximate factor by which specific activity can be increased for a given irradiation time, or by which irradiation time can be decreased for a given specific activity.

[b]B = The approximate factor by which the concentration of an extraneous (unwanted) radionuclide is reduced relative to the desired nuclide.

Table C-8. * TYPICAL USES FOR ENRICHED STABLE ISOTOPES

Target materials for preparing high-specific-activity radioisotopes in reactors and in accelerators. Enriched targets not only increase the number of activations per unit time of irradiation, but also reduce side activities introduced by other isotopes. Calcium-44, Cr^{50}, Cr^{54}, Fe^{54}, and Fe^{58} are examples of enriched isotopes used in producing radioisotopes.

Studying the isotope effects on bulk properties of elements. Specific heat, density, and the like have been studied for Li^{6} and Li^{7}; the isotopes of lead, mercury, and tin have been investigated in connection with superconductivity; Ni^{58} and Ni^{60} have been used to study the isotope effect on linear expansion.

Physicochemical studies, such as reaction kinetics and self-diffusion.

Starting materials in assigning masses to artificially produced radioactive isotopes. The results of a mass spectrometric analysis of irradiated material can be positively correlated to an activity only when the target material is mononuclidic. More than 100 such mass confirmations have been made in this way.

Isotope dilution analyses. Traces of enriched K^{39}, K^{41}, and N^{15} have been used in fertilizer uptake studies; K^{41} in geologic age determinations; V^{50}, Fe^{54}, and Ni^{62} in estimating trace impurities in crude oils.

Studying naturally occurring, low-abundance radioisotopes such as K^{40}, Sn^{124}, Rb^{87}, and Sm^{147}. It is much easier to evaluate the properties of these isotopes using enriched material than it is using the

normal element. This is particularly true in the case of the double-beta-decay studies of K^{40}.

Preparing calibration standards for mass spectrometers. "Blends" of various concentrations of essentially pure nuclides provide reference standards. An example is mixtures of enriched Li^6 and Li^7 to give a complete range of relative abundances.

Preparing radioisotopes for studies of decay schemes and characteristic radiations. Numerous highly enriched nuclides have been activated, and the investigators have thereby been spared the tremendous confusion which results from trying to unscramble the decay schemes of several species which are disintegrating simultaneously.

Medical use. Gallium-71 has been used in bone-cancer studies; K^{40} has been used in tissue studies of malignant lymphoma.

Sources of monochromatic light for optical calibration. Wavelength standards are preferable when uninfluenced by any isotope effect due to isotopic impurities. Cadmium-114, Hg^{198}, and Hg^{202} have been used as standards. The orange-red wavelength of Kr^{86} has been made the basis for the official definition of the meter.[7]

Optical studies of isotope shift, hyperfine structure, and nuclear spin. Hundreds of publications on these subjects exist.

Measuring neutron absorption, scattering, and total cross section. This probably involves the most concentrated efforts at the present time, using enriched samples of almost all the stable isotopes.

"Pushing back" the limits of natural abundance of certain isotopes. By analyzing material collected in the position for a "nonexistent" isotope, it is possible to ascertain with more assurance the lower

limits of possible abundance of a particular species. This has been done for Yb^{167}, Er^{172}, and Pb^{210}. The technique has also been used for Ta^{180} to help assign an actual value of abundance to an isotope previously thought to be nonexistent.

Spectrochemical standards. Spectrochemically pure Ca^{40} is recovered from the electromagnetic separation of calcium isotopes.

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*With minor changes, reprinted with permission from Chem. Eng. News 37(5): 64 (1959).

During the first ten years of the electromagnetic separations program, nearly one fourth of the isotopes were used to determine energy levels and another one fourth to measure various neutron cross sections.[8] Today much of the effort is involved in the various cross-section studies.[9,10] However, very interesting work continues on energy levels,[11] decay schemes,[12] and half-lives,[13] and on such other topics as hyperfine structure,[14] the measurement of bulk properties,[15] and Mössbauer studies.[16,17]

The use of enriched stable isotopes in isotope dilution as an analytical tool has good potential.[18] Their use in tracer work involving eventual activation analysis is also a powerful tool.[19] A unique use of B^{10} consists of introducing a soluble compound into the bloodstream of a patient with a brain tumor, allowing the isotope to concentrate in the tumor, and then irradiating the B^{10} with neutrons to provide an on-the-spot treatment of the tumor.

Targets

The versatility of the calutron is being exploited in still another way. Increasing demands for accelerator targets of high isotopic purity for use in nuclear studies led to investigation of a method for target preparation in the calutrons. Such a procedure would permit a one-step process and would result in less contamination of the target.

If an ion could be decelerated to an energy where it had a self-sputtering ratio of less than 1, a buildup would occur in the area of impact, thus offering a means of making targets within the calutron receiver. Based on this principle, a decelerating receiver (Fig. C14) was constructed to decrease ion energy by varying the potential of the receiver with respect to the source. A vertical grounded plate with a defining slit for beam entrance is established at the focal point of the imcoming beam, and the electric field that decelerates the ion beam is maintained between the collector target and the plate. Targets may be inserted in the receiver pocket so that direct deposition occurs, or they can be placed in such a way that material is sputtered onto the target backing.

Target size and lack of uniformity of the ion beam parallel to the magnetic flux made it necessary to devise means for moving target backing material through the beam. A mechanical device (Fig. C15) that uses rotational motion was constructed to systematically pass backing material through the beam impact area in order to produce targets of specific uniformity, size, and thickness. The target holder is offset

ORNL Photo. 36475

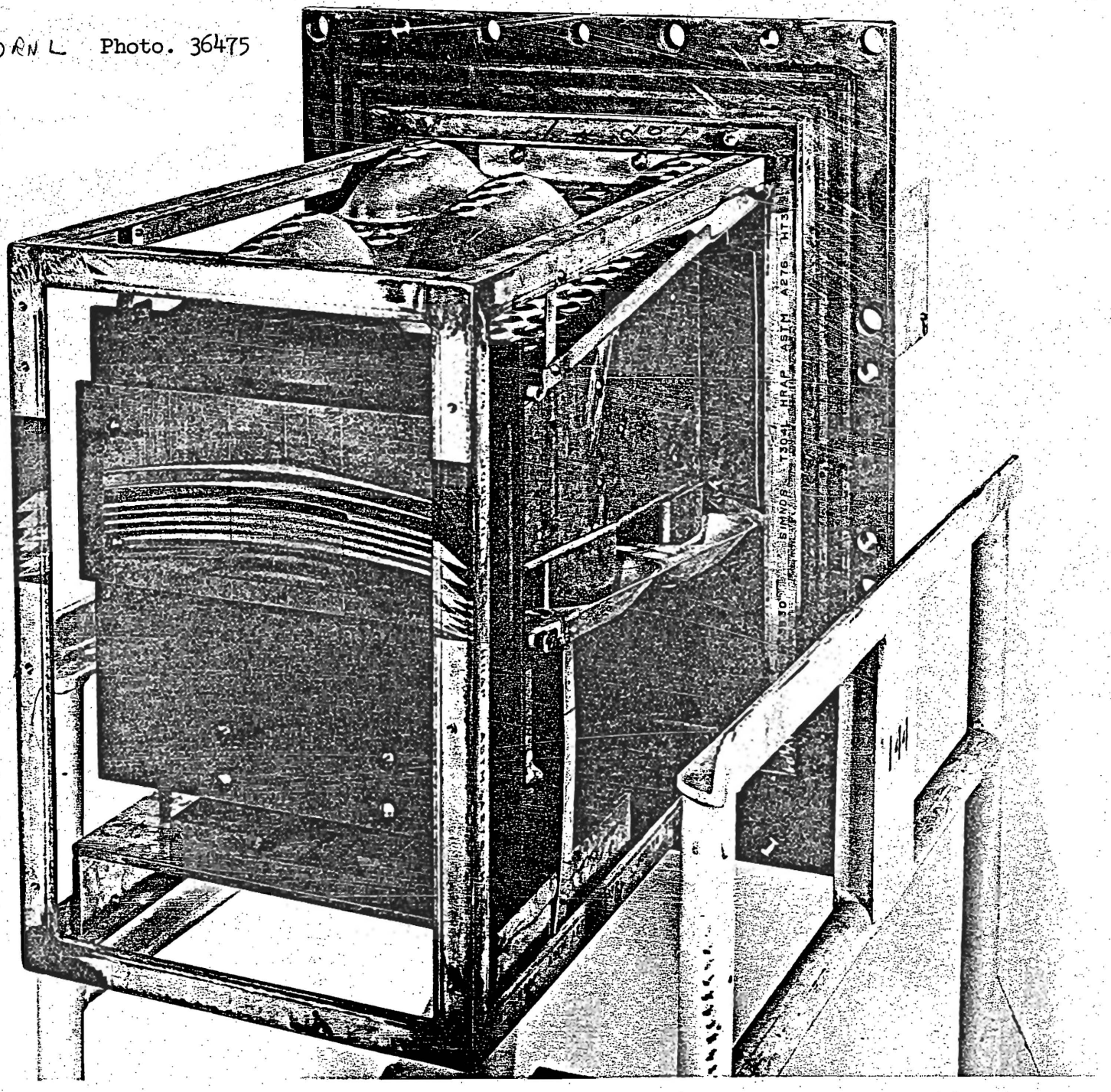


Fig. C-14 Decelerating Receiver for Calutron

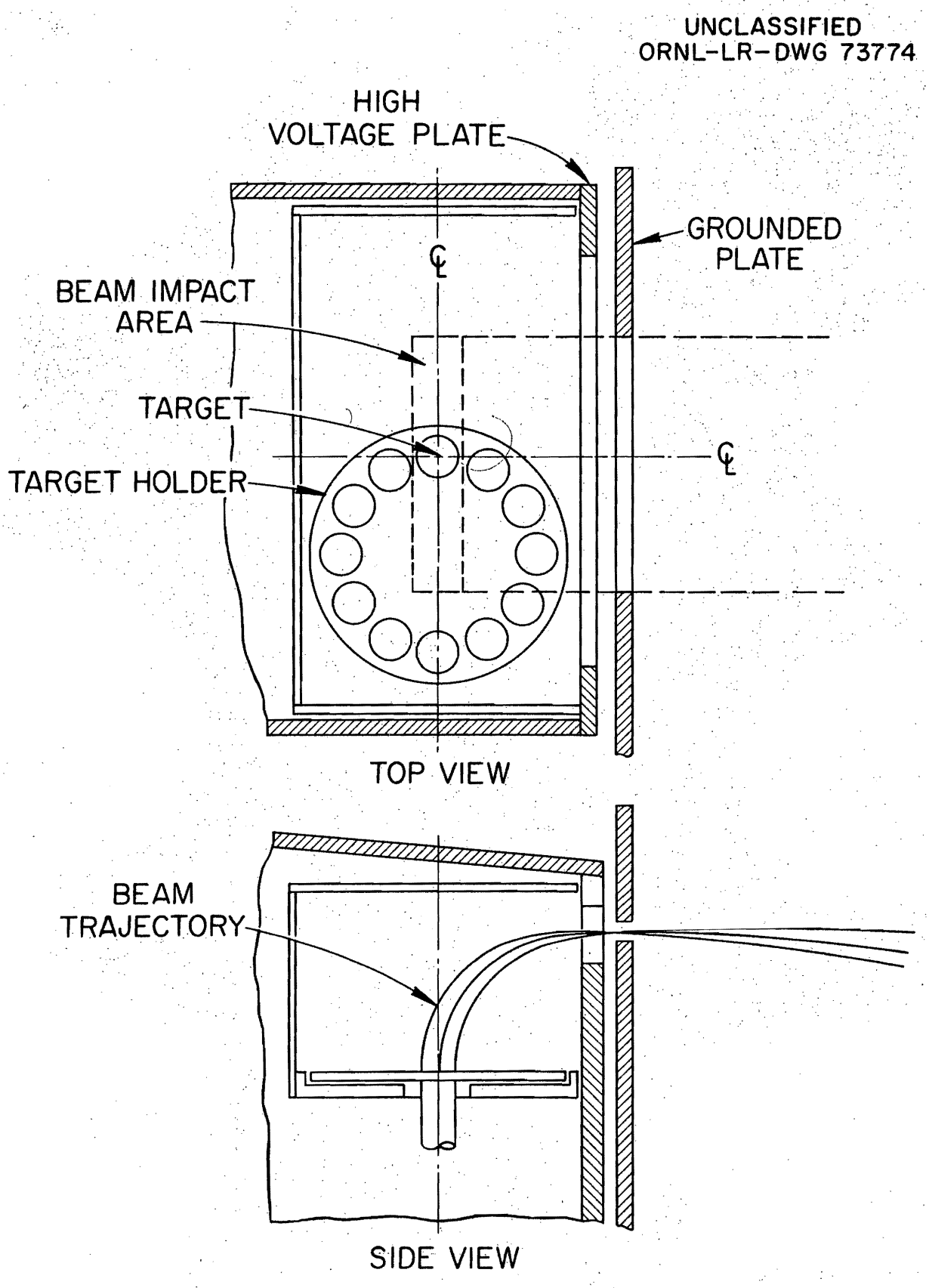


Fig. C-15 Target Holder in Decelerating Receiver

from the center of the receiver so that a series of targets mounted on it may be rotated through the center of the impinging, low-energy, ion beam. The device is powered by a synchronous motor geared to provide 33 rpm.

In early experimental runs, Ni^{58}, B^{10}, and B^{11} were deposited on tantalum backing material (Table C9). The technique was later used to deposit B^{10} on tungsten target substrates. The rotational device used here accommodated 12 tungsten disks each 0.75 in. dia and 0.030 in. thick, masked to 5/8 in. dia. The assembly was rotated so that each individual target remained in the ion beam for a specified time. Twenty-four target substrates were bombarded, each target being in the beam for 3 hr, during which the target area was struck by a 1-ma beam decelerated to 250 volts. The beam was accurately positioned by varying the decelerating voltage to produce the maximum current on a probe located adjacent to each target. The weight of material deposited on the target surfaces varied from 230 to 555 μg and averaged 387 μg.

Increasing requests for boron targets of high isotopic purity prompted investigation of the most feasible method of formation with respect to cost, uniformity, and purity. A grid type ion source used in the calutron for the mass range of boron produced a higher beam current with more uniformity than conventional sources. Also, the source potential through which the ionic particles were accelerated was lowered in the grid unit from 35 to 16 kv. This decrease directly affected the rate of

Table C9. Direct Deposition of B^{11}, B^{10}, and Ni^{58} on Tantalum

ISOTOPE	DEPOSITION TIME (hr)	SUBSTRATE	NO. OF SUBSTRATES BOMBARDED	AV. MONITORED CURRENT (ma)	WEIGHT DEPOSITED ($\mu g/cm^2$)	IMPACT ENERGY (ev)
B^{11}	25	Ta	5	6.5	254	200
B^{10}	27	Ta	5	2	196	200
B^{10}	3*	W	12	0.75	220	250
B^{10}	3*	W	12	0.75	165	500
Ni^{58}	8	Cu	2	3	150	200

RESULTS OF B^{10} DEPOSITION

ISOTOPE	PERCENTAGE ABUNDANCE	PERCENTAGE ISOTOPIC CONTAMINATION DUE TO B^{11}	METALLIC CONTAMINATION
10	92.2	7.8	>>1%

*Bombardment time per target.

target formation since the amount of material sputtered by particles striking a substrate is, in part, a function of the energy of the impinging ion.

For use with the large grid source (alpha type), a hexagonal cylinder ~24 in. long (Fig. C16) was designed to support the target substrates and to rotate them through the impinging ion beam. Target substrate material mounted on all six surfaces of the holder is continually exposed to the beam as the device is rotated. The cylinder is positioned in the collector so that the beam will dissipate energy over a wide area. Heat is removed from the target area by passing water through the rotating cylinder. The technique used for cooling involved design and development of "seal retainers" that would prevent water leakage into the high-vacuum region during rotation of the target holder.

The initial run in which the decelerating technique was used with the alpha grid and hexagonal target holder was made to determine whether very thick targets (1 mg/cm^2) could be formed. The targets on the rotating cylinder were bombarded for 50 hr at an average B^{10} current of 15 ma. A very thick layer was deposited, but the bond to the substrate was very weak and flaking occurred. In an 18-hr run, at an average B^{10} current of 12 ma to determine the qualities of targets with a B^{10} thickness of 100-200 μg/cm^2, deposits were uniform over each area. The bond between the deposited B^{10} layer and the substrate was quite strong, and the average B^{10} deposition on each of the 84 substrates was 100 μg/cm^2. The isotopic purities and chemical compositions of the deposits on several targets that were destroyed for analysis are shown in Table C10.

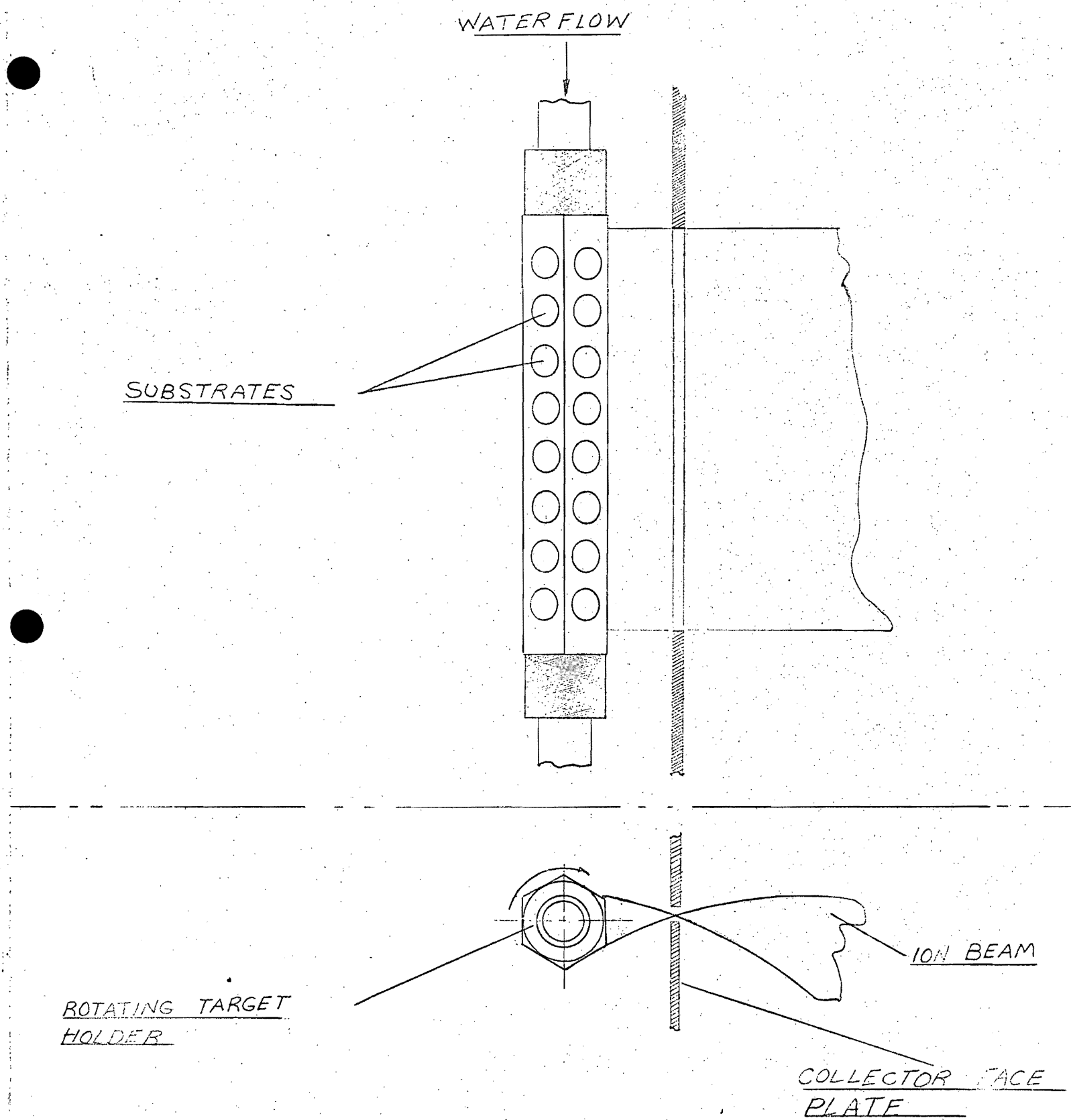


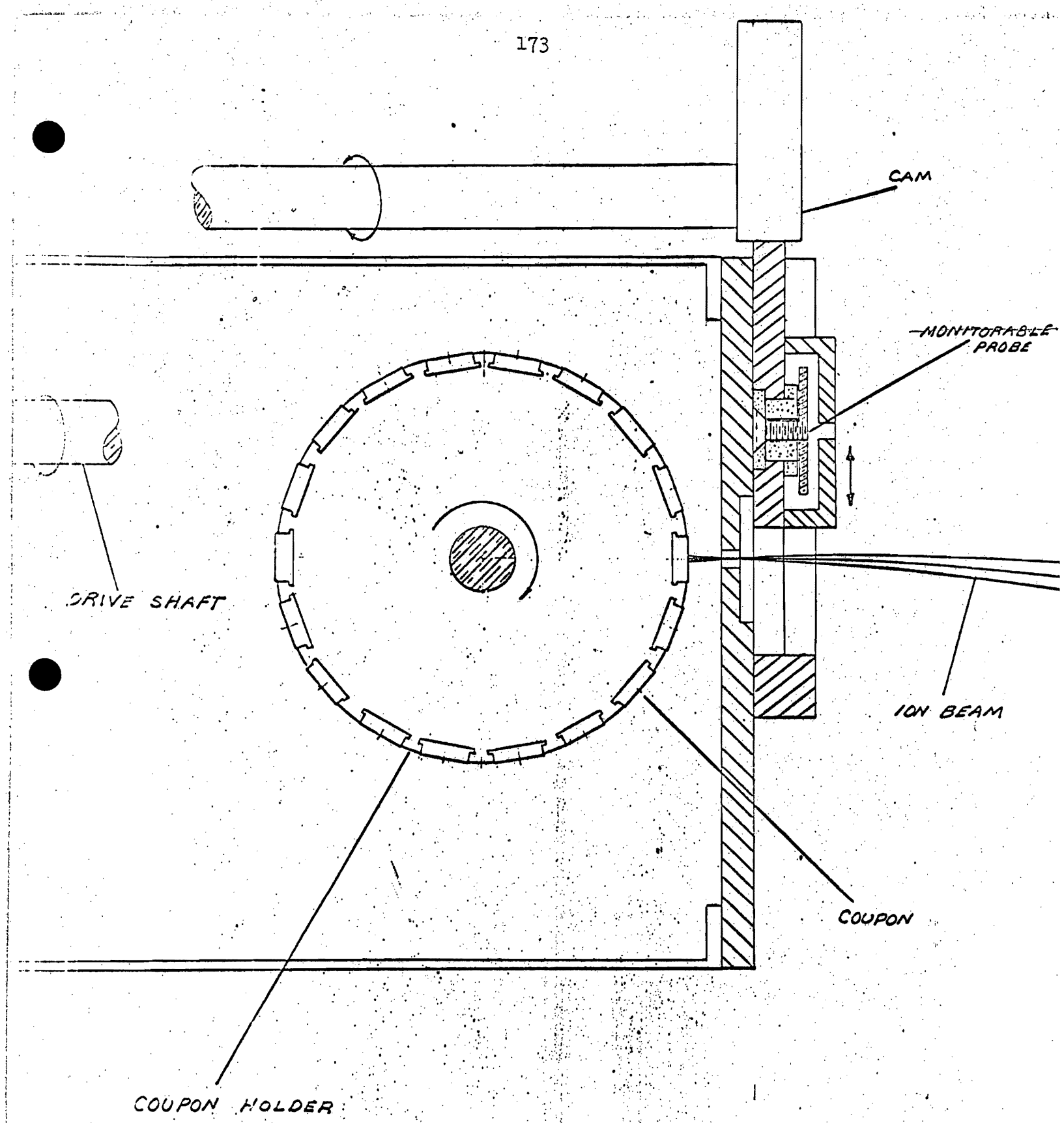
Fig. C-16 ALPHA GRID TARGET APPARATUS

Table C10. Chemical and Mass Analyses of Boron Targets

Isotope	Isotopic Purity (%)	Chemical Contamination (%)
	Alpha Grid Unit	
B^{10}	86	< 3
B^{11}	14	< 3
	Deceleration Unit	
B^{10}	92.2	< 1
B^{11}	7.8	< 1

Certain reactor problems are complicated by diffusion of elements through the graphite moderator. The study of this diffusion process might be expedited by using various materials containing other elements as surface layers. The electromagnetic separators have been used in preparing such samples for use in the experimental determination of diffusion coefficients for materials embedded in different substrates.

In order to achieve greater areal uniformity and make the process as economical as possible, a special collector was designed (Fig. C17). The substrates are supported in a motorized graphite holder 3 in. dia and 1.5 in. long. Fifty-four 3/8 x 3/8 in. graphite coupons 0.150 in. thick are supported simultaneously by the cylinder, which rotates about an



APPARATUS USED IN IMPREGNATING
GRAPHITE WITH THORIUM

Fig. C-17

axis parallel to the magnetic field. The beam bombards each coupon surface perpendicularly after passing through a 1/8 x 1.5 in. defining slot in the receiver faceplate. A movable probe (cam-actuated shutter type) is used to bring the beam up to proper intensity and stability before the shutter is opened and the substrates are bombarded.

In a 5-hr bombardment of 54 square coupons of pyrolytic graphite with Th^{232} ions, An 8.76-$\mu g/cm^2$ coating of Th^{232} was obtained. Uniform deposition was achieved by utilizing the rotating cylinder and accepting only a small centralized portion of the focused beam at the collector. The portion of the ion beam being accepted was 20 μa, and activation analysis showed that 88% of the monitored quantity of Th^{232} was retained by the surface. When Type C-18 graphite was bombarded 8 hr at 500 μa, the surfaces contained 65.6 $\mu g/cm^2$ of Th^{232}, which was equivalent to $\sim$17% of the material that impinged on the targets.

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FIGURE LI.

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	Fig. C7. Receiver Poc[illegible] Be[illegible]	[illegible]
	Fig. C8. Source (Lower Unit) [illegible] Upper Unit) Being Assembled on Jig Prior to Insert[illegible] into Vacuum Chamber	[illegible]
	Fig. [illegible] Source and Receiver [illegible] into the Vacuum [illegible]ber of Productio[illegible] Spectrometer (Calutron).	14[illegible]

ORNL-LR-DWG 69321	Fig. C10. Comparison of Purity and Quantity of Ca^{46} Recovered From Various Target Surfaces	152
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III. APPLICATIONS TECHNOLOGY

ANALYSIS

Process control by chemical or physical analysis is a very important and frequently a costly part of a manufacturing process. Much work is being done under the sponsorship of the Division of Isotope Development of the USAEC to develop ways for using radioisotopes to simplify and improve analytical methods both in industrial process control and in the determination of contaminants in our environment. New developments in the use of radioisotopes in analysis are reviewed biennially in Analytical Chemistry, and the number of references in these reviews reflects the large amount of activity in this field. The most recent review, by Leddicotte, in 1962,[A-1] and three earlier reviews by Meinke[A-2] contain over 4000 references; in the two most recent compilations, coverage is limited to selected articles rather than to all papers in the field.

The use of radioisotopes in analysis frequently offers some or all of the following advantages:

1. More sensitive because of the possibility of detecting very low radiation levels.
2. Faster because of the ease of detection of radioisotopes.
3. More easily instrumented and automated, which permits easy feedback for process control.
4. More specific, since radioisotopes of chemically similar elements may have entirely different properties.
5. Often nondestructive because the counting can frequently be done without separation.

These advantages make radioisotopes potentially very useful in process control and often result in analyses being moved out of the laboratory and into the process.

In this issue, several novel uses of the radio-release technique are discussed, and some of the newer developments in neutron activation analysis are surveyed.

Radio-release Techniques

In the radio-release method of analysis a nonradioactive substance is determined indirectly by the quantitative release of a radioactive material from a reagent with which it is treated. For example, ozone oxidizes hydroquinone in which Kr^{85} has been trapped:

$$\left[C_6H_4(OH)_2\right]\ 3Kr^{85}\ (s) + O_3\ (g) \longrightarrow 3C_6H_4O_2\ (s) + 3H_2O\ (\ell)\quad Kr^{85}\ (g)$$

Such "compounds," in which one compound or element is trapped in the crystal lattice of another, are known as "clathrate" or "cage" compounds. The Kr^{85}, which is released quantitatively, is easily measured by standard counting techniques, and from this measurement the ozone can be calculated. Since the analysis technique does not introduce radioactivity into the process, it is particularly attractive for process control.

Because of its relatively long half-life (10.6 y), its chemical and biological inertness, and its moderate-energy beta (0.67 Mev) and absence of gamma energy, Kr^{85} is an ideal isotope for such purposes.

Chleck and coworkers at Tracerlab have developed methods for making Kr^{85} clathrates suitable for use as reagents,[A-4] and, using these clathrates, have developed analyzers for small amounts of ozone[A-6] and sulfur dioxide.[A-4] Workers at Parametrics have developed methods for making radio-release reagents by trapping Kr^{85} in metals by ion bombardment or by high temperature and pressure diffusion.[A-5] Rotariu and coworkers at Booz-Allen applied Kr^{85} clathrates to the analysis of liquids,[A-7] and Richter and Gillespie at Research Triangle Institute have developed a radio-release method for determining small amounts of dissolved oxygen in water.[A-8]

Preparation and Properties of Kr^{85} Clathrates

In 1950 Powell published procedures for making hydroquinone clathrates of argon, krypton, and zenon.[A-9] An almost saturated aqueous solution of hydroquinone was heated to 90°C and then slowly cooled in contact with the inert gas at a pressure of about 40 atm. The product was a stable crystalline substance in which molecules of the rare gas were trapped in the hydroquinone crystal. Many "compounds" of this type can be prepared by similar techniques, as discussed in a recent book.[A-10]

Chleck, Ziegler, and coworkers,[A-4, A-11, A-12] to avoid the added manipulative steps of the solution method when working with the radioactive gas, devised the following procedure for crystallizing the Kr^{85} clathrate from the melt:

About 1 g of hydroquinone is put in a pressure bomb, atmospheric gases are removed, and Kr^{85} is introduced under pressure. The bomb is heated to 185°C to melt the hydroquinone and held at this temperature for 2 hr to allow the Kr^{85} to saturate the melt. The system is then cooled slowly to room temperature, the bomb is opened, and the sample is removed. With pressures up to 60 atm and cooling for as long as 72 hr, the greatest clathrate trapping efficiency* was 28.3%. This would provide 25.8 equivalent atmospheres of Kr^{85} at 15°C. With 20 atm pressure and 17 hr cooling, the efficiency was 17%. At 10 atm pressure and 4 hr cooling, efficiency was poor, but some clathrate formed.

The clathrates thus formed were a fused hard mass, with activities of 30 μc, 15 mc, and 1.5 c per gram, depending on the specific activity of

* The efficiency of a clathrate is defined as the amount of gas trapped compared to the theoretical amount possible.

the original krypton and the efficiency of the process. The leakage rate of Kr^{85} from the strongest sample (1.5 c/g) was about 3 μc/g.day. This rate (10 times that of the 30 mc/g sample on a per unit activity basis, but less than that of the weakest sample) is low enough that these clathrates can be kept in a closed room for weeks without exceeding the maximum allowable concentration. The samples are stable at room temperature and resistant to radiation breakdown. Wilson and Hughes[A-13] at Harwell, using Powell's solution procedure, obtained a clathrate efficiency of 80-90%. However, the compounds were not stable, with 10% of the Kr^{85} being lost in the first month and another 5 to 10% in the next 2 months. They attributed the instability of their compounds to the effect of the beta radiation from the Kr^{85} on the water trapped in the compound.

Mock, Myers, and Trabant at Purdue[A-14] studied crystallization from a number of different solvents, crystallization from melts, and sublimation. They found that crystallization from a solvent gave the best efficiency and from a melt, the poorest. They also tried a number of other crystals, but none was as good as hydroquinone. Nonradioactive samples of these clathrates exposed to 10^8 rads in the Purdue University gamma facility showed no weight change.[A-15]

Kryptonates as Universal Radioactive Indicators

Another type of solid compound of krypton, which should have wide application as a radio-release reagent, has been developed by Chleck and associates at Parametrics, Inc.[A-5] These workers incorporated krypton, containing 5% Kr^{85}, in solid materials both by bombardment with ionized krypton and by diffusion under high temperature and pressure. The ionization procedure is now used only for substances that cannot withstand the pressures or temperatures required in the more efficient diffusion process.

Typical conditions for the diffusion saturation of a substance with krypton are temperatures around 650°C and pressures up to 5000 psi for from several hours to three days. The possible saturation is a function of the structure of the compound, with substances having numerous interstices of the proper dimensions to contain the krypton atom becoming the most highly saturated. Pyrolytic graphite has been saturated to an activity of 1 curie/g, whereas with less favorable structures the activity is frequently limited to millicuries or microcuries per gram. The diffusion method has successfully induced measurable, though often small, permanent krypton activity in all substances tested so far. Therefore, the title of "universal" indicator is used. Uniform incorporation of krypton into a solid is possible if a fine powder is saturated with krypton and then compressed into a solid mass.

The temperature stability of the kryptonates varies. Some lose most of their activity at atmospheric pressure when at temperatures of 75-100°C, while copper kryptonate retains 75% of its activity to 900°C. Generally, the stability at room temperature is excellent.

Clathrate Ozone Analyzer

Using the Kr^{85} clathrate, Hommel, Chleck, and Brousaides at Tracerlab developed a very successful analyzer for ozone in the air.[A-6, A-16]

Analysis of air for ozone has received increasing attention due to acute problems of air pollution and the associated health hazard. It results in eye and nasal irritations, reduced visibility, injury to plant life, and breakdown of rubber articles. An almost explosive interest in the determination of atmospheric ozone in weather studies has also developed, because it appears that knowledge of ozone-meteorological relations will eventually lead to improved weather forecasting. The principal methods used until now for ozone determination are based on

the classical potassium iodide-iodine method, fluorescence, absorption of radiation by ozone, and special methods such as the cracking action of ozone on elongated rubber. Most of the chemical methods are relatively insensitive and nonspecific; for example, the limit of the potassium iodide method is about 1 part in 10^7, and the method is nonspecific. Spectrophotometric techniques require light-path lengths of several hundred feet, and the cracking action of rubber, though highly sensitive, is not specific or continuous. Thus, there has long been a need for a portable, light-weight, rugged, and specific ozone analyzer.

Specificity, sensitivity, and simplicity of design are inherent in the clathrate ozone analyzer system which is based on the reaction given above. The chief limit on the sensitivity is the specific activity of the radioactive salt. In some evaluation tests, when no effort was being made to get maximum sensitivity, 1 part of ozone in 10^{10} parts of air was detected. The specificity is due to the high oxidation potential of the hydroquinone-quinone couple. Only a few oxidizing agents can liberate krypton from the clathrate, and these do not include the airborne contaminants sulfur dioxide and hydrogen sulfide. The clathrate responds to water vapor in the air by dissolution of cage bonds, but the gas can be dried without changing the ozone concentration. Alternatively, the stream can be split and both the ozone and water measured in the first stream, and water in the second (after destruction of ozone by a silver foil catalyst) then ozone determined by difference.

In using the clathrate analyzer, 100 - 200 mg of Kr^{85} clathrate is placed in a 2- to 5-mm-dia capillary (the amount of clathrate and dimensions of the capillary are not critical). The liberated Kr^{85} is passed into a 50-ml chamber containing a Tracerlab TGC-6 Geiger-Mueller tube. Counts are observed on a Tracerlab SC-51 Autoscaler. Associated apparatus includes an automatic graphic recorder and rate meter.

One of the most difficult problems in the development of methods for the determination of traces of gases is the preparation of standard samples for analysis. The Tracerlab workers, after trying several methods for preparing air samples containing small concentrations of ozone, decided on the use of ultraviolet ozone lamps, which are sold for sterilization purposes under such names as "Sterilamp" and "Odorout". These lamps, after a break-in period and under carefully controlled conditions, gave an ozone concentration which varied less than 5% during a given day's operation. By use of the proper flow rate and combination of bulbs, ozone concentrations between 1 and 500 ppm could be obtained and analyzed chemically by the potassium iodide-iodine method.

The response of the clathrate analyzer to changes in ozone concentration was determined by connecting the ozone-air stream to a source of dry nitrogen which could be adjusted to give instantaneous changes in concentration. Ozone was passed over the clathrate and, after steady-state conditions were reached, the ozone concentration was dropped by increasing the proportion of diluent gas. After a new steady-state was reached, the same procedure was again followed for several different, and unmeasured, ozone concentrations. The plateaus on the curve (Fig. A-1) indicate regions of different ozone concentration and the arrows the points of dilution.

The 1 min between ozone dilution and counting response is about the same as the time lag due to dead volume. Thus, the clathrate reaction very nearly follows the change in ozone concentrations, the response being for all practical purposes immediate. No complete explanation has been given for the rise in the curve immediately after a change in concentration; however, the surge may result from a stripping action of the gas stream, caused by flow rate adjustments, on loosely sorbed krypton molecules on the surfaces of the clathrate.

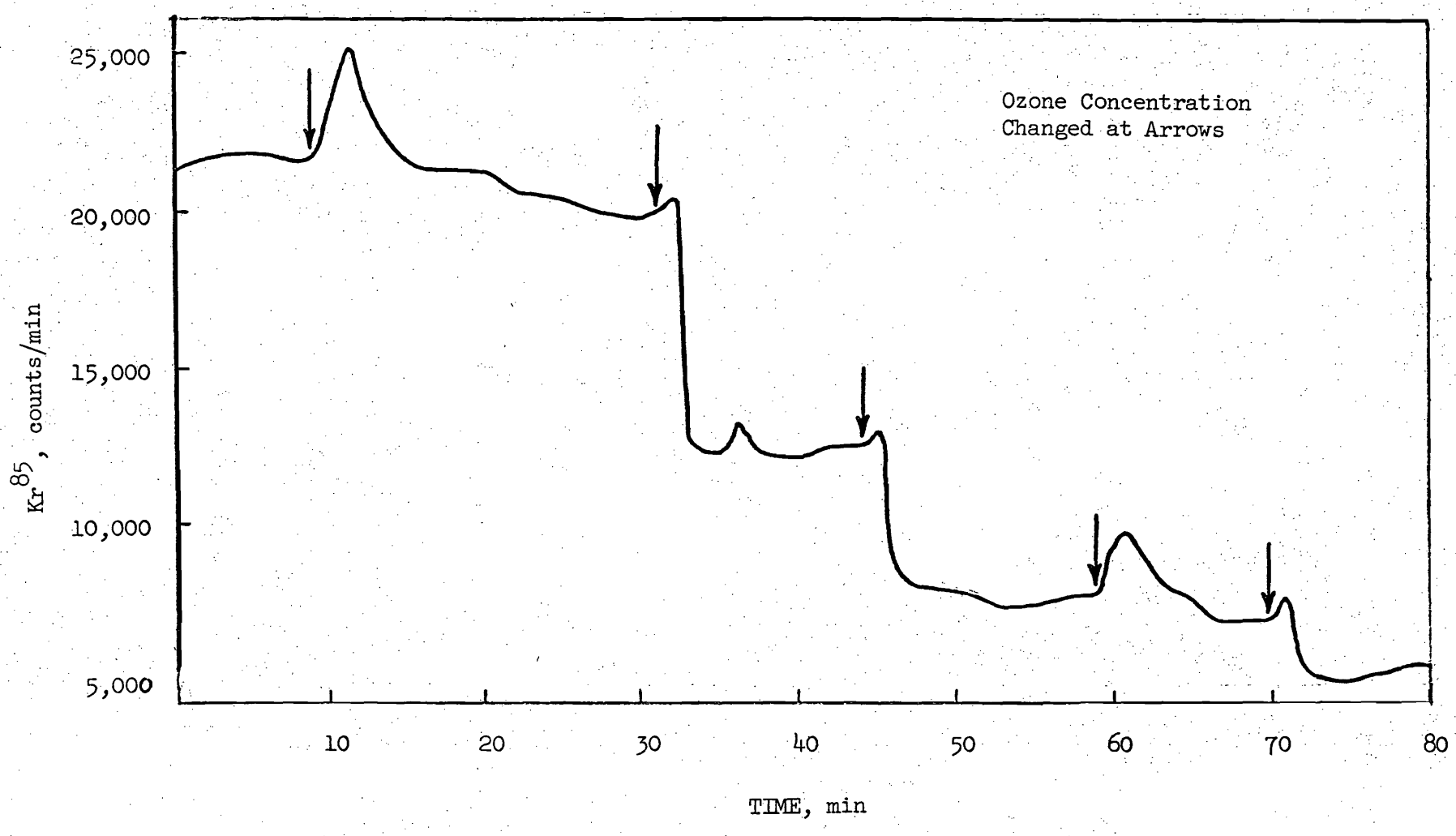

Fig. A-1 RESPONSE OF OZONE ANALYZER TO CHANGES IN CONCENTRATION

To study the variables in the ozone-clathrate reaction, a clathrate cell was installed in a 2-liter bell jar fitted with a manometer (Fig. A-2). A pump of the type used by the weather bureau in sonde balloons circulated the gas through the clathrate cell and into the detector. For static tests, the chamber is pumped to a high vacuum and ozonized air is admitted. After the system is closed, a pump circulates the gas through the clathrate and detector and returns the gas into the chamber. Although deozonized air and Kr^{85} are returned to the chamber, the interval for data gathering is small and the chamber volume is large enough to maintain an almost constant concentration during the measurement. In dynamic measurements, the observation period is unlimited since a fresh stream of the air and ozone is constantly fed to the chamber. Flow is maintained with a vacuum pump, and desired pressures are controlled by valves located at feed and exit ports.

The static system was used to test the response of the clathrate cell to a wide range of ozone concentrations at atmospheric conditions. The chamber was filled with 13 ppm of ozone at 1 atm and the counting rate of the Kr^{85} was taken. The chamber was then pumped to 76 mm Hg and repressured with dry nitrogen to 760 mm Hg, where the counting rate was again measured. Thus a given concentration of ozone was successively diminished by increments of 0.9, and an ozone concentration range of five orders of magnitude, from 0.1 ppb to 10 ppm, was obtained (Fig. A-3).

Using the dynamic system, with a constant ozone concentration in the chamber, the Kr^{85} count rate was studied as a function of pressure (Fig. A-4). No attempt was made to select or determine an ozone concentration. The important point was to maintain a constant concentration. The reproducibility of the reaction is shown by the nature of the slope.

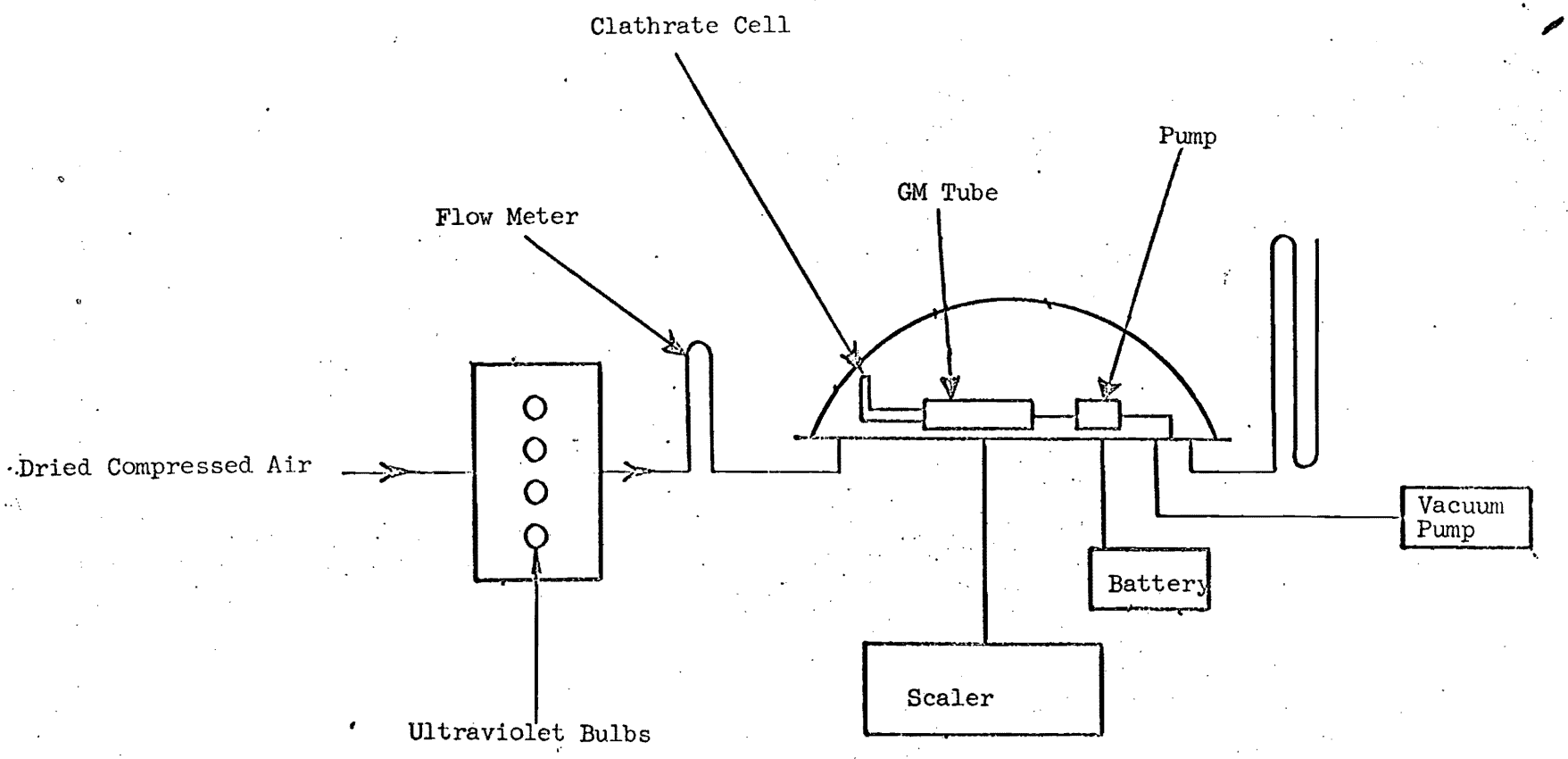

Fig. A-2 APPARATUS FOR ENVIRONMENTAL OZONE STUDIES

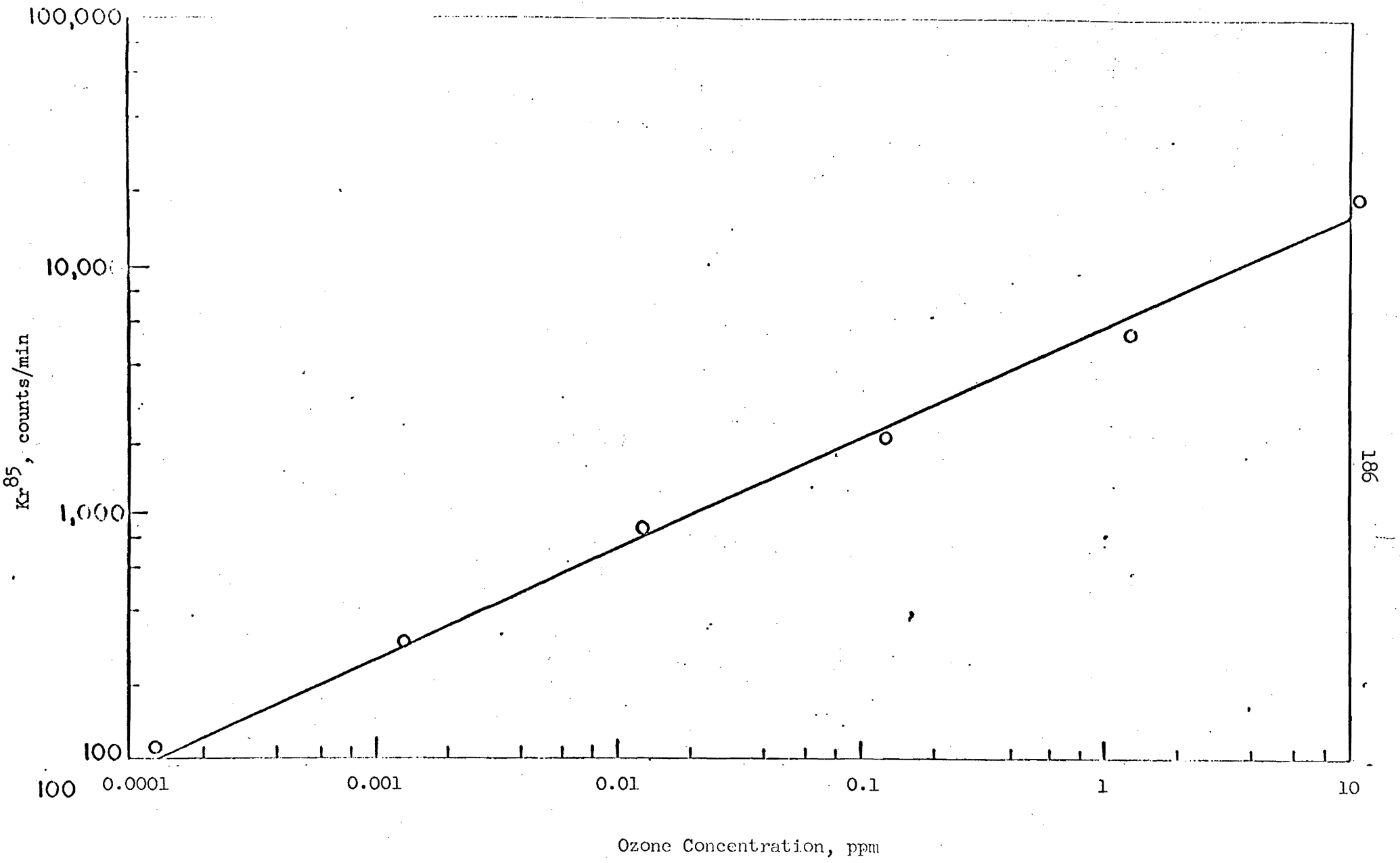

Fig. A-3 CLATHRATE ANALYZER RESPONSE TO OZONE

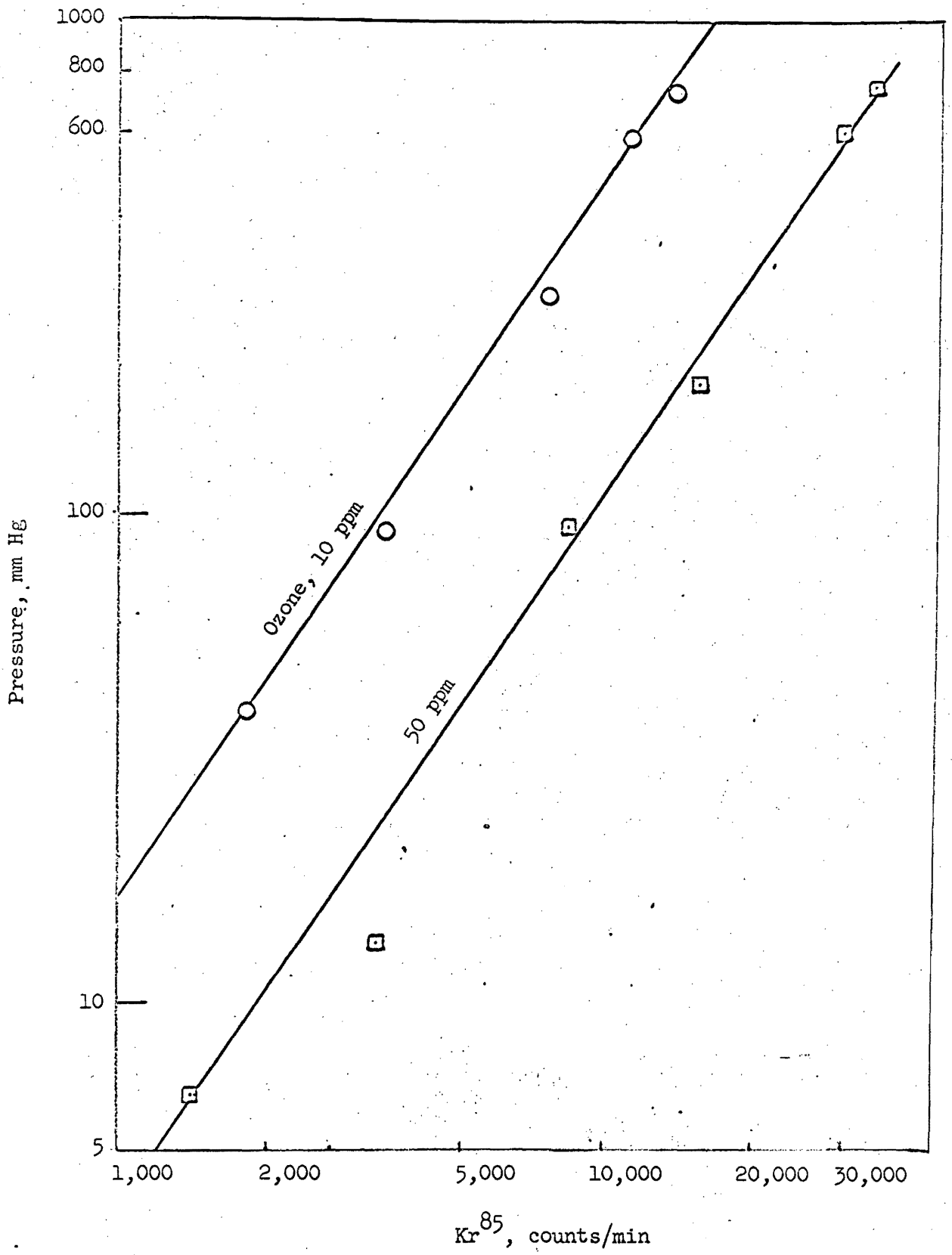

Fig. A-4 PRESSURE DEPENDENCE OF OZONE-CLATHRATE REACTION

An examination of Fig. A-3 and A-4 reveals interesting relations between count rate and ozone concentration. In Fig. A-3, a direct plot of ozone concentration against count rate, a 10-fold decrease in concentration caused a decrease in count rate of only a factor of 2.9. For Fig. A-4, which is a plot of pressure against count rate, the factor decreases by 4.8.

It may at first seem surprising that the reaction efficiency should increase with a reduction of pressure or ozone concentration. However, the reaction does not involve krypton directly and indicates only the breaking of bonds to release the gas. The rupture of any cage bond will release a krypton molecule while the rupture of two bonds on the same cage will still only release one molecule. Thus, if the ozone density about the clathrate is reduced, either by a pressure or concentration reduction, the probability that two or more ozone molecules will inefficiently effect the release of the same gas molecule is diminished.

Following these and other preliminary tests, the analyzer was tested in balloon flights. Two flights were made using conventional meteorological equipment. In the first flight, data were obtained on the temperature profile and radiation background between sea level and 75,000 ft. This information was needed to aid in interpretation of data from the ozone analyzer since the ozone-clathrate reaction is temperature sensitive and the final measurement of the radioactivity of the evolved Kr^{85} requires a knowledge of the background.

The test of the analyzer in the second flight went quite smoothly up to 75,000 ft. Although the signal-to-background ratio was bad, and the ozone concentrations were lower than those obtained by other methods, the data profile, shown in Fig. A-5, was obtained.

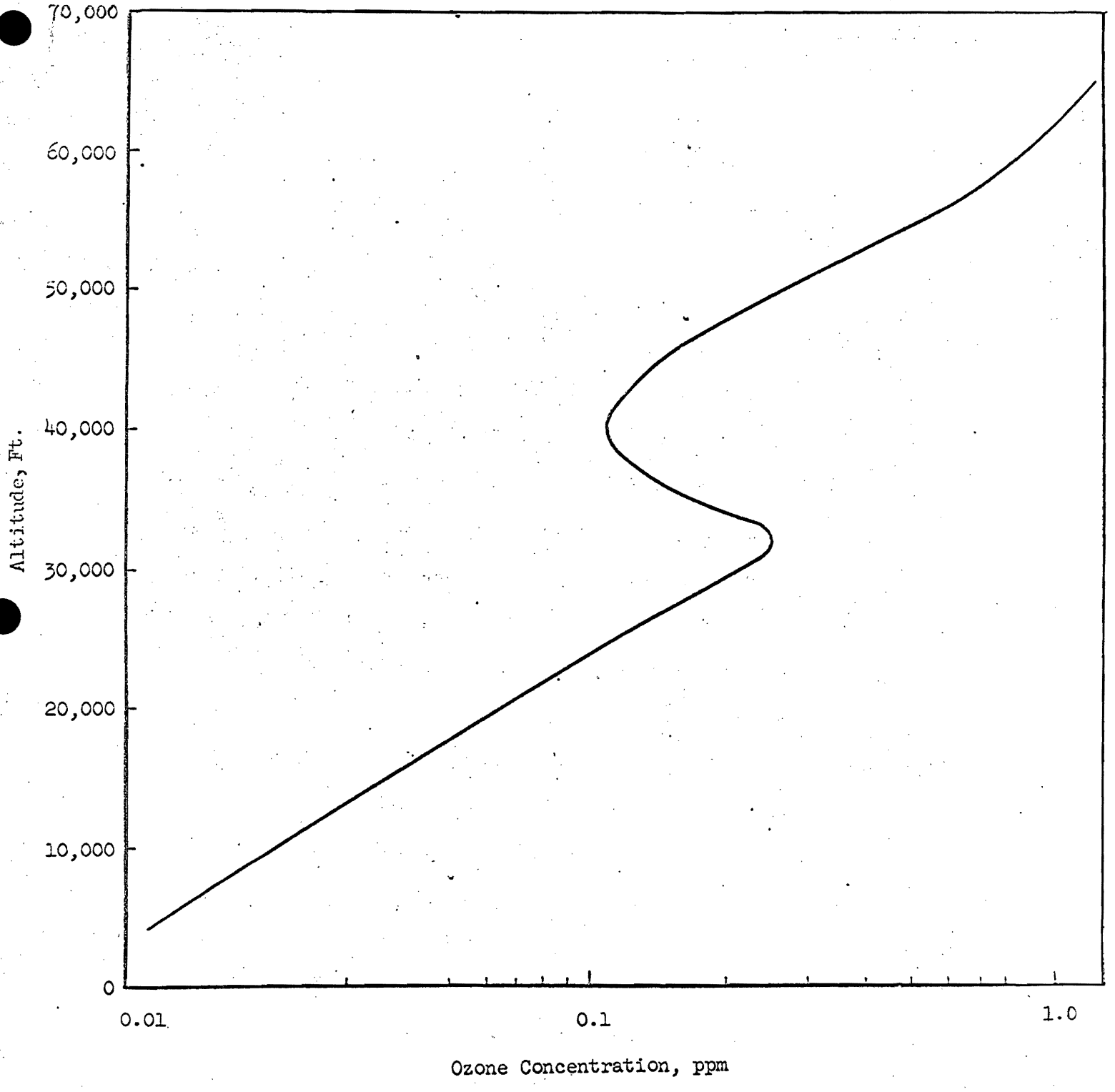

Fig. A-5 VERTICAL PROFILE OF ATMOSPHERIC OZONE

Sulfur Dioxide Analyzer

Workers at Tracerlab[A-4, A-17, A-18] have also developed an effective analyzer to determine traces of sulfur dioxide in air using the Kr^{85}-clathrate. Sulfur dioxide will not oxidize hydroquinone; however, it will oxidize sodium chlorite, releasing chlorine dioxide:

$$SO_2 + 2NaClO_2 \rightarrow 2ClO_2 + Na_2SO_2$$

(This equation is an oversimplification since experimentally it was found that one molecule of sulfur dioxide released between 4 and 8 molecules of chlorine dioxide). The chlorine dioxide will then oxidize hydroquinone-clathrate and release Kr^{85}. An instrument for the determination of sulfur dioxide was designed using these two reactions in series. In the original tests of the instrument, the response to varying concentrations of sulfur dioxide was linear; however, the slope and position of the entire line were displaced depending on the relative humidity of the air (Fig. A-6).

Bersin, Brousaides, and Hommel[A-18] carried out a very extensive investigation of this method of analysis at Tracerlab under the joint sponsorship of the Los Angeles Air Pollution Foundation and the Division of Isotope Development, USAEC. A number of variables in the operation of the instrument, such as the size of the reaction tubes, quantity of sodium chlorite and clathrate in the tubes, flow of gas, and humidity were studied. They showed that it was possible to pick conditions for the analyzer in such a way that for the lower sulfur dioxide concentrations the counting rate did not depend on relative humidity (Fig. A-7).

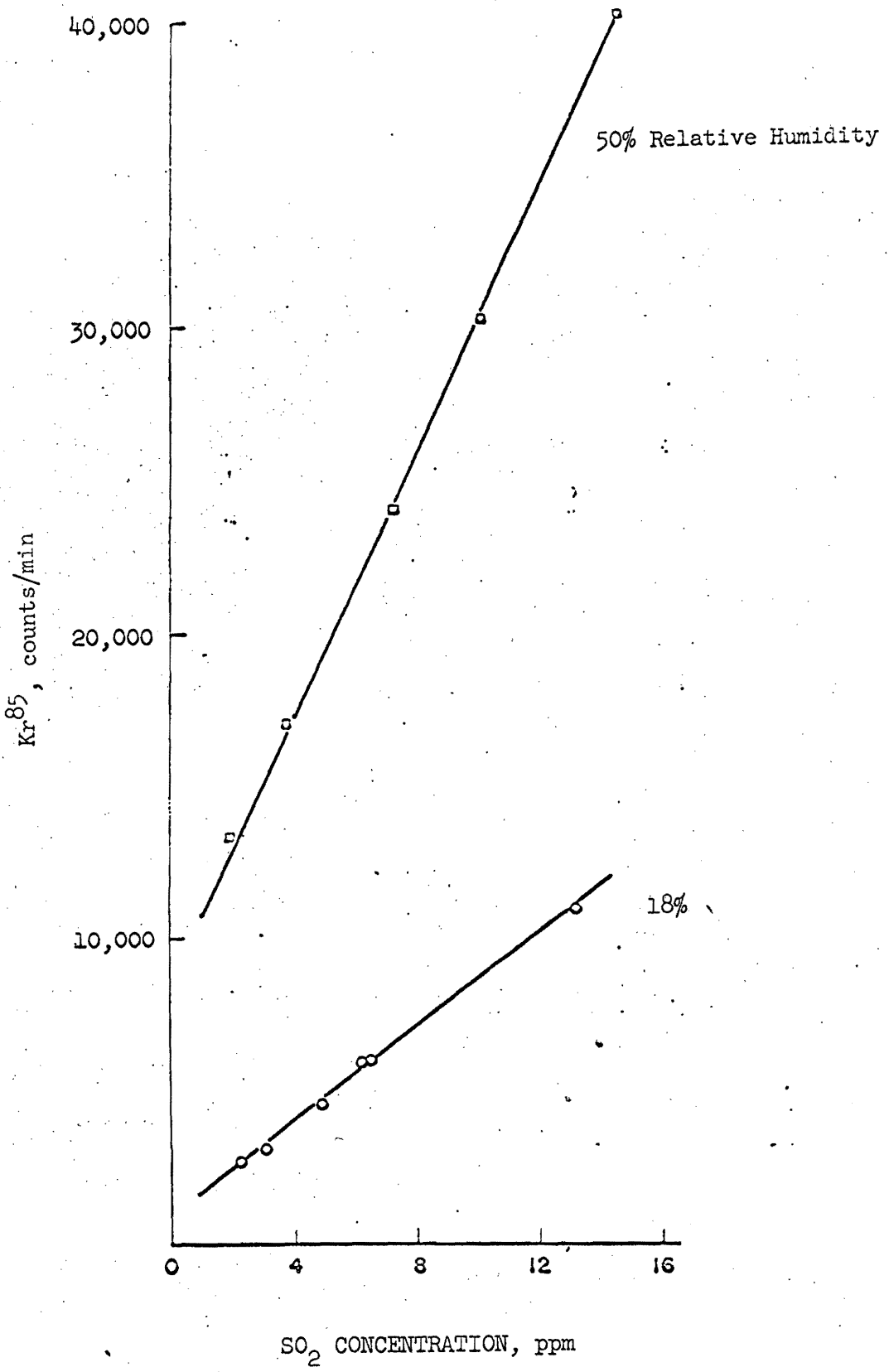

Fig. A-6 EFFECT OF HUMIDITY ON COUNTING RATE

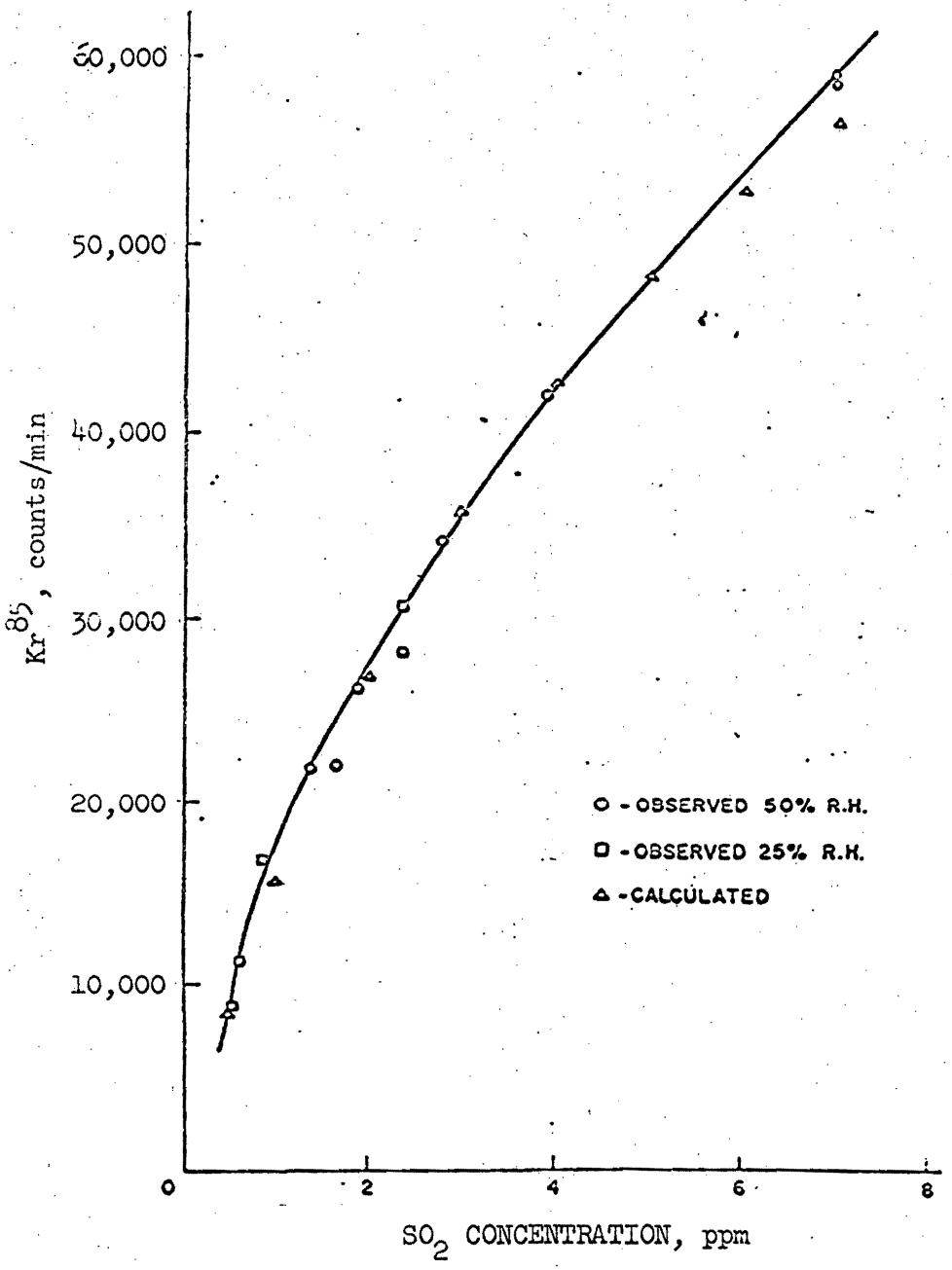

Fig. A-7 CALIBRATION CURVE FOR SULFUR DIOXIDE AT TWO RELATIVE HUMIDITIES

In an 8-day test to determine the stability in continuous operation, the data shown in Fig. A-8 were obtained. The only gases that should interfere with the operation of this analyzer are those that can oxidize either sodium chlorite or the hydroquinone in the Kr^{85} clathrate. The oxides of nitrogen and ozone are the only commonly occurring atmospheric gases which should be any trouble.

On the basis of these tests, an instrument for use in field monitoring was designed. A schematic drawing of this instrument is shown in Fig. A-9.

Kr^{85}-Clathrates for the Analysis of Solutions

Rotariu, Hoskins, and Hattori at Booz-Allen Applied Research[A-7] developed a technique for determining iron by the release of Kr^{85} from the clathrate. It was hoped that by this technique iron could be determined in the parts per billion range. The study was discontinued before this goal had been achieved; however, it was shown that it was possible to make determinations in the 2 to 20 parts per million range.

In this method, ferric ion is extracted into trifluorotoluene with thenoyltrifluoroacetone (TTA) as the chelating agent at a pH of 2 to 3. The organic ferric solution is then transferred to a closed system, gaseous hydrogen chloride is passed over it for 8 sec, and excess gas is removed with nitrogen. A weighed 1-mg portion of Kr^{85}-clathrate is then added to the solution and it is stirred for 15 min. The reactants are frozen in Dry Ice-acetone, and the Kr^{85} is pumped into an ionization chamber and counted.

Although this method will determine 2-20 ppm of iron in the trifluorotoluene-TTA solution, the results of studies on the distribution coefficient of ferric ion between water and this organic phase indicate

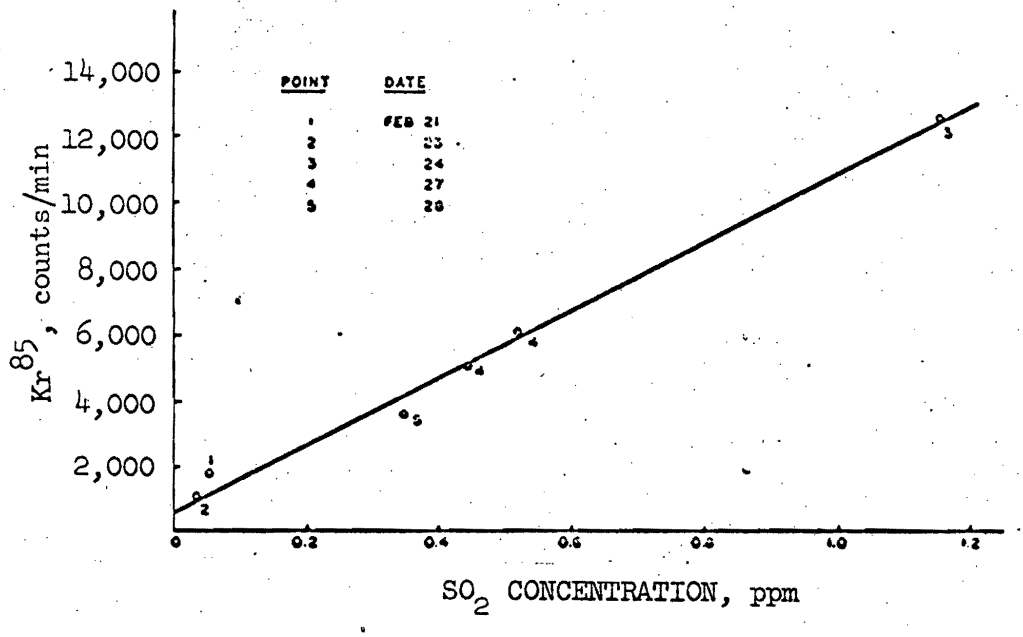

Fig. A-8 STABILITY OF ANALYZER OVER AN EIGHT-DAY MEASURING PERIOD

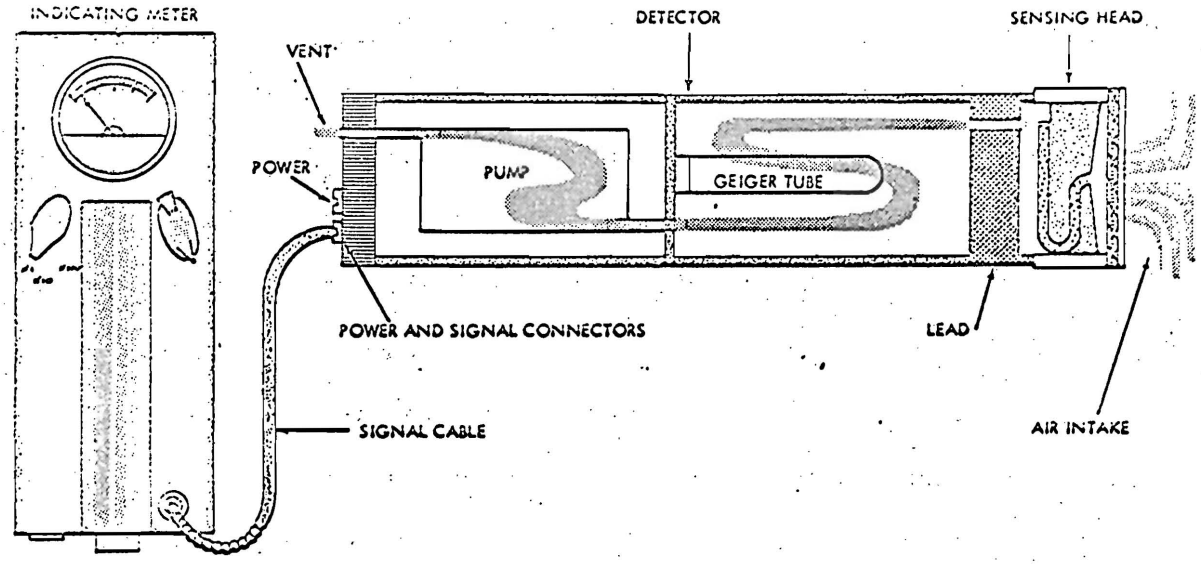

Fig. A-9 COMMERCIAL ISOTOPE AIR POLLUTION MONITOR

that 3 to 4 extractions would be needed to approach quantitative extraction. Also, no results of tests on the determination of iron in aqueous solution using the entire procedure are given; therefore, it is impossible to judge the overall precision and accuracy possible with such a method.

Determination of Active Hydrogen with Lithium Aluminum Tritide

Krynitsky, Johnson, and Carhart[A-19] in 1948 published a method for determining active hydrogen in such compounds as water, alcohols, and acids by reaction with lithium aluminum hydride at 0°C in an ether solution, and measurement of the pressure change in the system to determine the hydrogen released. Workers at Tracerlab[A-3, A-4] substituted lithium aluminum tritide for the hydride and determined the tritium released by counting with a proportional counter. In the apparatus used, (Fig. A-10) the 25-ml reaction cell, 6.5 in. long and 20 mm dia, was fitted with a graduated pipette for sample addition. The reaction mixture, 15 ml of approximately 0.2 M lithium aluminum tritide in diethyl carbitol, was introduced into the reaction cell. The pinch clamp was closed, and counter gas was bubbled through the cell at a flow rate of 40 cc/min to obtain the background count. The sample diluted in tetrahydrofuran was introduced, and the tritium counted for a 10-min period. Fine adjustment of counter gas pressure was made with a screw clamp between the reaction cell and the flow meter. A bypass line was provided around the reaction cell to allow for the adjustment of flows while reagents were in the reaction cell. The tritium detection equipment consisted of a Tracerlab Pulse Pre-Amplifier, Amplifier, and "1000" scaler; the counter gas was Tracerlab Proportional Counting Gas (90% argon, 10% methane).

The value of method is increased because it is not necessary to prepare a solution of a known specific activity of lithium aluminum tritide in diethyl carbitol. However, the specific activity must be high enough to yield adequate counting rates.

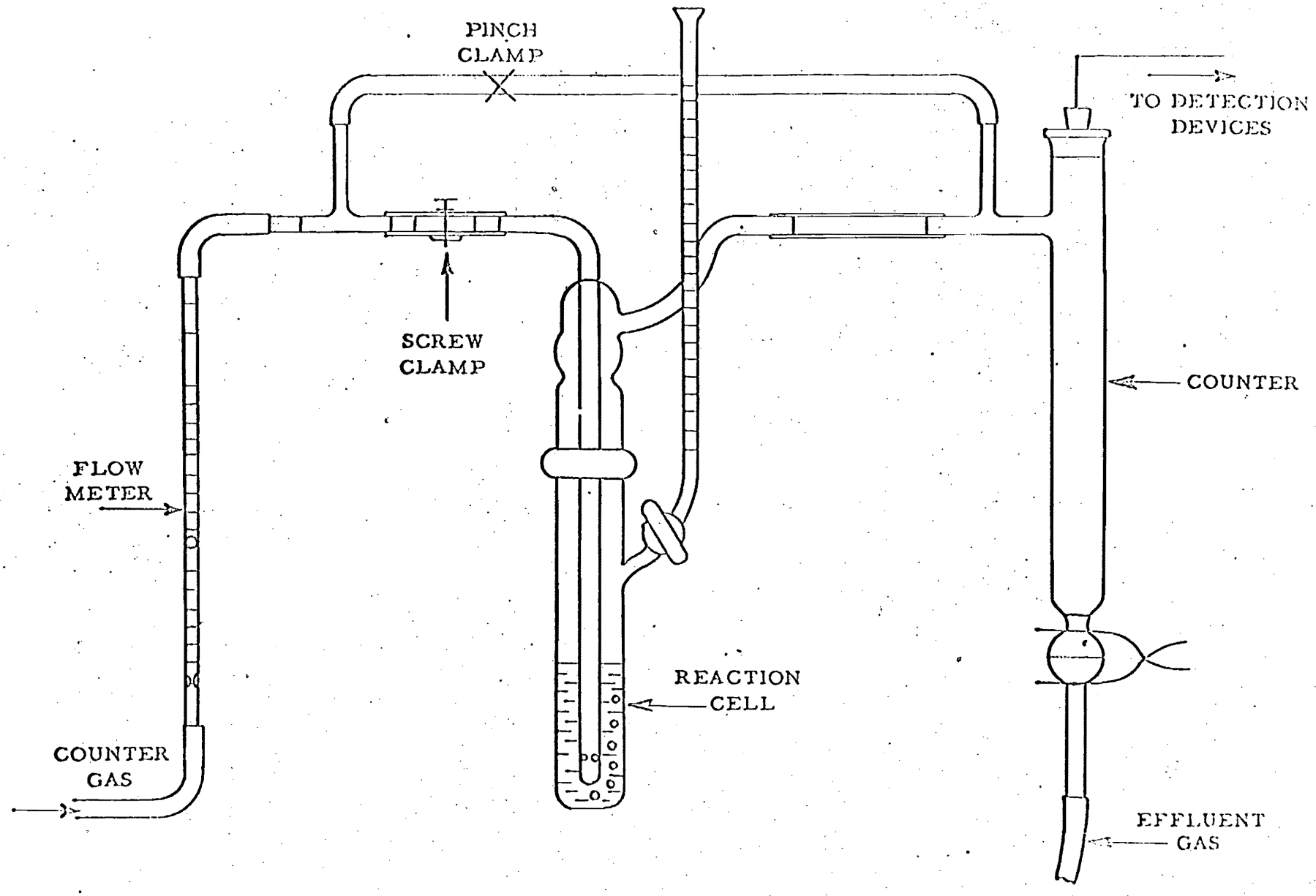

Fig. A-10 APPARATUS FOR ACTIVE HYDROGEN DETERMINATION

Normal butanol, benzoic acid, aniline, and water containing active hydrogen were analyzed, different specific activities being used for each. In each case, the plot of the amount of compound against counting rate of tritium was linear. Figure A-11 is a typical plot of the data obtained with water. Since the stoichiometry of the reaction had been studied by Krynitsky et al., no attempt was made to correlate data between compounds. Any of the four curves could be used to determine the active hydrogen content of an unknown by using the same specific activity of lithium aluminum tritide; or if the number of active hydrogens in the compound is known, the compound could be determined quantitatively. All the plots extrapolated to a point below the origin, probably due to fixed losses of hydrogen in the system. This error could be reduced with better cell design.

The sensitivity of the method is very high. The lithium aluminum tritide used in this study had a specific activity of 2.5 mc/mg, but was diluted with lithium aluminum hydride by a factor of 10^5. The ultimate theoretical sensitivity with lithium aluminum tritide without dilution would be of the order of 10^{-8} memole of active hydrogen compound. For practical purposes the sensitivity attainable is limited only by the ingenuity of the analyst in devising more efficient techniques for introducing small sample volumes and a means for sample protection from air moisture. Fifty or more samples may be analyzed on one filling of the reaction cell. In practice the cell should be refilled after half the available tritide has been consumed.

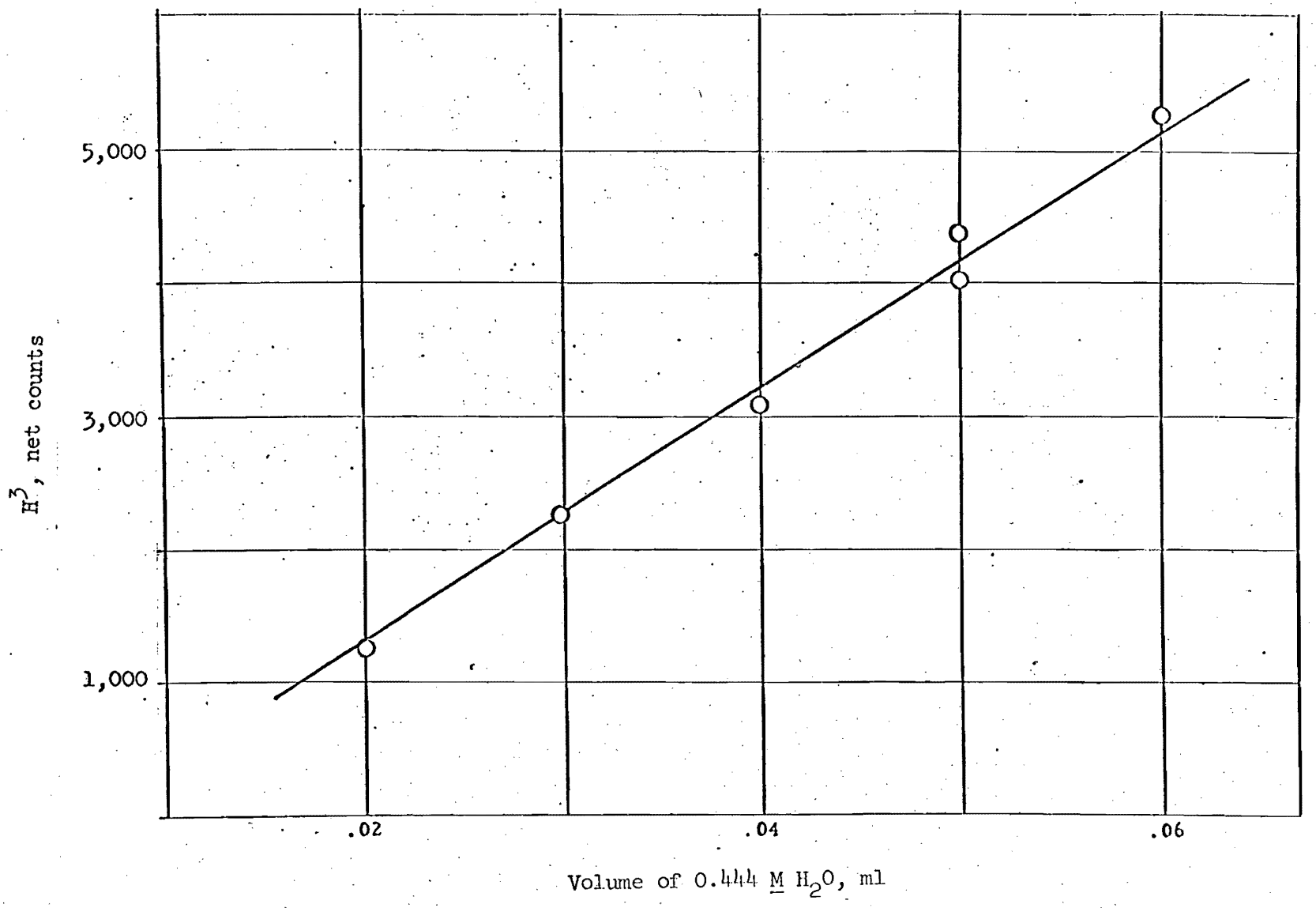

Fig. A-11 DETERMINATION OF H_2O IN TETRAHYDROFURAN

Following these successful experiments, the workers at Tracerlab attempted to change the method from a liquid to a gas-solid reaction.[A-6] It was hoped that the gas-solid reaction could be made the basis of a detector for water vapor similar to that described above for ozone and sulfur dioxide. The experiments were not successful on the microscale because the solid reaction products formed a film over the lithium aluminum tritide and prevented further reaction, but, on a semimicroscale, the reaction proceeded more smoothly and reproducibly. Because of this film formation, however, work with lithium aluminum tritide was discontinued and exploratory experiments with other reactions were started.

Determination of Dissolved Oxygen in Water

A new analytical technique for determination of dissolved oxygen in water was recently described by Richter and Gillespie of the Research Triangle Institute.[A-8] The method is based on the rapid and quantitative oxidation of thallium metal by oxygen dissolved in water, even in extremely low concentrations. This fact, discovered by Wright and Lindsay,[A-20] had been used in an industrial instrument in which the water is passed over thallium and the electrical conductivity of the effluent stream is measured. By substituting Tl^{204} and counting the effluent stream, Richter and Gillespie determined 0.2 ppm of oxygen in pure water and used the method for water containing many more impurities than in the original conductivity method. The authors believe that this method is potentially more sensitivity than any other known method for this determination.

The analyzer is a 10-mm dia glass column 20 cm long filled with radioactive thallium metal electrodeposited on copper turnings and a flow-type Geiger-Mueller tube. The sample is introduced at the top of the column and flows slowly through it. Tl^{204} ions released by the reaction of dissolved oxygen with the thallium enter immediately into the G-M tube and are detected. The Tl^{204} counting rate is a direct measure of the oxygen concentration.

For oxygen concentrations in the parts-per-million range, the radioactive thallium (4480 mc/g of Tl) was diluted with inactive thallium metal so that the final activity was 2.04 mc/g Tl. The Tl^{204} has a long enough half-life (3.6 years) that decay over several months does not greatly reduce the sensitivity of the technique. Also, since Tl^{204} is a pure beta-emitter, the glass walls of the column provide sufficient shielding that no radiation hazard exists. It is estimated that the analyzer, excluding scaling or recording equipment, can be built for about $100 in equipment and isotope costs.

Waters of known oxygen concentration were prepared by equilibrating a water sample with nitrogen and oxygen mixtures. Flow rates of the two gases were measured, and the gases were mixed and then bubbled through water for several hours. The sample so prepared was passed slowly through the thallium column (1 to 3 ml/min). known volume of water was removed from the reservoir for analysis by the well known Winkler method.

Response of the oxygen analyzer to waters of markedly different salt contents with varying oxygen concentrations is shown in Fig. A-12;

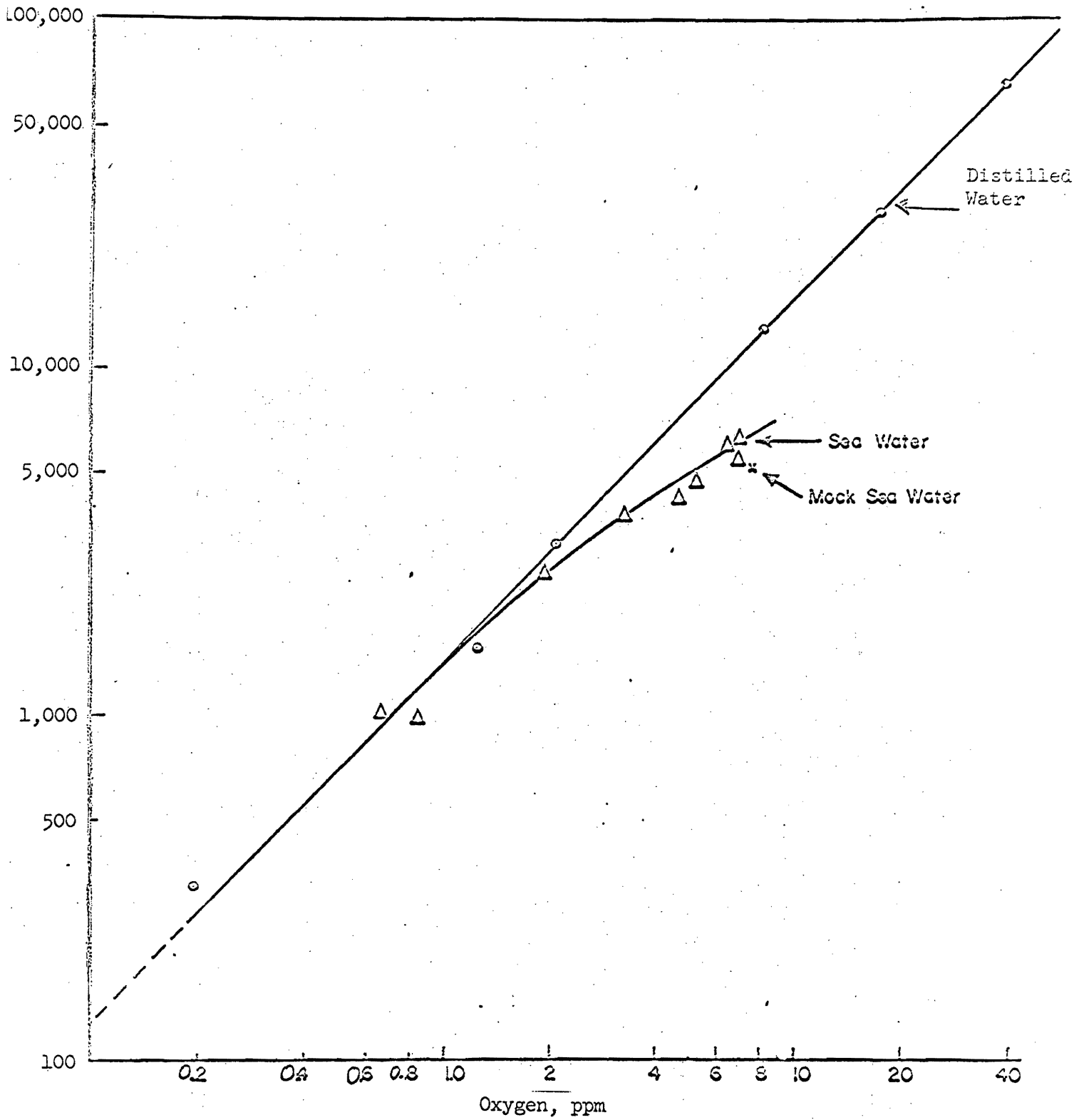

Fig. A-12 RESPONSE OF Tl^{204} ANALYZER TO DISSOLVED OXYGEN IN DISTILLED WATER AND SEA WATER

the count rate is corrected for dead time losses and for backgrounds and is plotted as a function of oxygen concentration. The linearity for oxygen in distilled water is excellent, but departure from linearity was noted for sea water and mock sea water containing only sodium chloride.

Thallium determinations on the effluent solutions indicate that oxidation of thallium is stoichiometric:

$$4Tl(s) + O_2(aq) + 2H_2O(liq.) \rightarrow 4Tl^+(aq) + 4OH^-(aq)$$

One gram of oxygen liberates 25.6 g of thallium; therefore, 1 g of thallium is sufficient to assay approximately 5 liters of water saturated with air. The column retains its calibration from beginning to end, and the thallium is completely inert to oxygen-free water.

The sea water used in this study contained 17.7 g of chloride per liter. Thallium analysis of the effluent solution from the column indicated that count rate corresponded to liberated thallium, but this amount did not agree with the expected quantity from the influent oxygen solution except at low oxygen concentrations. Mock sea water consisting of 29.6 g of pure sodium chloride per liter of distilled water gave results similar to those for sea water. Sodium chloride thus appears to cause the departure from linearity.

The results of several experiments performed in an attempt to understand the nonlinearity in sea water solutions indicated that interaction of thallium, copper, and oxygen was causing the

loss of thallium. Columns prepared by depositing thallium on a metal less active than copper should give satisfactory linearity. Although linear response by an instrument is desirable, nonlinear action does not preclude its usefulness in any applications. Since the thallium release in sea water is reproducible, if not linear, this technique can be a good analytical tool once the column has been calibrated.

Studies on the effect of the solution pH indicate that the method is suitable for solutions with pH values between 5 and 10 and that acidic solutions can be analyzed if the pH is brought into this range with a suitable buffer. Nitrate salts interfered, but chromates did not at pH 8.

The sensitivity of the column with a specific activity of 2.04 mc/g Tl is about 0.2 ppm, which produces a Tl^{204} counting rate equal to the background counting rate of the detector. If the 4478 mc/g material obtained from ORNL for the study had been used without dilution, the sensitivity of the column would have been 0.1 ppb. (The ORNL Isotopes Catalog lists Tl^{204} with a minimum specific activity of 1000 mc/g Tl.) Oxygen concentrations in this range were not investigated because of time limitations and the difficulty of obtaining solutions of known parts per billion oxygen concentrations. Sensitivity could be increased by at least a factor of 10 by using a larger counting tube. Still another factor of 10 in sensitivity should be attainable by a reduction in the background counting rate, which was about 100 cpm. Thus, a sensitivity of about 1 part of oxygen per trillion parts of water should be practical. This

speculation is based on the assumptions that metallic thallium is totally insoluble in oxygen-free water, and that radiolytic decomposition of water would produce no more oxygen than this.

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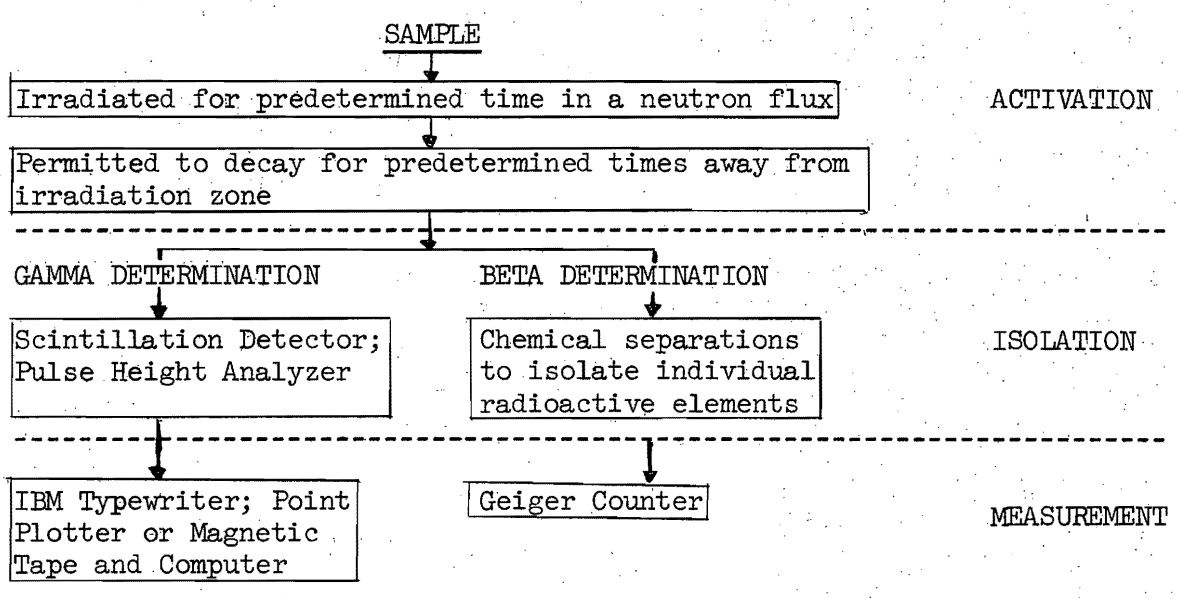
Activation Analysis

The basic principles of activation analysis are very simple. A sample of material is bombarded by some type of particle, usually neutrons, which produces radioactivity in either the matrix material, the impurities, or both; the activity may or may not be isolated; and the type and amount of the activity are determined. An outline of the general steps in activation analysis was given recently by Aebersold:[B-3]

STEPS IN NEUTRON ACTIVATION ANALYSIS



Neutron activation analysis was originated in 1936 by Hevesy and Levi,[B-1] who exposed samples of rare earths to a radium-beryllium neutron source to detect and determine impurities. The method, however, did not gain wide acceptance until after 1946 when nuclear reactors became available to provide high-flux sources of neutrons.[B-2] Since then, many analytical methods using neutron activation have been developed, but the majority of these are suitable only for laboratory use since they

require extensive radiochemical separation and take too much time for consideration in process control. Neutron activation is now moving out of the research laboratory and into broad applications throughout science and industry, mainly because of the development of new techniques and methods.

Meinke and Schideler[B-8] have suggested that activation analysis will come into general use only when neutron sources suitable for use in an individual laboratory are available and when it is possible to complete an analysis in a matter of minutes (or at the most an hour) instead of days. Current research in this field is directed toward solution of both these problems.

Although there have been many papers published on activation analysis, no textbook or monograph has appeared; however, several such books are said to be in preparation. Source Material in Radiochemistry[B-4] gives a reasonably good bibliography of the most important papers in the field up to 1960. Gibbons has published two bibliographies,[B-5, B-6] and Koch has compiled a handbook.[B-7]

The USAEC is continuing its strong interest in activation analysis by supporting a broad range of research and development projects, including preparation and evaluation of isotopic neutron sources; testing of small accelerators for production of neutrons; activation with fast neutrons; development of rapid radiochemical techniques for isolation of radioisotopes; use of multichannel gamma-ray spectrometers for determination of activities; application of instrumentation and computer techniques to provide rapid, automatic, and routine analyses of very large numbers of samples; and use of these techniques in such diverse applications as

analysis of the ocean bottom, the surface of the moon, and samples of interest in criminal investigations. Some of these newer developments are reviewed here.

Neutron Sources

Olive, Cameron, and Clayton of the UKAEA Isotope Research Group recently reviewed high-intensity neutron sources and their applications in industry.[B-9] Nuclear reactors are not considered, but radioisotope sources and accelerating machines are reviewed in detail.

Isotopic Neutron Sources. The UKAEA report concludes that radioisotope sources such as those shown in Table B-1 will usually be limited to a neutron flux of 10^6 to 10^7 n/sec. (In this section, neutron fluxes stated without an area term are based on a 4π geometry.) If higher fluxes are needed, small accelerators or reactors will probably be more economical.

Hennelly[B-10] published information on a 1000-curie Sb^{124}-Be source that has a neutron flux of 10^{10} n/sec. This source has a half-life of only 60 days, but it can be regenerated easily in a reactor. Hennelly suggests that two sources be prepared with one in the reactor being reactivated while the other is in use. (The Sb^{124}-Be source is the only one shown in Table B-1 which can be reactivated.) He estimates that sources could be operated for $5000 per year by this method. The British review[B-9] points out that the worst drawback from this type of source is its intense associated gamma-ray emission, about 10 r/hr at 1 ft from 1 curie.

Strain and Leddicotte[B-11] recently published a description of alpha, gamma, and neutron sources made from Am^{241}. They prepared Am^{241}-Be sources that had a total neutron emission of 4.5×10^6 n/sec, resulting in a flux

Table B-1 RADIOISOTOPE NEUTRON SOURCES

Source	Half-Life	Yield, n/sec. curie
Pb^{210}-Be(α,n)	22 y	2.3×10^6
Ac^{227}-Be(α,n)	22 y	1.5×10^7
Ra^{226}-Be(α,n)	1622 y	1.3×10^7
Po^{210}-Be(α,n)	138 d	2.5×10^6
Th^{228}-Be(α,n)	1.9 y	2.0×10^7
Sb^{124}-Be(γ,n)	60 d	1.0×10^6

of 2.36 x 10^4 n/cm.sec. Although this flux is low, they believe it would be suitable for determination of macro concentrations of a large number of materials. The Am^{241} was >99% pure radiochemically, containing ~0.1 wt % Pu. However, the 16 wt % stable contaminants (9% rare earths, 3% iron, 2% chromium, and 2% sodium-nickel-copper) had to be removed to provide maximum specific activity of the sources. This was done by a modification of a Dowex-1 resin ion exchange process, with elution by 5 M NH_4SCN.[B-12] The americium was extracted from the eluate into di-(2-ethylhexyl)phosphoric acid (D2EHPA), with decontamination factors of at least 10^5 from the rare earths, plutonium, and sodium and Am^{241} recovery ~90%. The 10% loss occurred in the ion exchange column separation because Am^{241} eluted only very slowly with any eluting agent. When the same column was used for processing additional amounts of americium, the retained americium was slowly eluted, at the rate of about 100 μg per liter of 5 M NH_4SCN.

Two types of neutron sources were produced, Am-B and Am-Be. The Am^{241}-Be sources were prepared by thoroughly mixing $Am(NO_3)_3$ in weak HNO_3 (pH 2) with 325 mesh beryllium powder and evaporating the mixture to dryness. The dry product was fired at 500°C to expel NH_4NO_3 and convert the americium to AmO_2. The Am^{241}-B sources were prepared similarly, except that the fired AmO_2-B powder was combined with 0.050 g of paraffin dissolved in CCl_4 and allowed to dry. The mixture was ground and pressed to a 0.72- by 0.73-in. right cylinder under a static load of 10 tons, and the cylinder heated to 500°C to remove the paraffin binder.

Both pellets were sealed in containers (Fig. B-1) in several stages. The pellet was first placed in the nickel, or inner, container and heated

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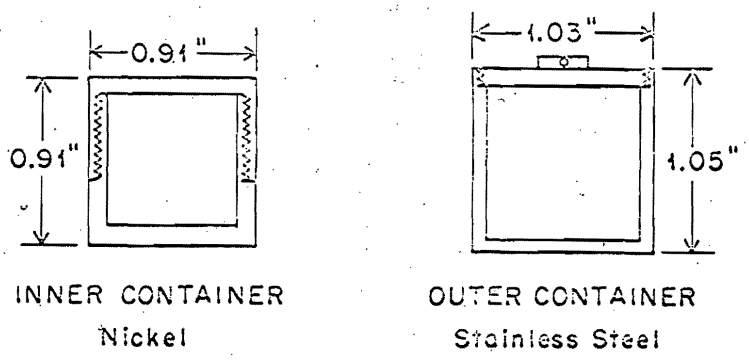

Fig. B-1 Am^{241}-Be Neutron Source Container

to 275°C on a hot plate. With ruby flux and standard solder, a uniform film or solder was placed on the threads of the cap of the nickel container, and the cap was then screwed onto the body of the container. After cooling, the sealed container was cleaned with 1 N HNO_3, dried, and checked for alpha contamination on its surface. When a smear technique removed less than 75 α counts/min from the outer surface, the source was placed in the outer stainless steel container, its lid screwed on, and the container sealed by Heli-arc welding.

Neutron activation analysis has been carried out with an Am^{241}-Be source having a total neutron emission of 4.54×10^6 n/sec. When enclosed in a paraffin moderator, the highest neutron flux attainable is 2.36×10^4 $n/cm^2 \cdot sec$ as measured by Au, Mn, and Co foil activation. The sensitivities of a number of elements have been tabulated with a series of pure oxides, nitrates and carbonates, for various irradiation times using a 3- by 3-in. NaI detector coupled to a 20-channel pulse height analyzer for discriminatory counting. Table B-2 gives a partial listing of these elements. While these neutron sources are not sufficiently intense to allow trace analysis for more than a few isolated elements, they offer a rapid and specific analysis method for macroelement concentrations in a wide variety of materials.

Accelerator Neutron Sources. The use of accelerators for production of neutrons was recently reviewed in Nucleonics.[B-13] Currently the chief interest in neutron sources for activation analysis seems to be in the small-particle accelerators that generate neutrons by the $H^3(d,n)He^4$ reaction. In the British review mentioned above,[B-9] data from the Nucleonics article on such machines, which cost about $20,000 (Table B-3), are summarized.

Table B-2. SENSITIVITIES OF A FEW ELEMENTS FOR NEUTRON ACTIVATION WITH AN Am^{241}-Be NEUTRON SOURCE

Thermal Flux of 2.4×10^4 n/cm^2.sec

Element	Isotope Produced	t 1/2	Gamma Energy, Mev	Irradiation Time	Energy, Counting Mev	Activity, c/min.mg
Hf	189 m	19 s	0.161	1 m	0.02 -- 0.21	2
Sc	46	19.5 s	0.14	1 m	0.02 -- 0.21	3
W	183	5.5 s	0.105	1 m	0.02 -- 0.21	0.01
Al	28	2.3 m	1.78	5 m	1.6 -- 1.88	0.04
I	128	25 m	0.455	30 m	0.72 -- 1.48	0.08
Ag	108	2.3 m	0.44, 0.60	5 m	0.36 -- 0.74	0.2
Mg	27	9.4 m	0.84 1.02	10 m	0.72 -- 1.48	0.002
Mn	57	2.58 h	0.845	60 m	0.80 -- 1.18	0.08
Au	198	2.70 d	0.412	2.6 d	0.34 -- 0.53	3
U^{238}	U^{239}	23.4 m	0.074	40 m	0.04 -- 0.42	1
U^{235}	FP's	Short	Many	40 m	0.04 -- 0.42	1

Table B-3 LOW-COST ACCELERATOR NEUTRON SOURCES

Manufacturer	Type	Voltage Supply	Operating Voltage, kv	Beam Current, ma
High Voltage Engineering Corp. Burlington, Mass., USA	AN 400	Van de Graaff	400	0.15
Texas Nuclear Corp., Austin 17, Texas	9500	Cockcroft-Walton	150	1
Societe Anonyme de Machines Electrostatiques, 21 Rue Jean Mace, Grenoble, France	Type J	Felici	150	1
20th Century Electronics, Croydon, Surrey, England	NGH 150	-	150	1

Two recent reports[B-14, B-15] describe the installation and use of Texas Nuclear Corporation Cockcroft-Walton neutron generators. The installation at the University of Michigan[B-8, B-14] is routinely producing a thermal neutron flux of 2×10^8 n/cm^2.sec, while the Oak Ridge National Laboratory[B-15] generates a fast neutron flux of about 3.5×10^8 n/cm^2.sec. The generator, which both these laboratories use,[B-16] consists of three units: the accelerator with vacuum system, a high-voltage power supply, and a control console. These units, interconnected by cables, can produce neutrons immediately upon installation with no further operations necessary except for mounting a tritium target in the target holder. This generator, which costs about $20,000, accelerates deuterons to 150 kv (Fig. B-2), the deuterons strike a tritium target, and the $H^3(d,n)He^4$ reaction takes place producing 14 Mev neutrons. Both installations are in suitably shielded rooms equipped with pneumatic tubes to introduce samples into the accelerator beam and to move the activated samples into a counter. The ORNL generator is equipped to do only fast-neutron activations. The Michigan generator has a 30- by 30- by 30-in. water tank so that the neutron beam can be moderated with water; it can therefore be operated as either a fast- or thermal-neutron generator.

One of the most difficult problems with this type of generator is the tritium target. The usual commercial target is about 1 curie/in.^{2} of H^3 adsorbed in a layer of titanium about 200 A thick on a 2-mil-thick stainless-steel backing. With a 150-kv stream of deuterons striking this target in a rather small area, it must be cooled, and even with cooling, damage and depletion of the target are rather rapid. To improve target life, Meinke and Shideler[B-14] developed a rotating target.

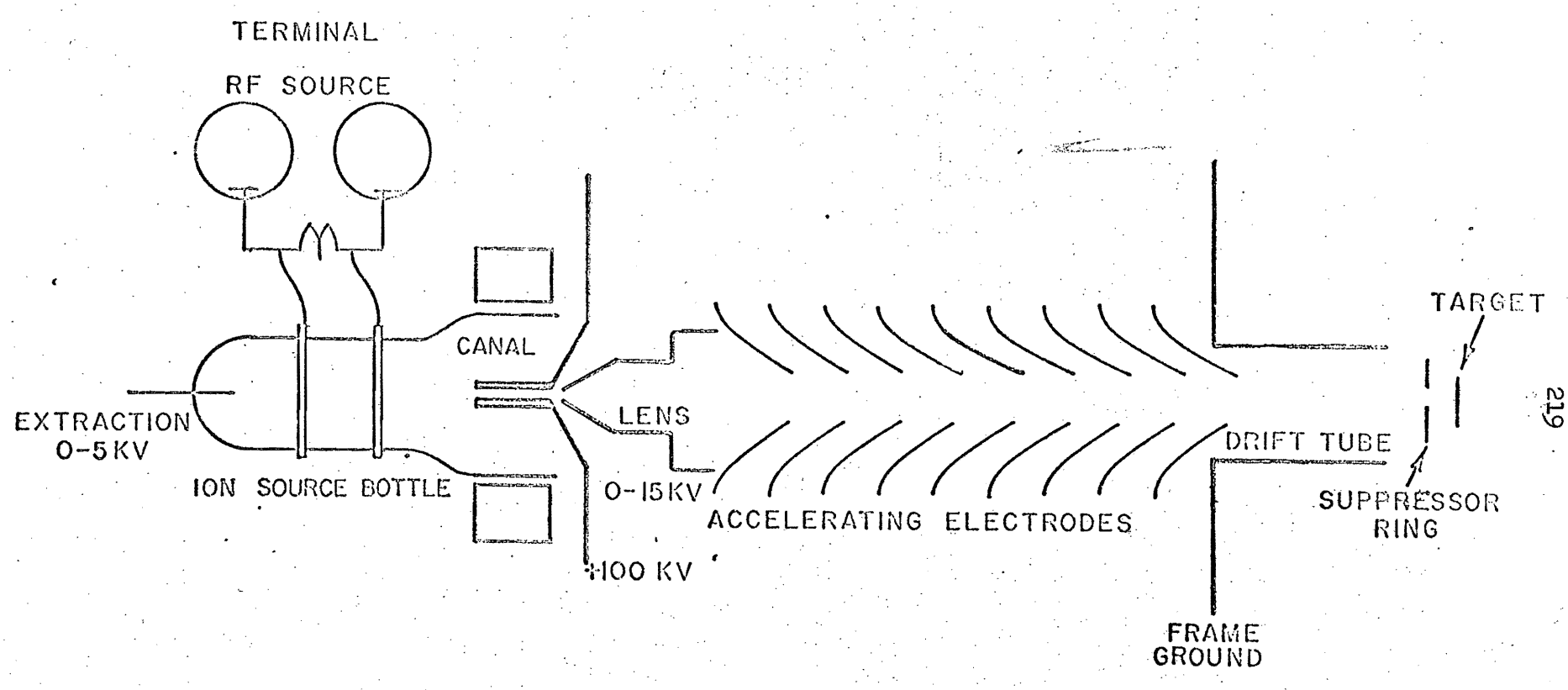

Fig. B-2 Schematic diagram of accelerator beam tube and ion source.

The target assembly (Fig. B-3) gradually rotates a long (10.5 by 2 in.) Ti-H^3 target in front of the beam and gives an even flux of $>10^9$ n/cm^2.sec for approximately 22 hr. The strip of target material is clamped to the inner surface of a stainless steel drum, the active side of the target forming the inner surface of a cylinder 3.5 in. dia. The entire back surface of the target is cooled by mercury, which is circulated through a water heat exchanger by a small magnetic pump.

An "O" ring seal on a rotary shaft transmits motion from a 24-hr clock drive situated outside the housing to rotate the drum at a rate of 0.00725 in./min; this corresponds to ~200 min for about 95% target use in a beam 1.5 in. wide at 1 ma. The drum target is situated with its axis inclined 45° to the beam axis so that the beam strikes the target through the open end of the drum. This angle also increases the area of the target exposed to the beam, thereby reducing the beam-current densities on the target.

Neutron Source for Activation of Flowing Stream. An interesting type of neutron source has recently been described by Gluck, McFarling, Kircher, Townley, and Sunderman at Battelle Memorial Institute[B-17] in a study done for the USAEC Division of Isotope Development. Designed for activation analysis of a flowing stream, the irradiator uses a Po-Be source that can be lowered into an activation cell.

A combination of neutron-source storage container and activation-vessel holder was built with provision for an activation cell, which is shown in Fig. B-4 with the source in the stored position and surrounded with 16.5 in. of borated paraffin deposited in 0.25-in.-thick layers. The paraffin provides adequate shielding for a 50-curie source. Because of

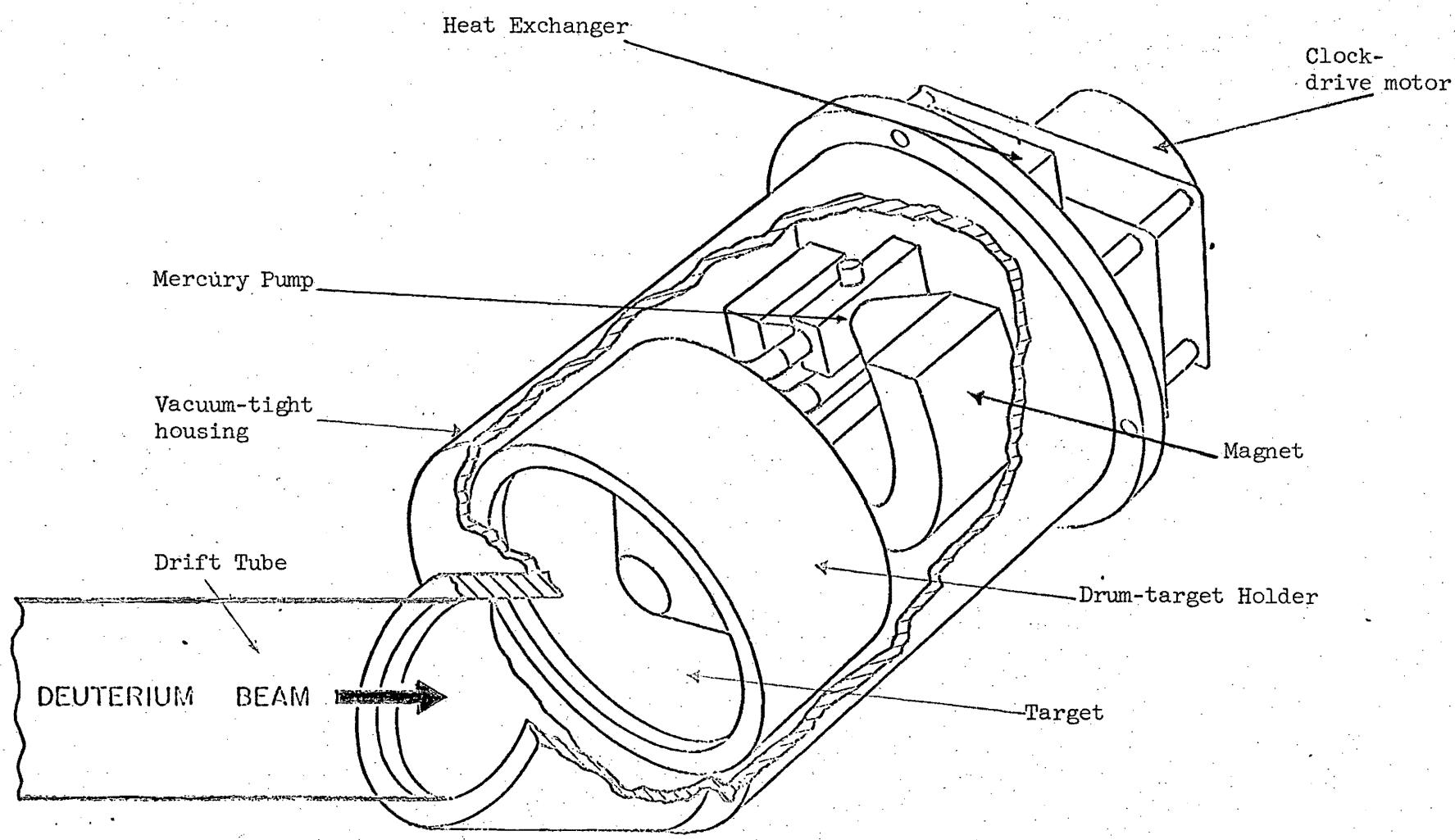

Fig. B-3 Long-lived target assembly.

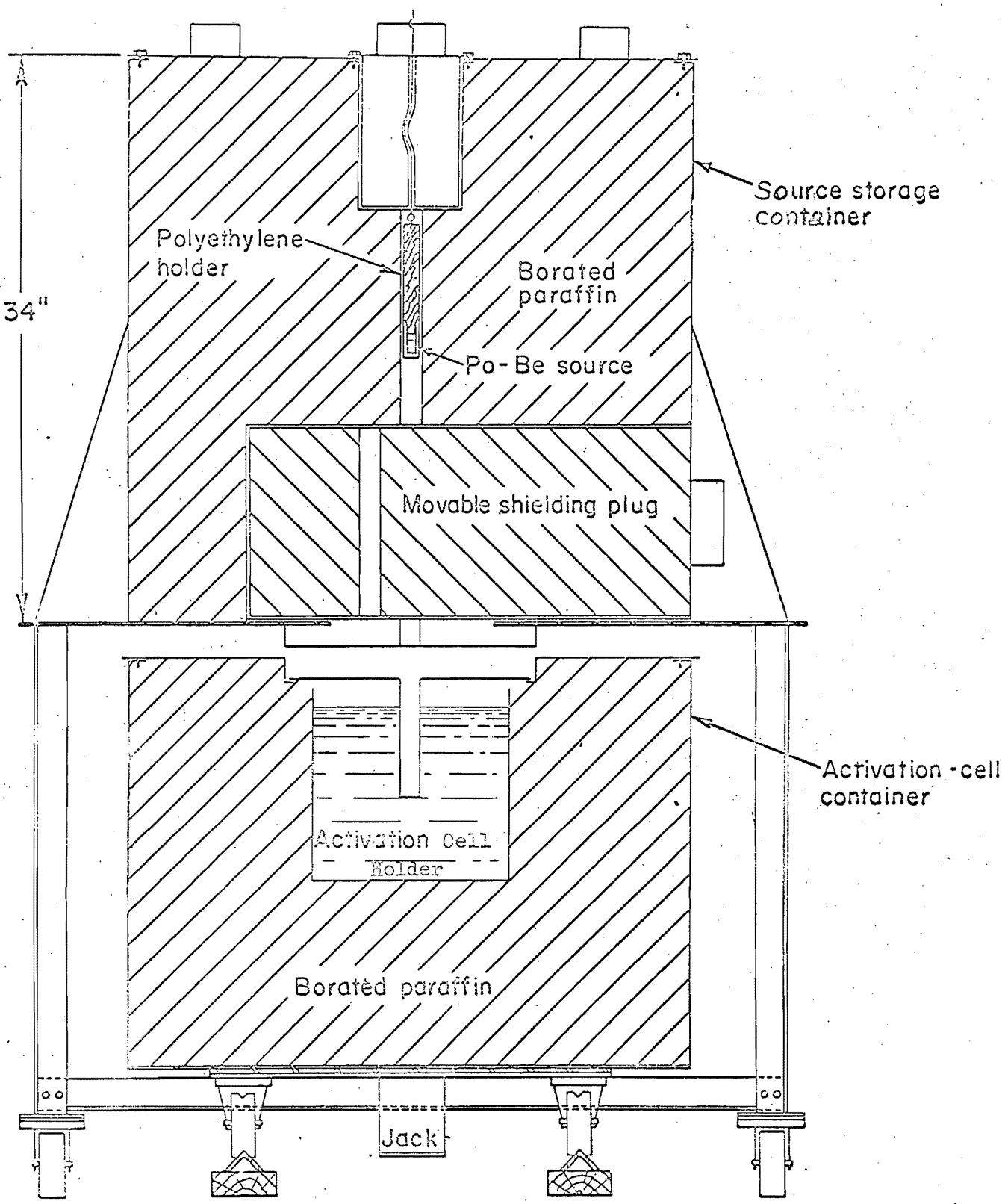

Fig. B-4 NEUTRON-SOURCE STORAGE SHIELD AND ACTIVATION CELL HOLDER

their weight the two containers were designed to move individually. An activation cell (Fig. B-5) with a volume of 2400 cm^3 was built to fit in this holder. This cell is constructed of 6-in.-wall stainless steel tubing, has a thimble suspended in the center, and overflows from the top. In order to achieve good mixing, a diffuser plate above the inlet and two baffle plates were incorporated. The outer surface of the activation vessel was coated with about 1.5 in. of paraffin and the vessel was permanently installed in the container. Provisions were made for washing and draining the cell. Void spaces around the installed cell were filled with polyethylene pellets for additional shielding.

To evaluate the system, the activation cell was installed in the cell holder. Figure B-6, a schematic of the flow system, shows the activation cell and the gamma-counting cell. The beta-counting cell can readily be substituted. During operation, inactive solution from the stirred process-solution tank was pumped through the system at a constant flow rate, entering through the bottom, overflowing from the top, and then passing through the counting cell and into the storage container. Solutions of four different elements were pumped through the cell, and their induced activity was determined as a function of the flow rate. The flow rates ranged from 50 to 490 ml/min, equivalent to activation times of from 48 to about 4 min.

Activation sensitivities for several elements -- manganese, indium, vanadium and silver -- were examined under flow conditions with the 50-curie source. Typical results for vanadium solutions are shown in Fig. B-7. The data plotted are the activities corrected for the different activation times by dividing the observed activity by $1 - e^{-\lambda t}$ where t is the irradiation

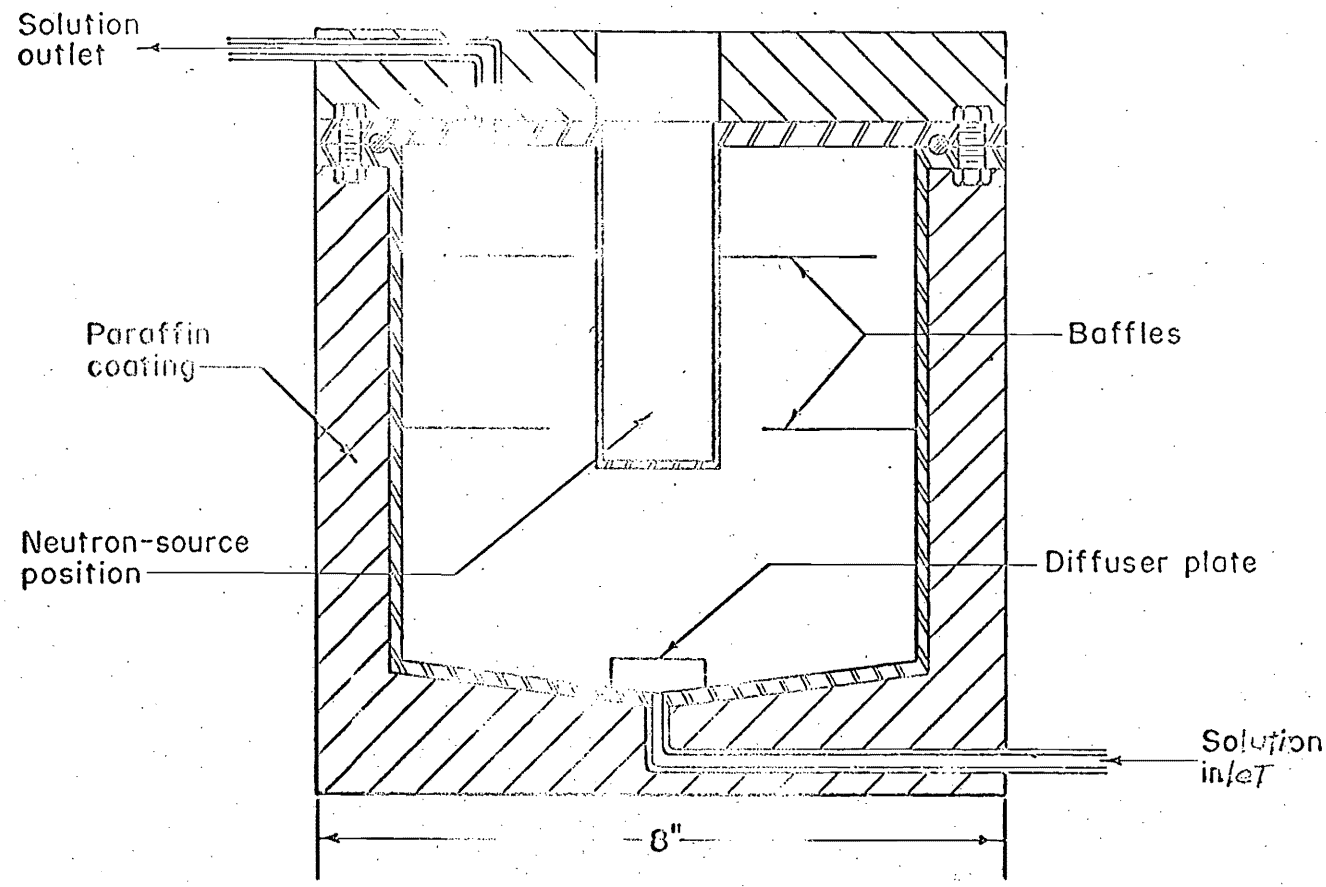

Fig. B-5 DETAIL OF ACTIVATION CELL

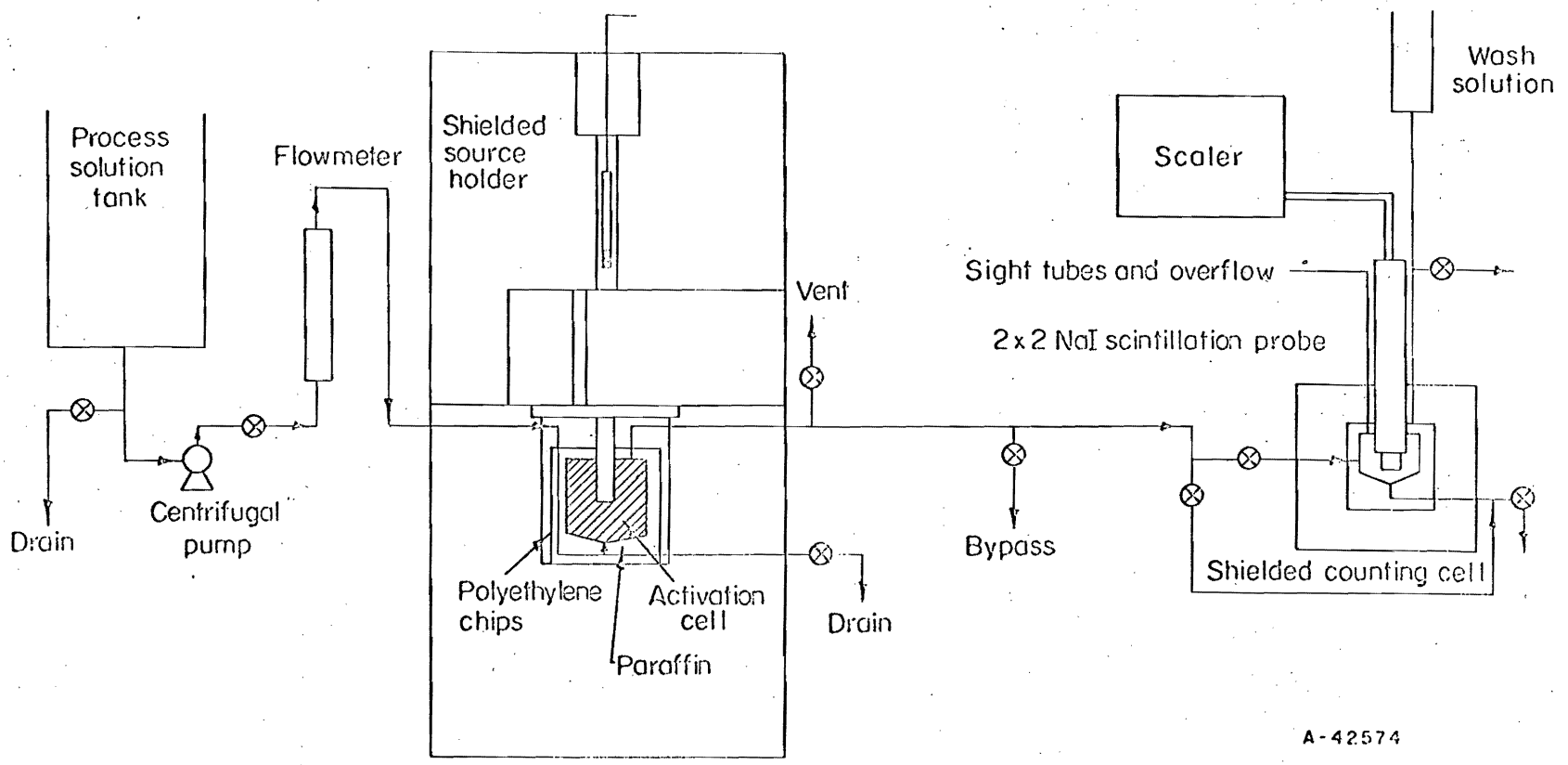

Fig. B-6 DIAGRAM OF THE CONTINUOUS-NEUTRON-ACTIVATION FLOW SYSTEM

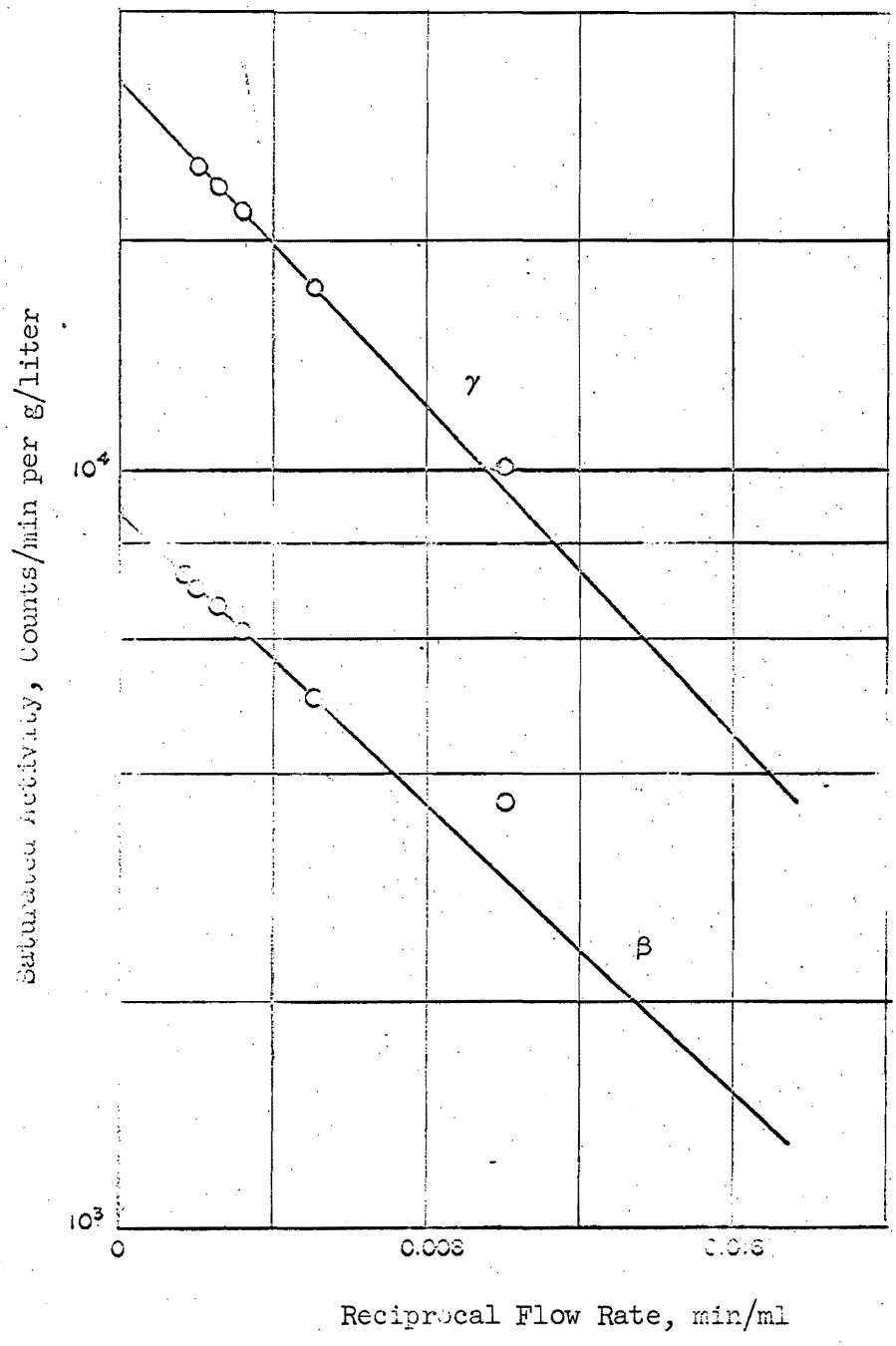

Fig. B-7 γ and β ACTIVITIES OF V^{52} PRODUCED BY ACTIVATION OF VANADIUM SOLUTIONS IN A FLOWING SYSTEM. $V^{52}t_{1/2}$ = 3.76 min
Apparent holdup volume = 675 ml for γ
= 620 ml for β

time. The effective irradiation time was calculated for each flow rate by dividing the volume of the activation cell (2400 ml) by the flow rate. Vanadium-52 with a half-life of 3.76 min was produced.

The corrected activity at the detector was found to decrease with decreasing flow rate, which is indicative of a time delay between activation and counting. Such a delay is inherent in this system for low flow rates or short-lived activities or both, because of the relatively large counting-cell volumes. The calculated volume necessary to account for the observed results, called "apparent holdup volume", was found to be 675 and 620 ml for data obtained with the gamma- and the beta-counting cells, respectively. It is difficult to account for these exact volumes, but they depend on the counting cell construction and method of injecting the solution to be counted, the activation-cell construction, and the amount of mixing in these cells. The assumption of this holdup volume, with the resulting radioactive decay as the solution traverses the volume, accounts very well for the results as shown in Fig. B-7 except at low flow rates. It is apparent that mixing of the solution is not good at the low flow rates.

Rapid Methods of Analysis

In addition to small portable neutron generators being needed, analyses should be done rapidly. Three different approaches are being investigated: (1) quick radiochemical separation methods following activation so that isotopes with short half-lives can be determined; (2) instrumental methods in which neutron activation is followed by gamma-ray spectrometry with no chemistry, the gamma-ray spectrum frequently being determined several times to measure half-lives of various

activities;[B-20, B-21] (3) very elaborate techniques in which the activation, counting, and calculating sequences are automatically controlled.

Fast Radiochemistry. Meinke and his coworkers[B-18, B-19] have been working for several years to develop and collect methods for the rapid separation of radioisotopes. While it is not possible in a brief review of this nature to condense or abstract his 125-page collection of methods,[B-19] a brief review of the guide lines and philosophy that have gone into the preparation of this collection is instructive and informative:

> "Radiochemical separations offer one major advantage over standard analytical techniques in that they do not require 100.0% yield. Since in radiochemistry we can determine the yield either by using carrier or by using tracers, we can utilize a much wider spectrum of reagents and procedures than can the classical quantitative analyst. The only limitation is that which is similarly imposed on the synthetic organic chemist, i.e., a separation step must give sufficient yield so that one can go on to the next step.
>
> Unfortunately, those who make use of radiochemical separations, whether in fission product analysis, nuclear decay schemes, or activation analysis, have too often tended to rely on adaptations of quantitative procedures which are lengthy and involved, to purify the desired radioisotope for measurement. Thus many of the early fission product procedures were developed in a hurry by combining many individual steps from quantitative work while often only a few steps of a multistep procedure were really effective. These early procedures were of necessity designed rapidly with the

information on hand to meet the emergencies of a war situation. Unfortunately many radiochemists today are blindly following these early procedures with little thought as to conservation of time and energy.

Workers in the field of activation analysis seem to be as guilty in this respect as those in other areas. A survey of published articles in this field shows two major classifications. In one group many investigators have sought to eliminate chemical procedures entirely with elaborate techniques of physical discrimination. In the other group the discrimination is chemical but is often quite involved and based usually on rather classical methods of separation.

Dr. Yusuru Kusaka and I have recently completed a rather thorough summary of radiochemical separation procedures reported in the literature for isotopes with half-lives shorter than 20 minutes. Although use of a number of ingenious devices was reported, particularly for isotopes of some of the lighter elements, the preponderance of the procedures was merely "speed-ups" of the standard analytical-type techniques such as precipitation, solvent extraction and ion exchange."

Instrumental Methods

In the "instrumental method" of Guinn and Wagner,[B-20, B-21] the gamma-ray spectra of samples are determined and compared with the spectra of known samples of simple compounds or elements to show the content of the unknown. The comparison of spectra by hand is a very tedious job, and programs are currently being prepared to do this work

on computers. Blotcky, Watson, and Ogborn at a recent symposium gave a paper describing a system using a 400-channel spectrometer and an IBM-1620 computer. At the same symposium, Guinn and Lasch described studies being done preliminary to preparing a program for the IBM-7090.[B-23]

Automated Methods. The most elaborate and sophisticated approach to instrumental activation analysis is the work being done at the Activation Analysis Research Laboratory of the A and M College of Texas by Wainerdi and coworkers.[B-22, B-24] This work is divided into two parts. In one part, an automated system is being designed to activate samples, count the samples with multichannel analyzers, and prepare the data in a form suitable for use on a computer. The second part of the work is the development of suitable computer techniques to resolve the gamma spectra and provide quantitative analytical data.

The original system for automated analysis[B-25] was designed to operate by either reactor activation of samples in groups of 100 followed by individual and automatic counting by individual and automatic introduction of the samples into the reactor for activation and automatic counting (Table B-4). With either of these operations, the samples are counted in a 256-channel vacuum tube gamma-ray spectrometer and the data are recorded on IBM cards for transfer to the computer. All sample transfers are pneumatic. In addition to its automatic features, this system also had built-in features that allow the operator to change almost any phase of the operation on each sample as it came through.

The speed of operation in the Mark I-Ia mode is determined by the irradiation and counting times. In a demonstration of operation in the Mark I mode, samples were processed at an average rate of 400 in 24 hr.

Table B-4 AUTOMATED ACTIVATION ANALYSIS

Mark I-Ia Mode of Operation

1. 100 samples are placed in a plastic storage block
2. The first sample is transferred from the storage block to the reactor
3. After proper irradiation time, the sample is transferred from the reactor to the 256-channel gamma spectrometer
4. The sample is counted for a predetermined time
5. The sample is transferred to a lead storage block and data are recorded on an IBM card
6. The cycle is repeated for each of the 100 samples

Mark I Mode of Operation

1. 100 samples previously activated in a reactor are placed in a lead storage block
2. The first sample is transferred from the storage block to the 256 channel gamma spectrometer
3. The sample is counted for a predetermined time
4. The sample is transferred back to the lead storage block and data are recorded on an IBM card
5. The cycle is repeated for each of the 100 samples

A downtime record for a period of several days showed that the machine was operating during 97% of the time. Table B-5 gives some results obtained with operation of the system in the Mark I-Ia mode. These are preliminary data, gathered and processed in an automatic mode with no corrections or smoothing processes applied outside the computer itself. The elements aluminum and silver were chosen to illustrate a good result and a rather poor one, respectively.

A new automatic system, designated MarkII, is under construction. It will take bulk irradiated samples, count them on one of three 400-channel transistorized gamma-ray spectrometers, and record the data on magnetic tape suitable for introduction into the computer.

The computer programs in this work have gone through several stages in their evolution. The original activation analysis computer program of Kuykendall and Wainerdi[B-26] used a highly complex, sophisticated spectrum-stripping technique which required many sequential measurements. This program, designed for an IBM-704, used both gamma-ray energy and measured half-lives for identification. Such a program was not readily adaptable for use in a fully automatic system. The simplified program of Fite, Gibbons, and Wainerdi[B-27] was therefore developed with the assumption that large numbers of similar samples would be run, and a general identification could be made manually on a few samples. This program has recently been revised again.[B-25, B-28]

Uses of Activation Analysis

Many applications of neutron activation analysis were reported at the International Conference on Modern Trends in Activation Analysis held at A and M College of Texas in December 1961.[B-34] A number of applications

Table B-5. Analysis of Standards by Mark I-Ia Mode of Operation

Amount of Element, mg		
Added	Found	Error,%
Aluminum (Al_2O_3)		
0.725	0.742	+ 2.3
0.704	0.700	- 0.6
0.694	0.685	- 1.3
0.699	--	--
0.736	0.709	- 3.7
0.715	0.751	+ 5.0
0.656	0.664	+ 1.2
		2.3 (avg)
Silver (Ag_2O)		
0.158	0.115	-27.0
0.140	0.089	-36.0
0.093	0.110	+18.0
0.130	0.067	-47.0
0.149	0.093	-38.0
0.102	0.069	-32.0
0.093	0.097	+ 4.3
0.102	0.109	+ 6.9
		26.1 (avg)

were also recently reviewed by Wainerdi and DuBeau in a recent issue of Science.[B-35] Only a few uses in crime detection and for analysis of oceanography cores are reviewed.

Law Enforcement. Several laboratories have recently started an evaluation of the possibilities of using activation analysis to aid in the examination of evidence for law enforcement purposes.

Guinn[B-29] of General Atomics has developed a technique to determine whether or not a suspect has recently fired a gun. The technique involves careful cleaning of the suspect's trigger hand, activating the removed material in a reactor, chemically separating the Sb^{122}, Ba^{139}, and Cu^{64}, and determining these activities by gamma-ray spectrometry. In some cases, it is possible to tell not only that the suspect has fired a gun, but also the type of ammunition. Guinn is now cooperating with the Los Angeles Police Department on the application of this method.

Bate, Pro, and Leddicotte[B-30] of Oak Ridge National Laboratory and the Alcohol and Tobacco Tax Laboratory, Internal Revenue Service, after examination of a number of samples of black automobile paint and some soil and soot samples, have concluded that activation analysis would be a useful technique for the forensic scientist to use in identification of these materials. After each specimen of paint had been irradiated for 1 hr in a reactor at a neutron flux of 8×10^{11} $n/cm^2 \cdot sec$ and the radioactivity measurements made of the short-lived radionuclides, the paint was allowed to decay for 1 day. Each sample was then reirradiated for an additional 140 hr to build up sufficient radioactivity of longer lived radionuclides. After this later irradiation, each specimen was allowed to decay for about 6 hr before radioactivity measurements were

made. This time was long enough to allow the short half-lived radionuclides in each sample to decay sufficiently so that radionuclides of longer half-lives could be more easily detected and determined. After these measurements, the samples were allowed to decay, with occasional radioactivity measurements, until the spectra of the induced radionuclide began to show strongly the longer lived radionuclides. This decay time was 30 hr.

The specimen was again analyzed by gamma spectrometry, and the spectral data recorded. Additional data were recorded 7 days after reactor discharge. Table B-6 lists the stable elements found in the finishes. The relative amount of each radioactive species present may be determined from the spectral data. Apparent peak-height ratios for Ba^{139} to Mn^{56} and Ba^{131} to Zn^{65} are also given. Since all the samples were irradiated in the same neutron flux, the photo-peak heights (radioactivity) for specific radionuclides in one sample may be compared to those found for the same radionuclides in another sample to show the relative abundance of one trace element to another. The intensity of these ratios can be used to identify or selectively separate sample materials.

The same type of procedure was used for soot and soil samples; and while promising results were obtained, identification of these materials is much more difficult because they contain many more constituents.

Jervis, Perkons, Mackintosh, and Kerr, working with the Crime Detection Laboratory, Royal Canadian Mounted Police, have used activation analysis to determine arsenic in human hair and other tissue to see if this analysis could be used to determine the cause of death in cases of

TABLE B-6

ELEMENTS FOUND IN AUTOMOBILE PAINT FINISHES

Paint	Mn	Ba	Zn	Co	Na	Cu	Cr	P	Ba^{139}/Mn^{56}	Ba^{131}/Zn^{65}
5	P	P	P	P	P		P		2	1.5
6	P	P	P	X	P	X			4	5
7	P	P	P	D	P	X			10	
8					P	P		P		
9	P			P	P					
10	P	P	P	D	P	X			10	5

P = elements definitely present.

X = possible but not definite.

D = present but at limit of detection.

suspected arsenic poisoning.[B-31] The results to date have been inconclusive. These same workers have made an investigation of the pattern of trace elements in human hair to try to use these patterns for identification purposes. In a case in which activation analysis of a hair was introduced in court,[B-32] the body of a young woman was found holding a single hair. Samples of hair from the victim and from a male suspect were compared with the hair found in the victim's hand by activation analysis, with the results shown in Table B-7. The difference between the P^{32}/Sb^{35} ratio in the two hairs was presented in the Canadian courts as evidence of identity.

Shipboard Activation Analyzer. A shipboard analyzer[B-33] performs rapid elemental activation analysis on core samples from the ocean floor on the vessel as they are collected. This aids effective exploration by making it possible for the ship to modify its survey pattern in accordance with the trends indicated by the data. The unit was developed by Dresser Research in conjunction with Lamont Geological Laboratory of Columbia University. The special neutron source is a small, rugged accelerator-type neutron generator which emits 10^8 neutrons/sec by the deuteron-tritium reaction. It is contained in a lead and paraffin shield designed to permit insertion of an entire ocean-bottom core sample so that intervals along the core length may be activated without cutting the core. A shielded 3- by 3-in. NaI (Tl) scintillation crystal is used to observe the activity induced in the core, and the gamma spectrum is analyzed in a 400-channel analyzer. Cores are analyzed for silicon, aluminum, magnesium, sodium, and iron contents by neutron activation and for potassium, uranium, and thorium contents by the spectra of their natural radioactivities.

Table B-7 NONDESTRUCTIVE ANALYSIS OF SINGLE HAIR IN EDMUNSTON, N.B., MURDER

	β-activity* total, c/min	P^{32}, c/min	S^{35}, c/min	P^{32}/S^{35}
Exhibit hair specimen (found in hand of victim)	3540	1783	1757	1.02
Male suspect hair	2462	1272	1190	1.07
Victim's hair	1908	1276	632	2.02

*β activity after 20 days decay

S^{35} low-energy component on β-absorption analysis; decay confirmed

P^{32} high-energy component on β-absorption analysis; decay confirmed

The activation conditions are: 10 min for irradiation of the core, 1 min for transfer of the core to the detector, and another 10 min for measurement of the induced activity. Analyses of standard samples have indicated the following sensitivities in mg/cm^3: aluminum and silicon, 0.3; magnesium, 1.6; sodium, 2.2; and iron, 3.8.

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RADIOISOTOPE SOURCES FOR RADIOGRAPHY

X rays in the energy range from 5 to 100 kev play an important role in the growing use of radiation in research and technology. Their principal areas of application are density and thickness gaging, industrial and medical radiography, and analysis by absorptiometry and fluorescent excitation techniques. The need for radiation in this energy range is currently covered by conventional x-ray machines. But, because these machines are not completely portable and require a source of electric power for operation, many investigations have been carried out to replace or supplement them with radioisotope sources. Two types of such sources are currently under investigation:[R-1] low-energy gamma emitting isotopes, and bremsstrahlung sources.

Low-energy Gamma Emitters

Very few long-lived isotopes emit low-energy gamma rays, and because of the complex decay schemes of most of them, they are of limited use where a narrow photon energy distribution is needed. Isotopes such as I^{125} and Fe^{55}, however, which decay by electron capture, give off monoenergetic photons. These isotopes, which have an appreciable half-life, have recently been prepared as effective low-energy gamma sources at ORNL.

Iodine-125 Sources. Iodine-125 decays[R-2] with a 57.4-day half-life by electron capture to the 0.0354-Mev excited state of Te^{125}. The Te^{125} decays by emission of a gamma ray which is largely converted in the K shell and results in emission of the characteristic 0.0273-Mev K_α x ray. Thus, the net result of the decay of I^{125} by these modes is the production of two coincident photons with energies of 27.4 and 35.4 kev (Fig. R-1).

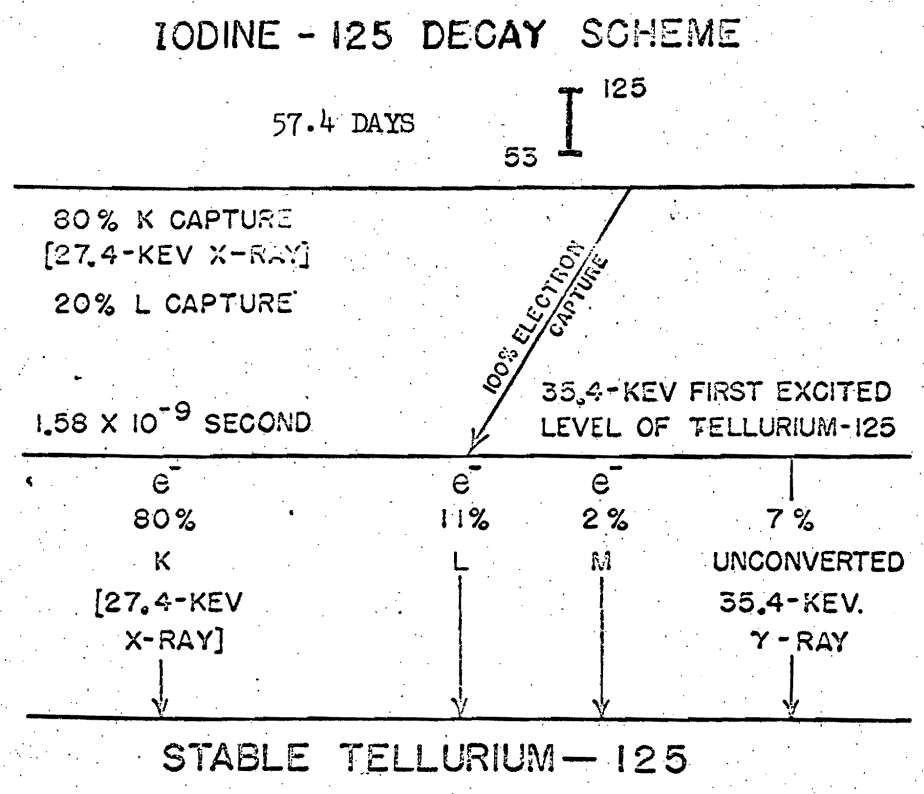


Fig. R-1 DECAY SCHEME OF IODINE-125[R-2]

These decay characteristics make I^{125} ideally suited for a portable x-ray source if a "point" source of relatively low self-absorption can be prepared.

Availability of high-specific-activity I^{125} makes possible the fabrication of concentrated sources. Although I^{125} can be produced in a cyclotron by the nuclear reactions $Te^{125}(p,n)I^{125}$, $Te^{124}(d,n)I^{125}$, and $Te^{125}(d,2n)I^{125}$, it is produced more efficiently in a reactor by the reaction

$$Xe^{124}(n,\gamma)Xe^{125} \xrightarrow[18\ hr]{EC} I^{125}.$$

The cross section for the xenon reaction is estimated at 74 barns.

A 425-cc sample (STP) of normal xenon gas (~0.1% Xe^{124}) was compressed in an aluminum can at about 100 psi and irradiated 21 days in the Oak Ridge Reactor at a flux of 2×10^{14} $n/cm^2 \cdot sec$ to produce 2500 mc of I^{125}. The time in the reactor was limited to limit the quantity of I^{126} produced.[R-3]

The irradiated sample was processed in a manipulator cell. The I^{125} was removed from the walls of the aluminum container by washing with 0.1 N sodium hydroxide solution; the iodine was oxidized to iodate by a slight excess of $KMnO_4$; and the solution was neutralized with H_2SO_4 while maintaining an excess of $KMnO_4$. The solution was heated to boiling, and saturated H_3PO_3 solution was added to reduce the iodate ion to elemental iodine. A small amount of H_2O_2 aids the reaction. The iodine was distilled into dilute sodium hydroxide solution.

Two sources, each of 2 curies, were prepared for study and evaluation. Two curies of I^{125} and 0.5 mg of normal I^{127} were precipitated together as silver iodide; the dried salt was sealed by welding in a stainless

steel capsule (Fig. R-2) with a 1-mil-thick 2-mm dia stainless steel window. The radiation through the window of this source was 55 r/hr at 3 in. from the center of the source as determined by a thin-wall ionization chamber.

Iodine and silver both have appreciable absorption coefficients for photons emitted by the I^{125}; therefore the efficiency of the second source was increased by using NaI^{125} instead of AgI^{125}. The I^{125} was distilled from a small volume into 1.0 ml of 0.0001 N sodium hydroxide solution. This solution was evaporated to dryness in an aluminum cup 0.090 in. i.d. by 0.070 in. deep and welded closed in an aluminum capsule 0.375 in. long by 0.140 in. dia and having a 0.010-in.-thick window. This capsule was placed inside another aluminum source holder with wall thicknesses of 0.020 in. The radiation from this capsule was 260 mr/hr at 16 in. as measured by a thin-wall ionization chamber. The capsule was placed in a G.M.R. Model 110 radiographic exposure device or "camera". A 5-mil stainless steel window in the radiographic exposure device decreased the radiation to 160 mr/hr at 16 in.

Several radiographs were made of the human hand at distances of 6 and 12 in. with exposures varying from 30 sec to 3 min.[R-4] Figure R-3 is a 2-min exposure at 12 in. using Kodak "Royal Blue" x-ray film with an intensifying screen. Some of the resolution is lost in reproduction, but the original pictures have a resolution comparable to that of standard x-ray machines. Figure R-4 is a hand exposure in which two pieces of glass are shown near the outermost joint of the ring finger. Also seen in the picture is a pocket dosimeter. This was a 30-sec exposure at 6 in. using Polaroid Type 3000X film.

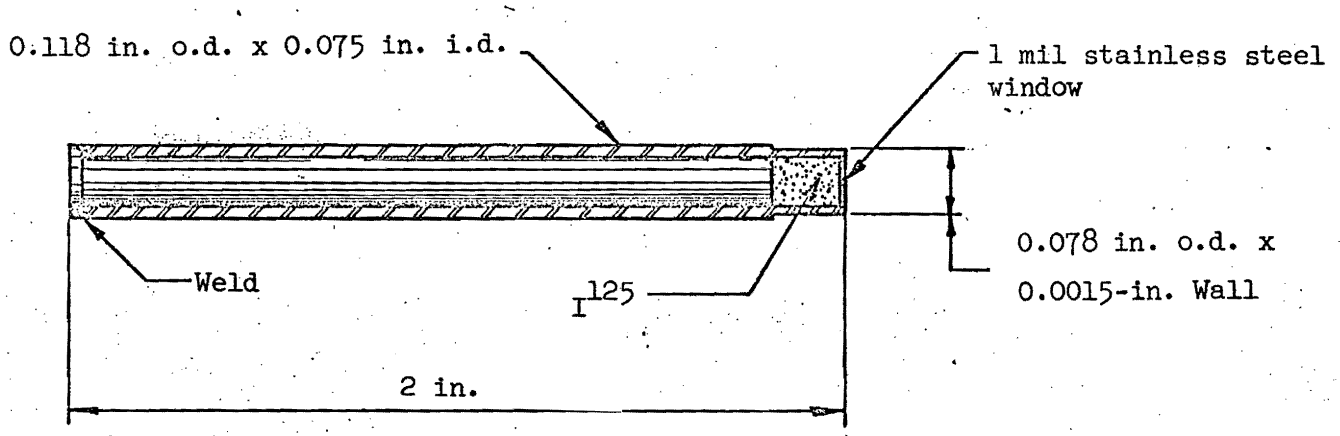


Fig. R-2 STAINLESS STEEL CAPSULE FOR I^{125} SOURCE

ORNL-Photo 60517

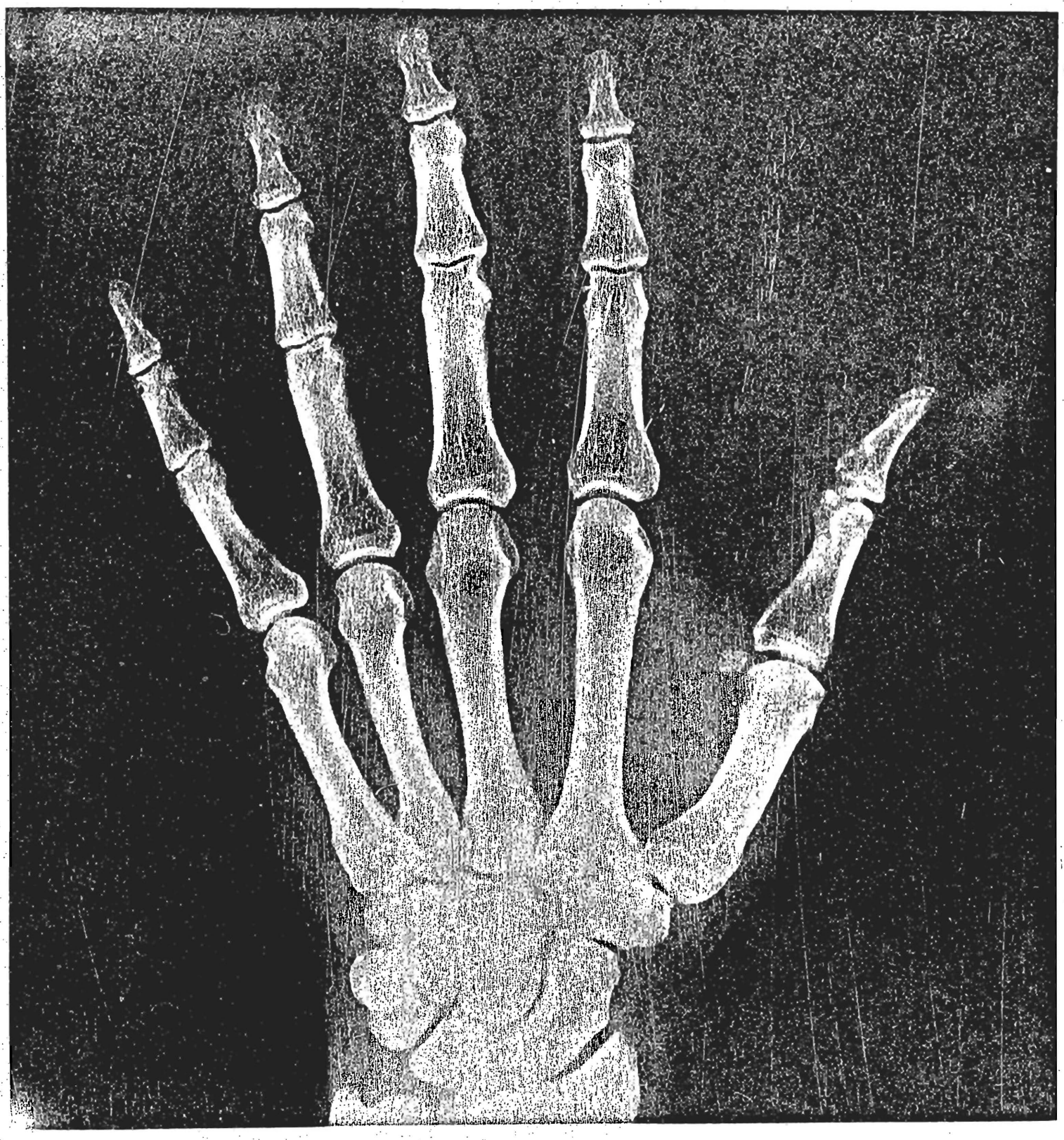
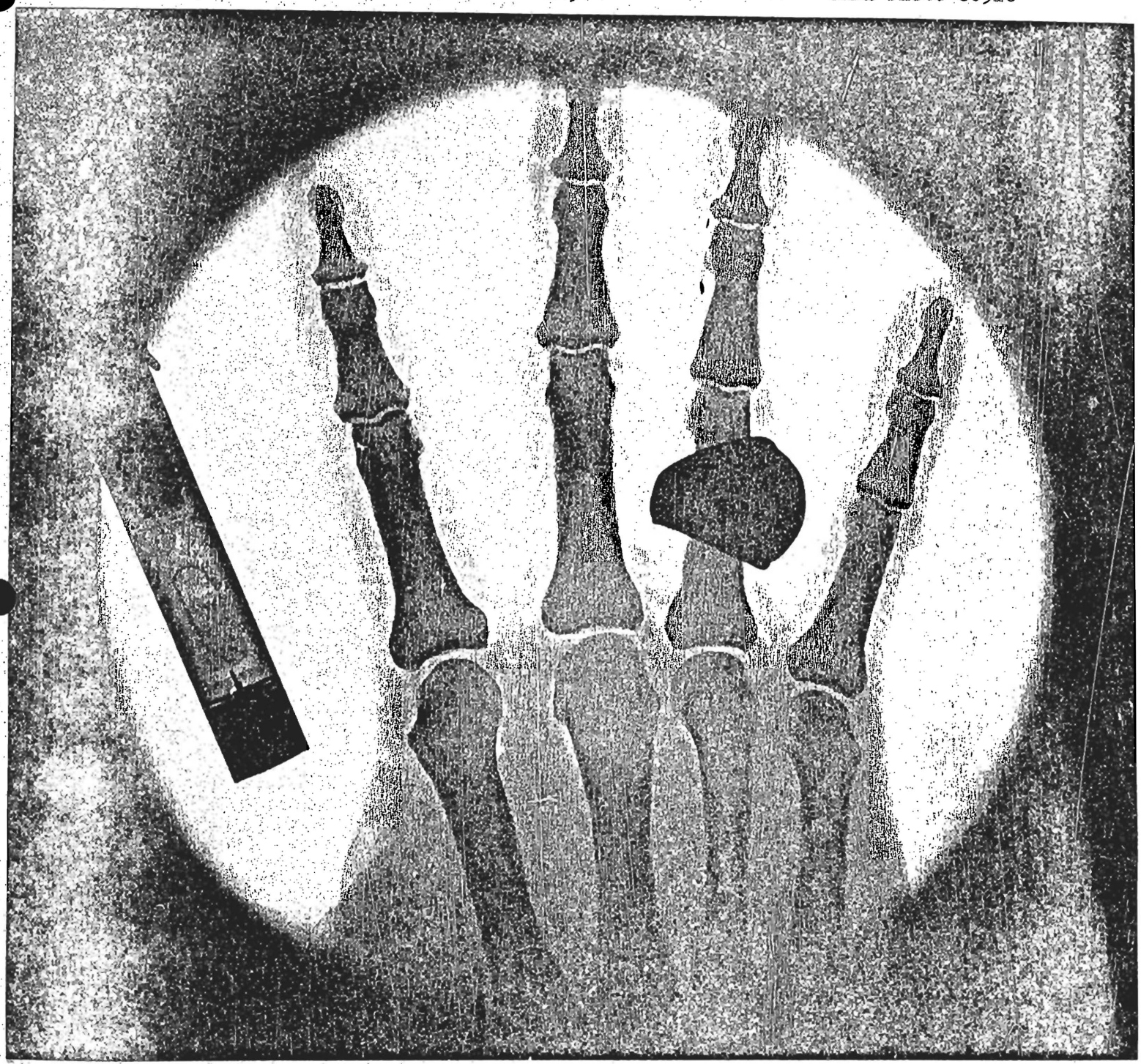


Fig. R3 SOURCE: I^{125} (1.96 curies) DISTANCE: 12 in.
TIME: 2 min. FILM: KODAK ROYAL BLUE X-RAY
DOSE: 9.5 mr

RADIOGRAPH OF HAND

ORNL Photo 60516



SOURCE : I^{125} (1.96 curies) DISTANCE: 6 in.

TIME: 30 sec FILM : POLAROID TYPE 3000X

Pi'8 R-4 RADIOGRAPH OF HAND

These radiographs show that this portable x-ray device can be effectively utilized as a diagnostic aid[R-5] without the necessity of elaborate equipment. Further testing will broaden the scope of usefulness of this source both in medicine and in industry.

Iron-55 - A Portable X-Ray Source. Iron-55 decays by electron capture with a 2.94-year half-life to yield the 5.9-kev manganese x ray. It is prepared by neutron irradiation of enriched Fe^{54}. This isotope has been especially useful for determining sulfur in petroleum[R-6] because of the high absorptivity of sulfur for this low-energy photon. Electron-capture isotopes with an appreciable half-life are also useful for x-ray-fluorescence analyses,[R-7] but can neither be prepared with high specific activities nor fabricated into point sources.

A source was made by spreading Fe^{55} oxide evenly over a circular area of a platinum strip by evaporating a nitric acid solution of $Fe(NO_3)_3$ and thermally decomposing the nitrate to the oxide. The oxide was reduced to metallic iron with hydrogen at 800°C, and the platinum strip was heated until the platinum on the surface started to melt and the metallic iron became coated with a thin layer of platinum. The area of platinum containing the radioactivity was cut from the platinum strip and rolled into a thin disk. Repeated rolling of the platinum increased the radiation from the source because the iron was spread more uniformly and the thickness of the platinum coating layer was reduced. The area of the source was ~2.4 cm^2, and the radiation, as measured by a thin-wall ionization chamber, was 800 mr/hr at 1.5 in. from the center of the source.

Radiographs of flowers were made with Polaroid 3000X film and Kodak Royal Blue x-ray film to illustrate that very small differences in densities can be resolved (Fig. R-5). No image intensifiers were used with the films. A helium atmosphere blanketed the flowers to decrease the absorption and scattering of the low-energy photons by air, and the exposure time was adjusted to give the best radiographs. These pictures are comparable to similar photographs taken with a low-voltage x-ray machine.[R-8] The same techniques can also be used to radiograph watermarks in stamps and valuable papers.

Bremsstrahlung X-Ray Sources

Isotopic x-ray sources can also be made which use the bremsstrahlung x rays produced when β rays strike a target. When high-energy betas are brought to rest in a target they lose their energy by radiative and ionization processes, most of the energy being dissipated in outer shell ionization of the target material. However, a substantial fraction of the energy of the electron goes into production of bremsstrahlung x rays and characteristic x rays following inner shell ionization. The energy distribution and flux of the x rays depend mainly on the nature of the target, the electron energies, and the strength of the beta source.

Several studies have been made recently on the preparation and use of bremsstrahlung x-ray sources. Brech of Jarrell-Ash Co. made a brief study of the use of Pm^{147} in analytical fluorescence spectroscopy.[R-9] Bersin and coworkers at Tracerlab studied the use of Kr^{85} clathrates with seven different targets[R-10] chosen to give a band of K_α energies between 10 and 100 kev. Reiffel,[R-11] Filosofo,[R-12] and Ezop[R-13] at the Armour Research Foundation, who have been especially active in this field,

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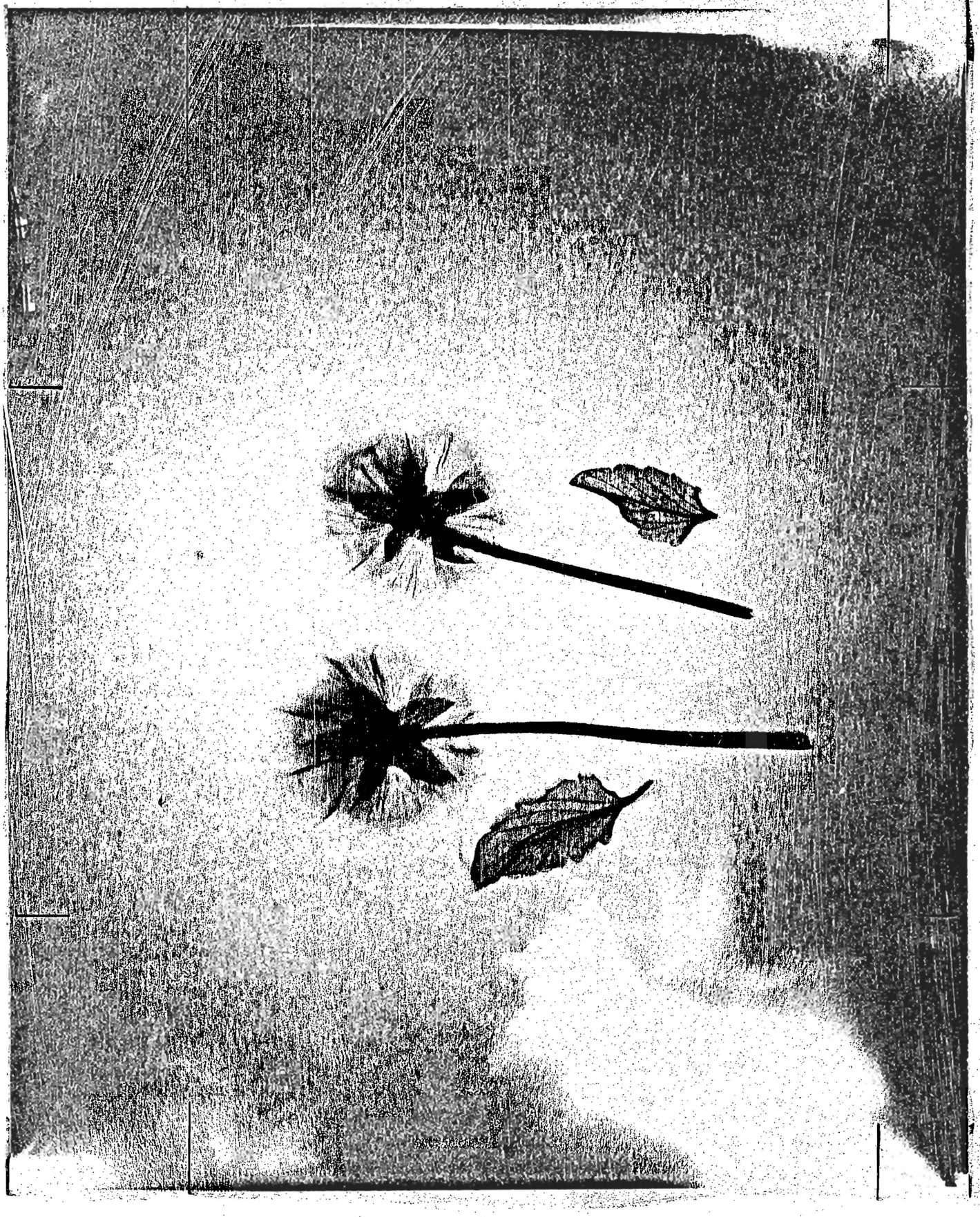

Fig. R-5

have concentrated their attention on the beta-emitting fission products Pm^{147}, Kr^{85}, and Sr^{90}-Y^{90}. They used tin and lead targets in most of their studies.

Promethium-147 X-Ray Sources. Promethium-147, a fission product now available in kilocurie quantities, decays by emission of a 0.223-Mev beta ray to Sm^{147} with a 2.65-year half-life. There is no known stable isotope of promethium. Sources were fabricated by compressing high-purity promethium oxide into thin pellets and encapsulating in aluminum source holders. Three hundred curies of promethium formed a cylindrical pellet 7.0 by 2.5 mm. A window thickness of 0.015 in. in the source holder allowed 94% transmission of the 38-kev photons.

The x radiation from the encapsulated 300-curie Pm^{147} at 16 in., as measured by an ionization chamber, was 280 mr/hr. Radiographs were taken of the human hand (Fig. R-6) with Polaroid 3000X film and of certain small objects to show how fine structures can be revealed (Fig. R-7).

Ezop[R-13] at Armour Research Foundation radiographed a human hand in 40 sec using a 250-curie Pm^{147} source 9 in. from Polaroid 3000X film.

The radiation from a Pm^{147} source can be increased by increasing the radioactive area of the source. If the pellet is made 2.0 by 0.2 cm, the output can be increased by a factor of ~8. Such a source may produce a detrimental penumbra effect; to improve the sharpness of the image, the distance from source to film would have to be increased.

The important advantages of using beta radioactivity for the production of secondary x rays have been demonstrated. In particular, Pm^{147}, because of its long half-life, low-energy beta emission, and freedom from decay products that produce higher energy photons, is particularly suitable for portable x-ray use.

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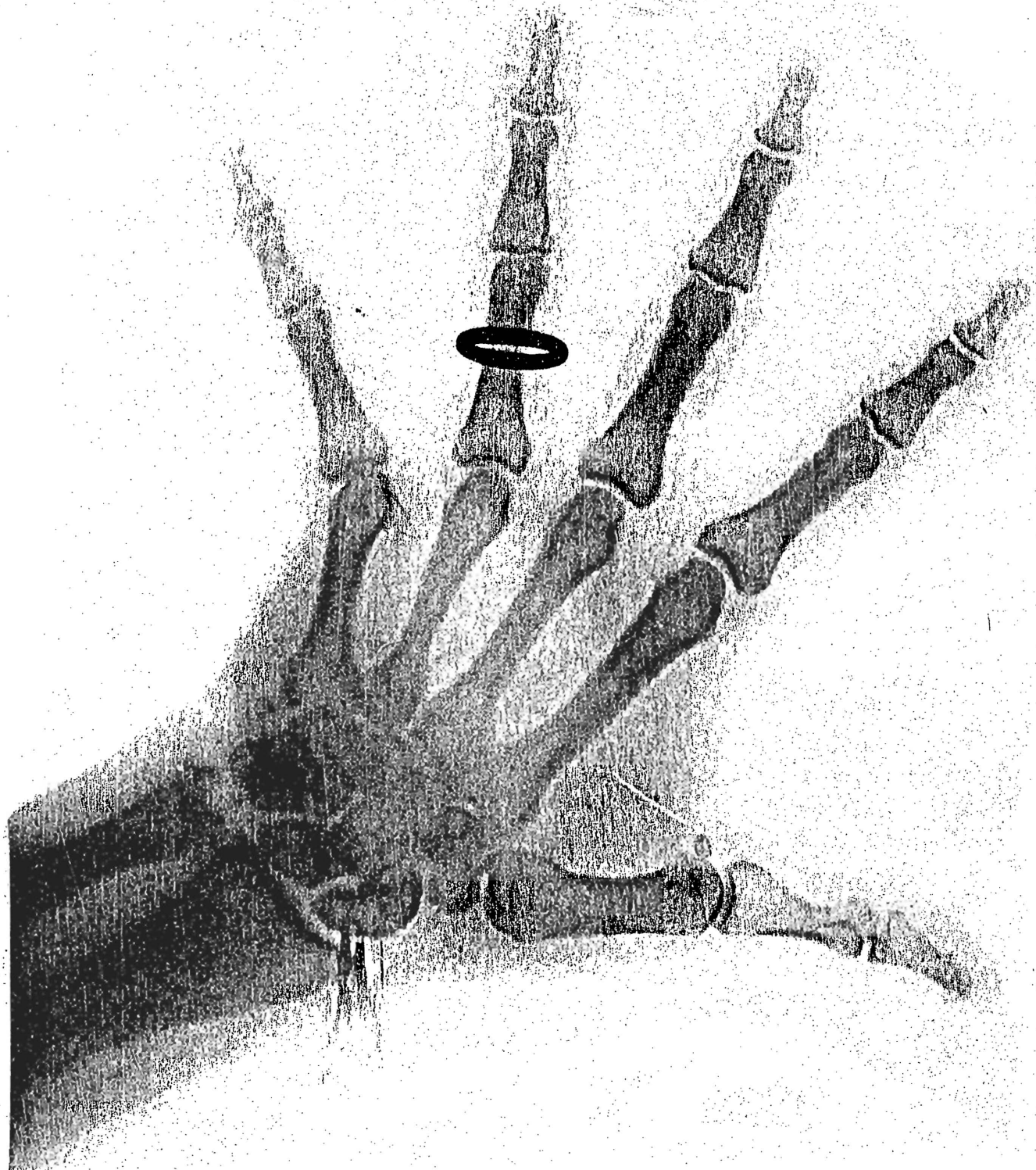

Fig. R-6

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Fig. R-7

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IV. PROCESS RADIATION DEVELOPMENT

One of the exciting applications of the radiant energy from radioisotopes is in the field of process radiation. This includes a broad spectrum of investigations as outlined in the talk by Fowler and Aebersold* and in another talk by Machurek.[1] For example, Yavorsky and Gorin have evaluated beta radiation as a hydrogenation catalyst.[2] Investigators at Battelle Memorial Institute have reported on the influence of radiation on graft copolymerization,[3-5] on radiation-induced nitration of hydrocarbons,[6] and on enhancement of catalytic activity from internal irradiation.[7] Armstrong and Rutherford at North Carolina State College have studied the modification of textile fiber properties by gamma radiation.[8,9] Other reports cover radiation-induced reactions in swollen polymers,[10] the accelerating effect of additives on radiation-induced graft polymerization,[11] and the mechanism of radiation-induced gelation.[12] The design of beta irradiators is the subject of a report by weinstock, Karalis, and Dell'Abate of Radiation Applications Incorporated.[13] A rather comprehensive report from William H. Johnston Laboratories, Inc. deals with basic studies in radiation technology.[14]

Considerable promise is also held for the future of food irradiation and there is much work going on in this field.[15] The preservation of seafoods is reported by Slavin, Steinberg, and Connors of the Bureau of Commercial Fisheries,[16] by Nickerson, Goldblith, and Masurovsky of MIT,[17] and by Novak and Liuzzo of Louisiana State University.[18] An excellent survey and bibliography of packaging requirements for radiation pasteurized foods has been published by Continental Can Company,[19] and the University of California, Davis Branch, has evaluated radiation

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*See p 39, this issue.

technology in conjunction with postharvest procedures for extending shelf-life of fruits and vegetables.[20] Many of these topics will be covered in detail in subsequent issues, with the food problem receiving some attention in the next issue of I&RT.

Brookhaven National Laboratory has been designated by the AEC as the Commission's center for studies of the effects of high-intensity radiation. A report of their laboratory, the High Intensity Radiation Development Laboratory (HIRDL), follows.

RADIATION ENGINEERING ACTIVITIES AT BROOKHAVEN NATIONAL LABORATORY

The BNL high-intensity radiation facility,[21] recently completed, and the program currently being carried out in the facility are described here. Discussions of the economics of the large-scale application of radiation in industrial processing invariably are inconclusive because of the lack of extensive, reliable data on source efficiency and of engineering experience in the fabrication and handling of megacurie quantities of radiation. The Division of Isotopes Development's program at Brookhaven is directed toward fulfillment of these needs.

Facility Design

The facility (Fig. IV-1) houses offices, laboratories, and a hot cell and canal complex. The cell complex (Fig. IV-2), designed for remote handling of 1 Mcurie of Co^{60}, is composed of a work preparation cell 13 x 15 x 14 ft high, an experimental irradiation cell 14 x 20 x 14 ft high, and a connecting canal with two bays. The preparation cell, with its associated special equipment[22] was designed for use in source decanning, sorting, encapsulating, welding, leak testing, calibrating, and assembling of about 600 kcuries of Co^{60} now on hand. However, a great deal of versatility has been built into the cell to permit handling of other isotopes and a wide variety of specialized irradiation experiments. The experimental irradiation cell is also versatile, having been designed for large-scale batch, as well as continuous, irradiations of gas, liquid, and solid targets.

The necessary shielding for this high activity is provided by 4.5 ft of 240-lb/ft^3 concrete on the three walls of each cell on which

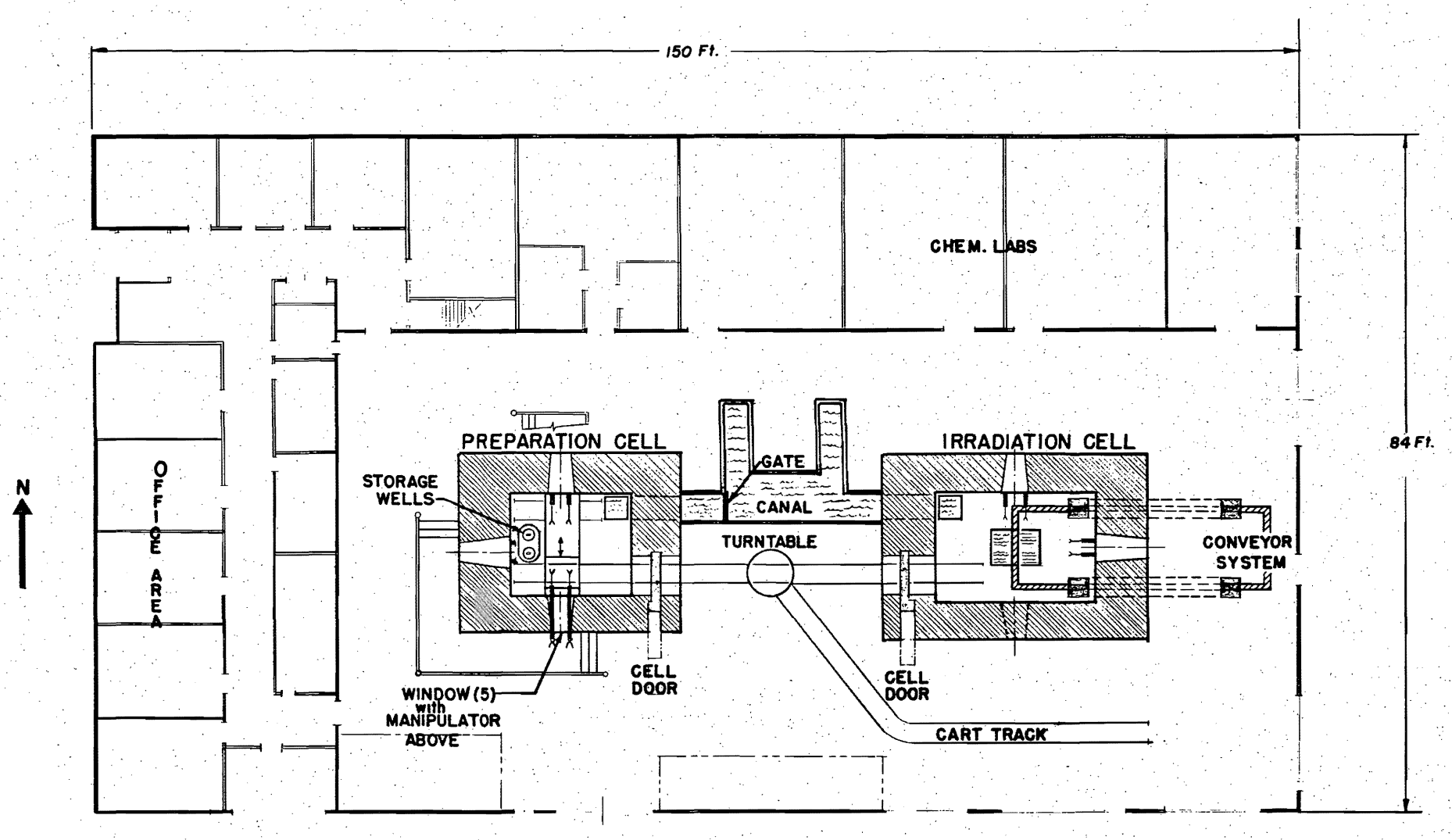

Fig. IV-1. Plan view of High-Intensity Radiation Development Laboratory at Brookhaven National Laboratory.

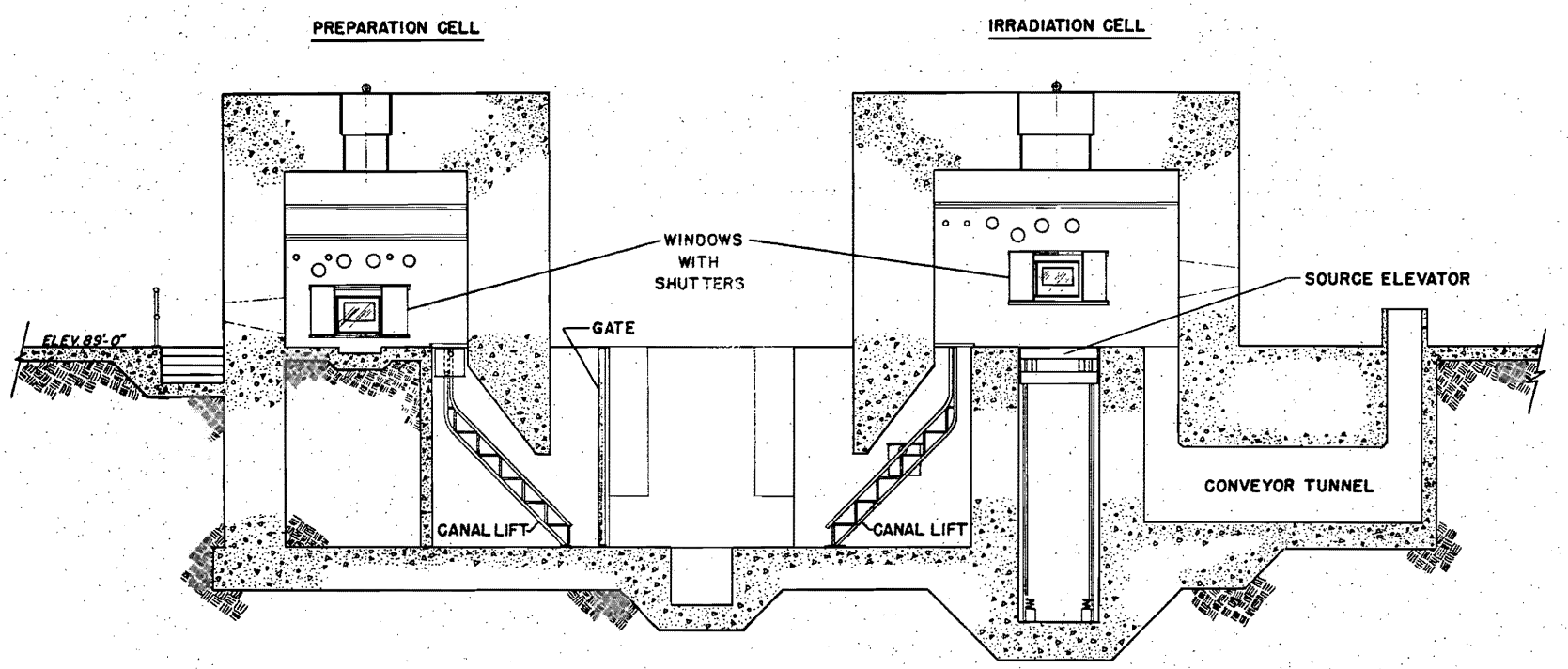

Fig. IV-2. Section view of cell-canal complex of the High-Intensity Radiation Development Laboratory at Brookhaven National Laboratory.

equipment controls and viewing windows are located. On the remaining walls and the ceilings the 145-lb/ft^3 concrete is 6.33 ft thick. Openings in the ceilings through which isotope shipping casks may be moved are covered with plugs. Uniformity in shielding is maintained by such devices as overlapping laminated steel plates in the recesses for installation of lights, panels, piping, and air ducts. In the preparation cell, below the floor level, are two water-cooled wells, with a capacity of 300 kcuries of Co^{60}, for storage of source elements during sorting, calibrating, and encapsulating. Both cells are equipped with heavy-duty, Model 8, manipulators and General Mills mechanical arms, which provide complete cell coverage. Provisions have been made in both cells for future installation of periscopes.

There are three viewing windows in the preparation cell and two, with provision for a third, in the irradiation cell. All cell windows are 3 x 3.5 ft on the inside, 1.5 x 2.5 ft on the outside, and 54 in. thick. Remotely operated lead shutters on each window will minimize radiation exposure of the lead-glass shielding windows when the windows are not in use and protect them against possible damage in case of an explosion inside the cells.

Each cell is equipped with a steel-encased lead access door, 7 ft 3 in. wide, 8 ft 3 in. high, 15 in. thick, and weighing 24 tons. The doors are remotely operated by an electrically powered chain drive, which slides them into and out of the entranceways. Pressure-sensitive safety stops on the forward edges of the doors prevent injury to personnel or equipment by moving doors. The mechanisms that operate the doors and elevators are connected to an interlocking safety system and to

high- and low-activity-level area-monitoring devices. To prevent trapping of personnel in the cell, the door is designed so that it cannot be closed from inside. To close the door, the operator must first energize a timing device inside the cell with a key, thus checking on occupancy. The same key actuates a switch located outside the cell to complete the closing.

Material is moved into and out of the irradiation cell and through the irradiation arrangements therein by a conveyor system. The two labyrinth entranceways that accommodate this system are three-segment U-shaped ducts with two 90-deg bends, extending downward into the floor to a depth of 14 ft, then horizontally in the direction of the cell for 22 ft, and finally up to grade inside the cell. The shaft portions are 2.5 x 2.5 ft and the tunnel portions, 2.5 x 6 ft. This cell also contains a remotely operated elevator inside a water-filled pit, 5 x 6 x 22 ft deep. Sources to be used for experimental irradiations may be assembled into arrays on the elevator and lowered into the water tank when not needed.

A stainless steel-lined canal, 45 ft long, 3 ft wide, and 16 ft deep (with a 20-ft-deep pit in the center), connects the preparation cell with the irradiation cell. It can be used for storing and transporting radioactive material. Remotely operated lifts move in the vertical water-filled shafts that go from the canal to each cell. At right angles to the canal are two irradiation bays, 3 ft wide, 12 ft long, and 12 ft deep, in which general laboratory irradiation services will be conducted. Conduit ducts from the bay area to the laboratory areas make possible the setting up of instruments in the laboratories to facilitate remote

recording of data. These underwater facilities will also be used for exploratory experiments and pilot runs prior to larger scale in-cell experimentation. A gate at the end of the canal, adjacent to the work preparation cell, may be used to isolate a portion of the canal water for testing and storage of newly encapsulated sources. This will minimize the possibility of spreading contamination from the work preparation cell.

A water-conditioning system treats the water in the canal and in the source elevator pit. A holdup tank connected to the preparation cell collects any contaminated waste.

Program

The program for the use of these facilities will develop data to be used in preparing a radiation engineering design manual. Gamma shielding and dose-distribution data will be compiled and correlated with theory in such a way that an engineer may arrive at sound irradiator designs. The manual will contain the following chapters:

I. Introduction

II. Radiation Physics

III. Computational Procedures

IV. Source Design and Fabrication

V. Irradiation Design and Fabrication

VI. Health Physics

VII. Source Shipping and Miscellaneous Problems

VIII. Economic Considerations

IX. Safety

X. Appendix

In BNL work with large-scale sources of low activity levels, a large number of experimental dose-distribution data have been determined for point, cylindrical, and slab sources. These have been obtained principally for homogeneous, 1 g/cm^3 density, infinite, semi-infinite, and finite targets.[23,24] A simultaneous program for determining experimental buildup factors for these geometries is continuing.[25] The data are being handled empirically, and experimental boundary correction factors are being formulated for one, two, and three boundaries.

The problem of gamma radiation scattering through air ducts in concrete shields has been investigated, in conjunction with efforts of the National Bureau of Standards, for large, rectangular, two- and three-segment ducts with 90-deg turns. Empirical calculations based on experimental data involving the duct parameters have been formulated.[26,27] Although these studies have not yet led to analytical techniques for duct design, preliminary results are promising. The results of recent BNL experimental determinations of the spectral distributions of scattered gamma rays inside air-concrete ducts hold promise of development of an analytical technique.

With regard to both the duct and the dose-distribution problems, the scattered photon spectra are, of course, of considerable interest. An experimental approach has been developed by which the theoretical and experimental response matrix of a scintillation spectrometer is used to convert the observed spectrum to the actual spectrum of the target. The scintillators are small, 0.5 x 0.5 in., plastic and sodium iodide crystals. The IBM-7090 spectrum-unfolding technique used, which tests the corrections to the observed spectrum, yields the most probable

spectrum, with an estimated accuracy of 10%. Measurements based on these techniques will be useful in studying the effect of boundaries and inhomogeneities and in multiple-scattering gamma-ray problems, which thus far have been solved only mathematically by Monte Carlo techniques.

A large part of the analytical and experimental effort is on development of computational procedures for isotropic Co^{60} and Cs^{137} sources and finite water, water-air, and water-aluminum targets. The penetration accuracy range is 0-10 mfp* ±5%. Source geometries are point and finite plane, slab, line, and rod. Target geometries are homogeneous, heterogeneous, and multilayer slab and cylinder. For many of the applications under consideration, specifications require that the ratio of the maximum-to-minimum dose at any two points in the target not exceed 1.25. In most cases the target is less than one mean-free-path thick. The calculational problem is similar to that in shielding problems except that a knowledge of the distribution of the radiation flux within the target to 5% accuracy is required instead of knowledge of the quality and quantity of radiation escaping from the shield with perhaps 20% accuracy.

Techniques are being developed by the Monte Carlo method, the only one appropriate for the cases under consideration. These techniques, in the development of which the Bureau of Standards and Yale University will collaborate, will ultimately make possible the handling of a wide variety of dose-distribution problems. The method of successive scatterings is too limited to be appropriate here. Calculations by the moments methods, primarily those of Goldstein, although appreciable, apply only

*Mean free path.

to infinite media.

A sizeable effort has been expended on the development, testing, and evaluation of new high- and low-activity-level dosimetry systems. Among the high-level systems developed at BNL are two silicon solar cell dosimeters[28,29] and two ferrous-ferric sulfate chemical dosimeters.[30] The solar cell dosimeters are the "n" on "p" type dose rate meter with a range of 10^2-10^9 rads/hr, and the "p" on "n" type integrating dosimeter, with a total dose range from 10^5 to 10^8 rads. The chemical dosimeters are modifications of the conventional Fricke dosimeter and may be called the "aerated" and "deaerated" Fricke dosimeters. They extend the Fricke range from 10^4 to well beyond 10^6 rads.

The need for reliable low-activity-level dose rate measuring instruments has led to development of a scintillation dose rate meter with a range from 0.1 mr/hr to 1 r/hr. It contains a very small, 0.25 x 0.25 in., anthracene crystal connected by a long, thin, Lucite light pipe to a lead-shielded photomultiplier tube and is suitable for point measurements.

Analytic and experimental efforts for developing generalized methods for irradiator design are in progress at BNL. A considerable amount of data has been generated, aimed at evaluating irradiator design parameters, e.g., source energy and overlap, source-to-target airgap, source element design, and standardization, encapsulation, testing, and safety.[31] Design parameters have been studied for Co^{60}, Na^{24}, and Cs^{137} slab irradiator systems.[32] The effect of heterogeneous targets on design parameters has been considered in these studies. Analytical methods are also being developed to treat a limited number of standard

cases of combinations of moving sources and/or moving targets by equating them mathematically to static cases.

In general, these design and analytical efforts are aimed at optimizing the efficiency of gamma-ray and gamma-energy utilization, not only for developing a particular design or obtaining a particular piece of information, but also for gaining a better understanding of the fundamentals involved. This is illustrated by the study of electrical breakdown phenomena in cerium-protected lead shielding glasses under intense gamma irradiation, which has been known to cause catastrophic internal electrical discharge. The study of this phenomenon is practical since it may occur in the window of a high-intensity-radiation cell. It is also a means for studying some of the basic mechanisms of radiation damage in solids. Both experimental and theoretical studies in this field are underway.

At BNL a theoretical calculation involving classical electromagnetic theory and a theory of separation of charge has been made to determine the buildup of electrical potential inside typical shielding glasses as a result of gamma irradiation. The experimental program is designed to test the predictions of the theory and to correlate the breakdown process with other effects such as cerium color center formation and fading, the buildup of temperature gradients, and the time duration and pattern of the discharge. Experiments to date indicate that glasses with high cerium content experience electrical breakdown more readily and at lower total doses than those with low cerium.

D. B.

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FIGURE LIST

Dwg. or Photo Number	Caption	Page
BNL-7-733-61	Fig. IV-1. Plan view of High-Intensity Radiation Development Laboratory at Brookhaven National Laboratory	
BNL-7-734-61	Fig. IV-2. Section view of cell-canal complex of the High-Intensity Radiation Development Laboratory at Brookhaven National Laboratory	

V. SOURCE DEVELOPMENT

The decision to separate this section on "Source Development" from the next one on "Power Sources" was purely arbitrary. It was felt that the latter would be oriented toward the large-scale applications, whereas the former could emphasize the research and development activities (with the realization, of course, that overlap is inevitable).

Recent trends are toward preparing radioisotopic sources as insoluble compounds or in insoluble matrixes to decrease the contamination hazard if the primary encapsulation is destroyed. Low leach rates and high abrasion resistance make glass attractive as a radiation source material. This is especially true for radioactive monovalent ions, such as Cs^{137}, whose salts are very soluble. Much of the recent work at ORNL has been directed toward preparing glasses from the fission products, with strong emphasis on the preparation of cesium glasses and evaluation of their properties. Glasses prepared at ORNL include silicate,[1-3] borosilicate[4] and aluminosilicate,[5] with major emphasis on incorporating cesium into these glasses for use as radioactive sources.

ALUMINOSILICATE GLASSES

Aluminosilicate glasses are prepared by adsorbing a radioactive cation on a synthetic aluminosilicate ion exchanger (Decalso), mixing with a flux, and fusing at a high temperature. Although especially good for making cesium sources, any radioactive cation can be adsorbed on Decalso. The ion exchange capacity of Decalso is $\sim$2.0 meq/g; however, the amount of a carrier-free tracer that can be adsorbed on the ion exchanger depends on the distribution coefficient of that ion, which in turn depends on the salts present, their concentrations, and the selectivity of the ion exchanger rather than entirely on the capacity of the exchanger.

Adsorption Properties of Decalso

Cations that have been found to be strongly adsorbed on Decalso are Cs, Rb, Ca, Sr, Ba, Sc, Ti, Cr, Mn, Fe, Co, Ni, Cu, Zn, Y, Zr,

Ru(III), Ag, In, Cd, the rare earths, Au, Hg, Tl, Pb, Am, U, and Np. Capacity is maximum at the pH of incipient hydroxide precipitation. Anions in general do not adsorb on Decalso, although $P^{32}O_4^{\equiv}$ adsorbs on the zinc form of Decalso at pH 10 with a distribution coefficient of $\sim$ 30.

The Decalso used as a source matrix is first ground to $\sim$ 200 mesh, and the desired amount of material is contacted with the desired radioisotope solution for 1-2 hr. Exchange is complete for tracer concentrations in 30 min, but greater concentrations of cations require up to 2 hr for 90 - 95% adsorption and 24 hr for $\sim$ 99% adsorption.

Glass Preparation Studies

Decalso (7.5% Na_2O, 15.9% Al_2O_3, 49.5% SiO_2, and 26.1% combined water) melts at 1450 - 1500°C to a glass. Compounds tested as fluxes to lower the melting range and viscosity of the glass include B_2O_3, CaF_2, and CaO; however, only CaO gave a glass resistant to leaching. The melting points of Decalso-CaO mixtures are shown in Table V-1. Since 8-12% CaO gave the lowest melting glass, 10% CaO was used in the glass prepared for leaching tests. The composition of this glass after firing is 8.8% Na_2O, 18.7% Al_2O_3, 58.1% SiO_2, and 13.1% CaO. The glass is harder than ordinary plate glass, and very resistant to thermal and mechanical shock.

Table V-1 Melting Points of Decalso-CaO Mixtures

Composition, wt %		
Decalso	CaO	Melting Point, °C
100	0	1400
96.5	3.5	1240
92.5	7.5	1180
90.0	10.0	1160
88.5	11.5	1180
85.0	15.0	1210
80.0	20.0	1230
75.0	25.0	1250
70.0	30.0	1280
67.0	33.0	1290
56.5	43.5	1320
53.0	47.0	1400

Table V-2 Leachability of Decalso Glass Sources

	Isotope Leached from 1-cm^2 Source in 68 hr at 25°C, %			
Leaching Solution	Cs^{137} *	Tl^{204}	Sr^{89}	Ce^{144}
Distilled water	$<1 \times 10^{-5}$	$<1 \times 10^{-5}$	$<1 \times 10^{-5}$	$<1 \times 10^{-5}$
Process water	$<1 \times 10^{-5}$	4×10^{-5}	1×10^{-5}	1×10^{-5}
8 M HNO_3	1×10^{-2}	3×10^{-2}	2×10^{-2}	1×10^{-2}
6 M HCl	3×10^{-2}	3×10^{-3}	3×10^{-3}	3×10^{-3}
12 M H_2SO_4	1×10^{-2}	6×10^{-2}	2×10^{-2}	1×10^{-3}
25% HF	7.0	10.0	3.0	4.0
6 M NaOH	2×10^{-2}	1×10^{-3}	3×10^{-2}	1×10^{-2}
4 M NH_4OH	1×10^{-5}	2×10^{-4}	2×10^{-3}	1×10^{-4}
4 M $NaNO_3$	1×10^{-3}	1×10^{-2}	4×10^{-2}	1×10^{-4}
4 M NH_4NO_3	1×10^{-3}	1×10^{-2}	4×10^{-2}	5×10^{-5}
1 M NH_4F	5×10^{-3}	5×10^{-3}	-	-
Simulated sea water	1×10^{-4}	1×10^{-3}	1×10^{-3}	1×10^{-4}

* Loading equivalent to 6 curies of Cs^{137} per gram

Leaching Tests

In a 400-hr leach of the glass at room temperature, 2-5 times as much activity was removed as in a 68-hr leach (Table V-2). The glass was resistant to all acids, bases, and salts tested except HF. The tests were made on 0.1-mc sources of Cs^{137}, Sr^{89}, Tl^{204}, and Ce^{144}.

Source Preparation

Decalso can be used to prepare glazes which have high abrasion resistance and low solubility, yet are thin enough to be useful as sources of soft β or α radiation.

Sr^{89}. Ninety-nine percent of the activity of 7.4 mc of Sr^{89} was adsorbed on 0.5 g of Decalso in 24 hr. The Decalso was mixed with 10% CaO and fused at 1300°C in an alundum cup to give a glass layer 0.01 in. thick. There was no detectable loss of Sr^{89} on ignition. The source surface smeared 10^3 counts/min, and after 2 days' equilibration with 2 M HNO_3, 8 x 10^3 counts/min, indicating that etching does not decrease the count. The observed radiation output was 3.1 r/hr at 20 cm, the calculated value being 3.5 r/hr assuming no self-absorption in the source.

Alpha Sources. Alpha sources can be prepared from heavy elements Am^{241}, $U^{233}O_2$, or $Np^{237}O_2$ ions by adsorption on Decalso. Thin films of glass (glazes), 0.3-5.0 μ thick, are fused to ceramic or stainless steel. Finely ground glass mix (75% Decalso + 25% Be_2O_3, m.p. 1000°C, is used with stainless steel), 0.1 mg/cm^2, in benzene containing 1-2% Canada balsam, is spread on the backing material, and, after drying, is fused at 1000°C. The efficiency of the alpha sources as a function of the glaze thickness is shown in Table V-3.

Abrasion Resistance

The abrasion resistance properties of the Decalso-B_2O_3 sources are shown in Table V-4.

Table V-3 Efficiency of Am^{241} Alpha Sources of Various Thicknesses

1 mg Decalso = 6885 dpm α

Decalso in Glass, mg/20 cm^2	Calculated Thickness, μ	Measured Alpha dpm	Measured Alpha dpm/mg	Efficiency
1.20	0.3	8,088	6,735	0.978
2.20	0.6	14,773	6,714	0.975
4.60	1.3	30,200	6,564	0.953
4.90	1.5	34,280	6,996	1.016

Table V-4 Abrasion Resistance of Decalso - B_2O_3 Glaze

Source	Activity per cm^2	Thickness	Smear Material	Smear, dpm/cm^2
Cs^{137}	1 μc	1 mm	Smear paper	< 10
			Crocus cloth	< 10
Cs^{137}	100 μc	1 mm	Smear paper	< 10
			Crocus cloth	100
Sr^{90}	1 mc	1 mm	Smear paper	2000
			Crocus cloth	4000
Sr^{89}	7 mc	1 mm	Smear paper	4000
			Crocus cloth	4000
Tl^{204}	100 μc	1 mm	Smear paper	< 10
			Crocus cloth	100
Ce^{144}	100 μc	1 mm	Smear paper	< 10
			Crocus cloth	100
U^{233}	10^6 dpm	~~10 μ~~ 0.1 μ	Smear paper, dry	< 5
			Smear paper, wet	5
			Crocus cloth, dry	120
			Crocus cloth, wet	240
			Carborundum paper (000)	600

Granular Sources

Small sources capable of giving a high radiation dose can be prepared by absorbing a beta emitter (Pm^{147}, Sr^{90}, Cs^{137}, or Ce^{144}) on 325-mesh Decalso and firing to about 1000°C for 1 hr. At this temperature, although the Decalso particles do not fuse, the grains become glassy and the radioactivity is not leached. Since the particles are smaller than 325 mesh (44 μ), there is little beta self-absorption, especially with Sr^{90} and Ce^{144}. Since this type of source is easily prepared, it would be useful for short-half-life fission products as well as for the longer lived isotopes.

Assuming complete beta absorption, the maximum dose rate may be calculated in rads per hour per gram of absorber per curie for Pm^{147}, Sr^{90}, Ce^{144}, and Cs^{137}, where T_o is the maximum beta energy:

$Pm^{147} \rightarrow Nd^{147} + \beta$, $T_o = 0.23$ Mev/dis

$Sr^{90} \rightarrow Y^{90} + \beta$, $T_o = 0.54$ Mev/dis
$Y^{90} \rightarrow Zr^{90} + \beta$, $T_o = 2.27$ Mev/dis (total 2.81 Mev/dis)

$Ce^{144} \rightarrow Pr^{144} + \beta$, $T_o = 0.30$ Mev/dis
$Pr^{144} \rightarrow Nd^{144} + \beta$, $T_o = 2.98$ Mev/dis (total 3.28 Mev/dis)

$Cs^{137} \rightarrow Ba^{137} + \beta^-$, $T_o = 0.52$ Mev/dis

Assuming 1 g of adsorber, 1 curie of activity, and 100% absorption of the activity, calculated dose rates are:

Isotope	Dose Rate,* rads/hr.curie	Approximate Specific Activity, curies/g	Decalso Capacity, curies/g	Maximum Source Strength, rads/hr.g Decalso
Pm^{147}	1.6×10^5	400	35	5.6×10^6
Sr^{90}	1.9×10^6	80	7.2	1.4×10^7
Ce^{144}	2.3×10^6	300	27	6.2×10^7
Cs^{137}	3.6×10^5	33	8.7	3.1×10^6

$$* \frac{\text{rads}}{\text{hr.curie}} \frac{\text{Mev}}{3\ \text{dis}} \frac{(2.2\times10^{12}\ \text{dis})}{(\text{min.curie})} \frac{(60\ \text{min})}{(\text{hr})} \frac{(1.6\times10^{-6}\ \text{ergs})}{(\text{Mev})} \frac{\text{rad}}{100\ \text{ergs}}$$

SILICATE GLASSES

Cesium silicate glasses were prepared from SiO_2, carbonates of the divalent metals, and cesium carbonate; the $CsCo_3$ was prepared from cesium alums by the cesium tetraoxalate process.[7] Silicate glasses have the general formula[8] $xR_2O \cdot yRO \cdot zSiO_2$, where $x + y = 1$ and z varies from 2.5 to 4.0. The R_2O component can be Na_2O, K_2O, or, in the case of Cs glasses, Cs_2O. The function of the R_2O group is to decrease the viscosity of the glass; however, the proportion of R_2O should not be greater than 0.8 or the leaching rate of the glass will be excessive. The function of the RO group is to decrease the leaching rate and to improve the annealing properties of the glass. The SiO_2 is the major glass-forming constituent, and should be present to the extent of 2.0 - 4.0 moles per mole of the basic oxides (R_2O, RO). Glasses containing less than 2.0 moles of SiO_2 tend to crystallize, while those containing greater than 4 moles of SiO_2 are too viscous to flow well at 1200-1300°C.

Table V-5 shows the effect of various divalent oxides on the leaching rate, melting range, and density of the glass. Although there is little difference in the leaching rates of glasses of similar compositions, some of the oxides have disadvantages. Zinc oxide dissolves slowly in the glass melt and has a strong tendency to devitrify. Barium oxide glasses low in SiO_2 also have a strong tendency to devitrify after long heating at 800°C, but this is not true with > 3.0 SiO_2. Cadmium oxide volatilizes from the melt at 1150°C or higher, especially if a stream of air is passing over the melt. Lead glasses must be made in an oxidizing atmosphere to prevent formation of lead metal. Calcium and strontium oxides give good cesium glasses melting at 1300-1350°C, whereas barium oxide glasses melt lower, at 1200-1250°C. All the glasses in Table V-5 contain 35-45 wt % Cs. Table V-6 shows the effect of the R_2O:RO ratio on both the leaching rate in water at 25°C and the volatilization of Cs at 1200°C in the Cs_2O-CaO-SiO_2 system.

Table V-5

Effect of RO Group on Leaching Rate, Density, and Melting Range

Empirical Formula				Cs Content %	Cs Content c/g glass	Density, g/cm^3	Cs^+ Leaching Rate, $mg/cm^2.d$	Melting Range, °C
0.15 Li_2O	0.55 Cs_2O	0.3 CaO	2.7 SiO_2	41.5	41.0	3.00	1.7	1300-1350
0.15 Li_2O	0.55 Cs_2O	0.3 ZnO	2.7 SiO_2	40.5	43.1	3.22	2.4	1250-1300
0.15 Li_2O	0.55 Cs_2O	0.3 BaO	2.7 SiO_2	38.0	40.3	3.21	2.0	1200-1250
0.15 Li_2O	0.55 Cs_2O	0.3 CdO	2.7 SiO_2	38.8	41.8	3.26	2.7	1100-1150
0.15 Li_2O	0.55 Cs_2O	0.3 PbO	2.7 SiO_2	36.0	44.0	3.78	4.7	1050-1100
0.15 Li_2O	0.55 Cs_2O	0.3 SrO	2.7 SiO_2	39.8	41.5	3.17	2.0	1300-1350

Table V-6

Effect of the R_2O:RO Ratio on the Leaching Rate and Volatility of Cs^+ from $Cs_2O \cdot CaO$ Glasses

Glass			Volatilization Rate at 1200°C, $g/cm^2.hr$	Cs Leaching Rate, $mg/cm^2.d$
0.5 Cs_2O	0.5 CaO	2.7 SiO_2	0.5	< 1.0
0.55 Cs_2O	0.45 CaO	2.7 SiO_2	0.7	< 1.0
0.60 Cs_2O	0.4 CaO	2.7 SiO_2	1.2	1.0
0.65 Cs_2O	0.35 CaO	2.7 SiO_2	1.5	1.3
0.70 Cs_2O	0.30 CaO	2.7 SiO_2	2.0	1.7
0.75 Cs_2O	0.25 CaO	2.7 SiO_2	5.0	5.0
1.00 Cs_2O		2.7 SiO_2	9.0	V.S.

Generally, Cs leaching and volatilization rates from silicate glasses are low. However, glasses with a high calcium oxide content have a lower volatilization rate at 1200°C and a lower leaching rate than those with less divalent oxide. The SiO_2 content also has a great influence on the leaching and volatilization rates of cesium oxide from the glass. Table V-7 shows that increasing the SiO_2 content lowers the leaching and volatilization rates at 1200°C of cesium from the $Li_2O \cdot Cs_2O \cdot BaO \cdot SiO_2$ system.

A good cesium glass for molding glass bars is $0.8\ Cs_2O \cdot 0.2BaO \cdot 3.5SiO_2$.[9] This glass contains 45.7% Cs and has a density of 3.1 g/cm^3, giving a Cs^{137} activity concentration of 47.4 curies/cm^3. The leaching rate is < 2mg/cm^2.day and the volatilization rate < 2mg/cm$^2 \cdot$hr at 1200°C. The glass does not have a tendency to devitrify, and can be annealed at 500°C. This glass would be useful for molding cesium rods or bars for a radiation source or right cylinders for heat sources.

BOROSILICATE GLASSES

Cesium borosilicate glass, $Cs_2O \cdot B_2O_3 \cdot 4.2SiO_2$, has been prepared at ORNL[4] by precipitating cesium tetraphenylboron with 240-mesh SiO_2, calcining at 700°C to form $Cs_2O \cdot B_2O_3 \cdot 4.2SiO_2$, and melting at 1200-1300°C. The glass contains 44 wt % Cs and has a density of 3.1 g/cm^3. At 1200°C it volatilizes at a rate of 35 mg $Cs_2O \cdot B_2O_3$/cm$^2 \cdot$hr. The Cs leaching rate of such a glass is 0.2 mg/cm$^2 \cdot$d. Cesium borosilicate glasses are difficult to anneal and have a tendency to crystallize if cooled too slowly in the temperature range 1100-1200°C.

Table V-7

Effect of SiO_2 on Properties of $Li_2O \cdot Cs_2O \cdot BaO\ SiO_2$ Glasses

Glass				Cs_2O Volatilization at 1200°C, $mg/cm^2.hr$	Cs Leaching Rate in H_2O, $mg/cm^2.d$	Remarks
0.15 Li_2O	0.55 Cs_2O	0.3 BaO	2.0 SiO_2	10.0	10.0	Glass tends to be devitrified
0.15 Li_2O	0.55 Cs_2O	0.3 BaO	2.5 SiO_2	4.0	5.0	Glass tends to be devitrified
0.15 Li_2O	0.55 Cs_2O	0.3 BaO	2.7 SiO_2	2.0	2.0	Glass tends to be devitrified on long heating
0.15 Li_2O	0.55 Cs_2O	0.3 BaO	3.0 SiO_2	1.5	1.1	Good glass
0.15 Li_2O	0.55 Cs_2O	0.3 BaO	3.5 SiO_2	1.0	0.5	Good glass
0.15 Li_2O	0.55 Cs_2O	0.3 BaO	5.0 SiO_2	0.4	0.3	Viscous glass at 1200°C
0.15 Li_2O	0.55 Cs_2O	0.3 BaO	10.0 SiO_2	0.2	0.1	Very viscous glass at 1200°C

Table V-8

Properties of Radioactive Cesium Glasses Prepared at ORNL

Glass	Cs, %	Density, g/cm^3	curies/cm^3	watts/cm^3*	Leaching Rate in H_2O at 25°C	Volitization Rate**	Remarks
$0.8\ Cs_2O \cdot 0.2BaO \cdot 3.5\ SiO_2$	45.7	3.10	46.7	0.215	1 $mg/cm^2 \cdot d$	2 $mg/cm^2 \cdot hr$ at 1200°C	low volatility, low solubility, easily annealed, can be molded
$(Cs_2O,\ Na_2O) \cdot M_2O_3 \cdot 6\ SiO_2$	26	2.70	24.0	0.110	2.5 $\mu g/cm^2 \cdot d$	2 $\mu g/cm^2 \cdot hr$	high melting, very viscous, very low solubility and volatility
$Cs_2O \cdot B_2O_3 \cdot 4.2\ SiO_2$	44	3.10	44.9	0.207	0.2 $mg/cm^2 \cdot d$	35 $mg/cm^2 \cdot hr$ at 1200°C	high volatility, low solubility, difficult to anneal, tends to be devitrified

* 4.6 watts/curie Cs^{137}

** Volatility is controlled by diffusion and not by vapor pressure.

CONCLUSION

The properties of these three types of glasses are summarized in Table V-8.

Much of the present work has been done with fission products, but many of these same cesium glasses are amenable to use with other radioactive elements, such as those produced in cyclotrons and reactors; this is especially true of the aluminosilicate glasses. Incorporation of many of the more "exotic" tracers into glass for use as radiation sources will greatly increase the utilization of various radioactivities, such as positron, alpha, and soft x-ray emitters, as well as widen the field of gamma sources available for radiation studies. Incorporation of radioactive materials into glass, followed by encapsulation, approaches the ultimate in safety from contamination for the user of the radioisotope.

R. E. L.

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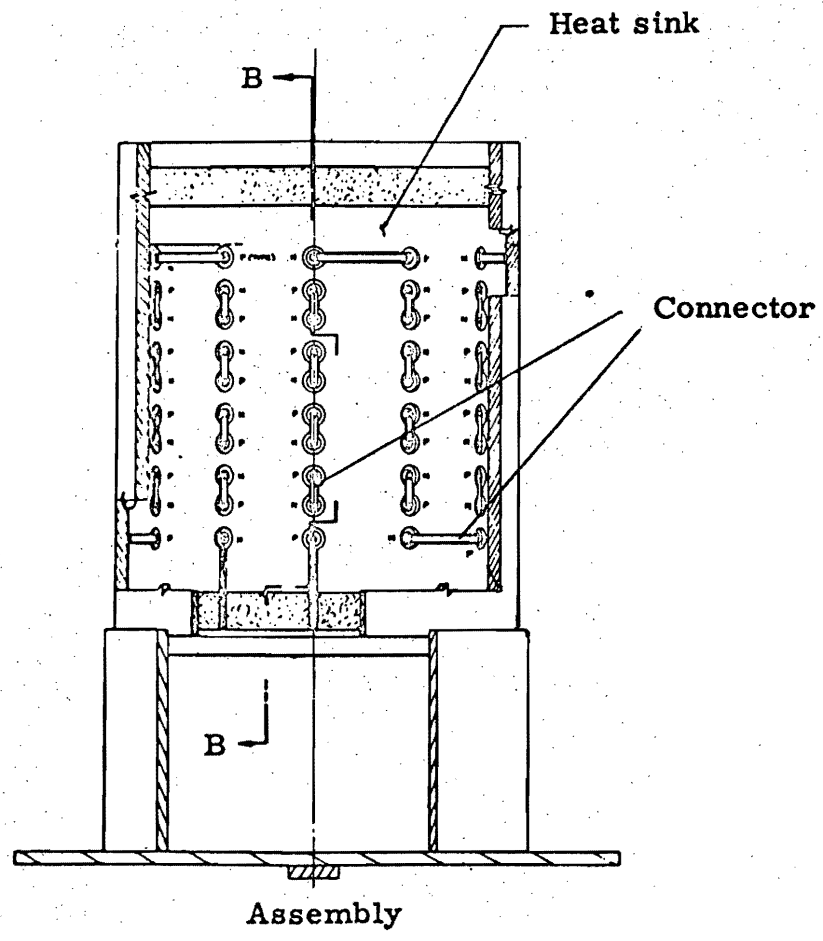
VI. POWER SOURCES

The heat generated by radioisotopes during decay may be partially converted to electricity and thus used as a power source. Such thermoelectric generators are suitable for both terrestrial use in remote areas and space use.[1] The generators for remote-area terrestrial use are discussed here, which, in the present state of development, are based largely on fission-product radioisotopes. Most of the current space applications, SNAP 1A being an exception, use alpha-emitting isotopes[2] and are not discussed here. Properties of radioisotopes that are of interest for making power sources are reviewed in references 3 and 4. Continuing work on fission products is reported regularly.[5]

RADIOISOTOPE POWER SOURCES

In 1957, Hittman[6] presented availability and cost studies that showed the feasibility of specialized low-power-output applications for radioisotopes. Subsequent research carried out under contract with the Division of Isotopes Development in cooperation with other Government agencies has resulted in development of radioisotopes-fueled generators.

A 5-watt thermoelectric generator (Fig. VI-1) fueled with 17,500 curies of Sr^{90} and an automatic nuclear-powered meteorological data-transmitting radio station for unattended service were designed, manufactured, and tested by the Martin Company for the Division of Isotopes Development and the U.S. Weather Bureau. After extensive testing, the system was installed on Axel Heiberg Island in the Arctic in 1961, and is still operating satisfactorily.[7, 8] The system automatically measures air temperature, barometric pressure, wind direction and two wind-speed averages. Once every 3 hr it transmits the data in binary digital form to a manned receiving station, where it is recorded on a two-channel



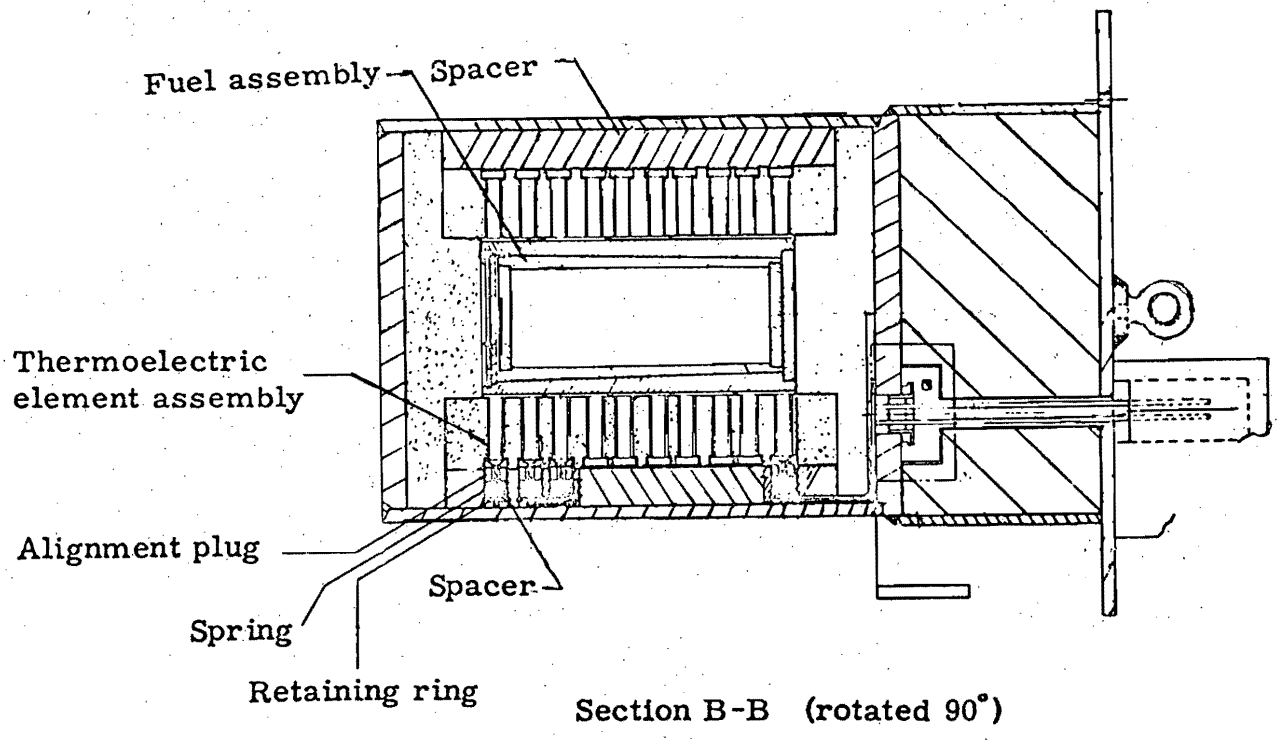


Fig. IV-1. **Strontium-90 Generator Assembly**

recorder. Temperature, wind direction, and barometric pressure are read at the instant of transmission. Wind speed is averaged over an 8- and a 1-min interval immediately before transmission. The generator and data telemetry package are in an insulated cavity 8 ft below the ground surface. Although the permafrost temperature varies from -10 to +30°F, the 95 watts of waste heat manufactured keeps the cavity between 40 and 80°F.

The strontium is in the form of strontium titanate, whose solubility in 0.1 N acid and base at 120°F in 750 hr was shown to be less than 5 ppm. Tests in both fresh and sea water indicated that more than 900 years would be required to dissolve the pellets tested. Strontium titanate melts at 1910°C,[8a] while 600°C is the maximum expected fuel temperature in a $Sr^{90}TiO_3$ thermoelectric generator. The Hastelloy C capsule used for the fuel is not pitted in sea water and is corroded at a rate of 0.0001 in./year.

A Sr^{90} titanate-fueled generator (SNAP 7A, Fig. VI-2) was also developed for a U.S. Coast Guard light buoy.[9] The generator supplies a nominal 10 watts of electric power at the end of the 10-year design life with an operating voltage of about 5 volts and is designed for operation without maintenance or attendance for 2 years. The fuel is encapsulated in Hastelloy C and is shielded with depleted uranium. The SNAP 7D 60-watt generator [10] is also fueled with Sr^{90} titanate, which is sealed in Hastelloy C. It is designed to provide electrical power for the U.S. Navy floating weather station in the Gulf of Mexico without attendance or maintenance for 2 years.

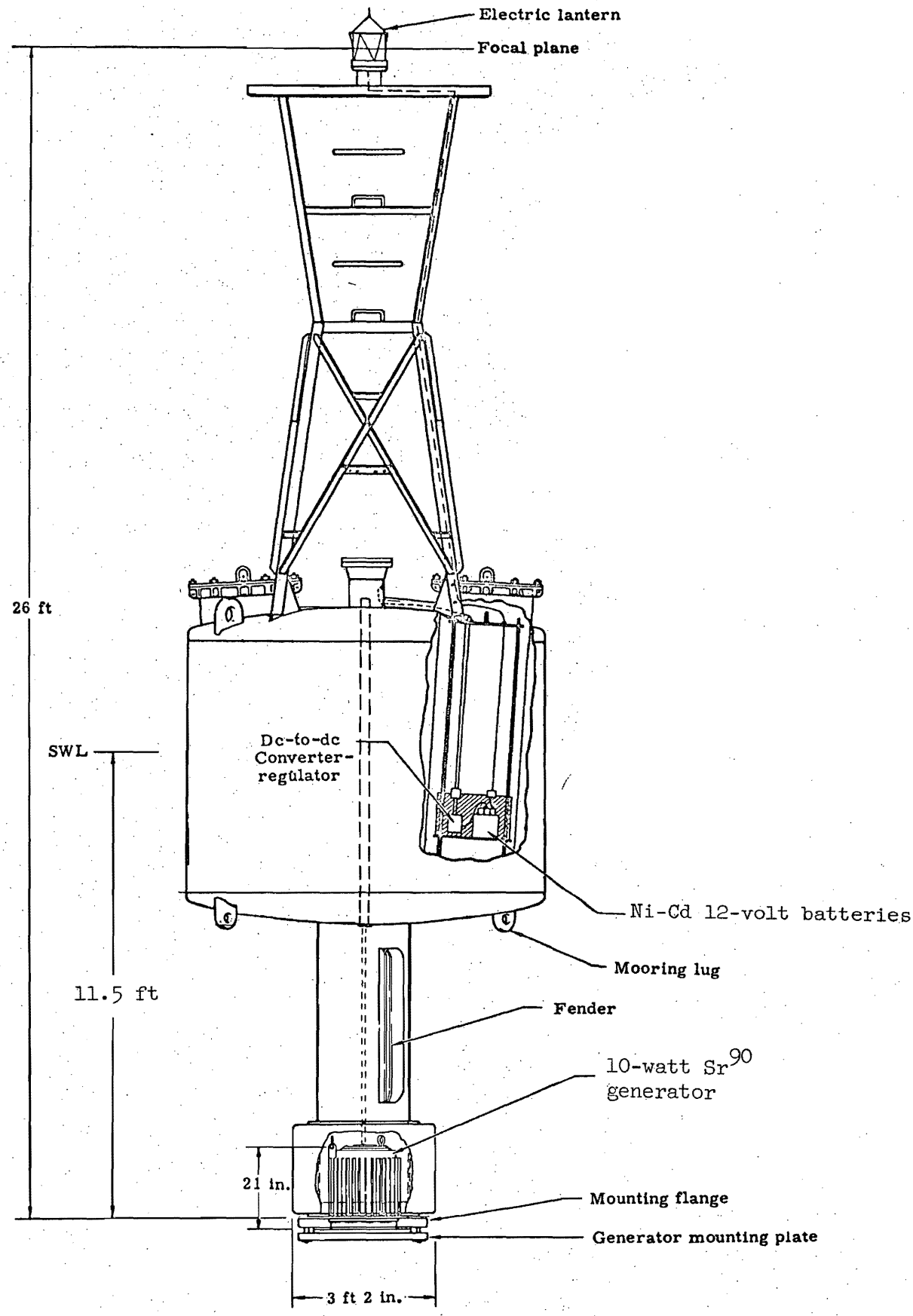


Fig. VI-2. Sr-90 Fueled Thermoelectric Generator System Mounted in Standard U. S. Coast Guard Flashing Light Buoy

The Chance-Vought Corp established both the economic and technical feasibility of a radioisotope-powered heat source for water desalination (Project Artesia).[11] A laboratory test model source capable of producing 250 gal of water per day was designed, in which 1 Mcurie of Ce^{144} and a 6-stage flash evaporator are used. The most economical source is considered to be the calcined gross fission product (CGFP) waste from processing of spent reactor fuel elements. At a distance of 400 miles from the fuel processing plant, such fuel would cost $0.55 per million Btu, which is the same as the cost for conventional fuels used to produce steam. The use of various separated fission products was also investigated.

Cs^{137} as a power source has been investigated by the Royal Research Corp., with special emphasis on underwater applications.[12] A complex borosilicate glass, containing 43 wt % cesium, was found to have a very low solubility and to be suitable as the heat source. After numerous safety tests, a prototype thermoelectric generator was designed to convert the decay heat from 27,000 curies of Cs^{137} (124 thermal watts) to 5 watts of electricity (Fig. VI-3). On the basis of this preliminary work, Royal Research Corp. and ORNL fabricated a source for an underwater seismograph device.

CALORIMETRY FOR MEASURING RADIOACTIVITY

The use of calorimetry at the Isotopes Division of ORNL was started as a part of the program to produce large radioactive heat sources. A calorimetric method was desired since it would give direct measurements of the rates of heat production and thus would eliminate errors associated with uncertain knowledge of the isotope decay schemes. It is the only

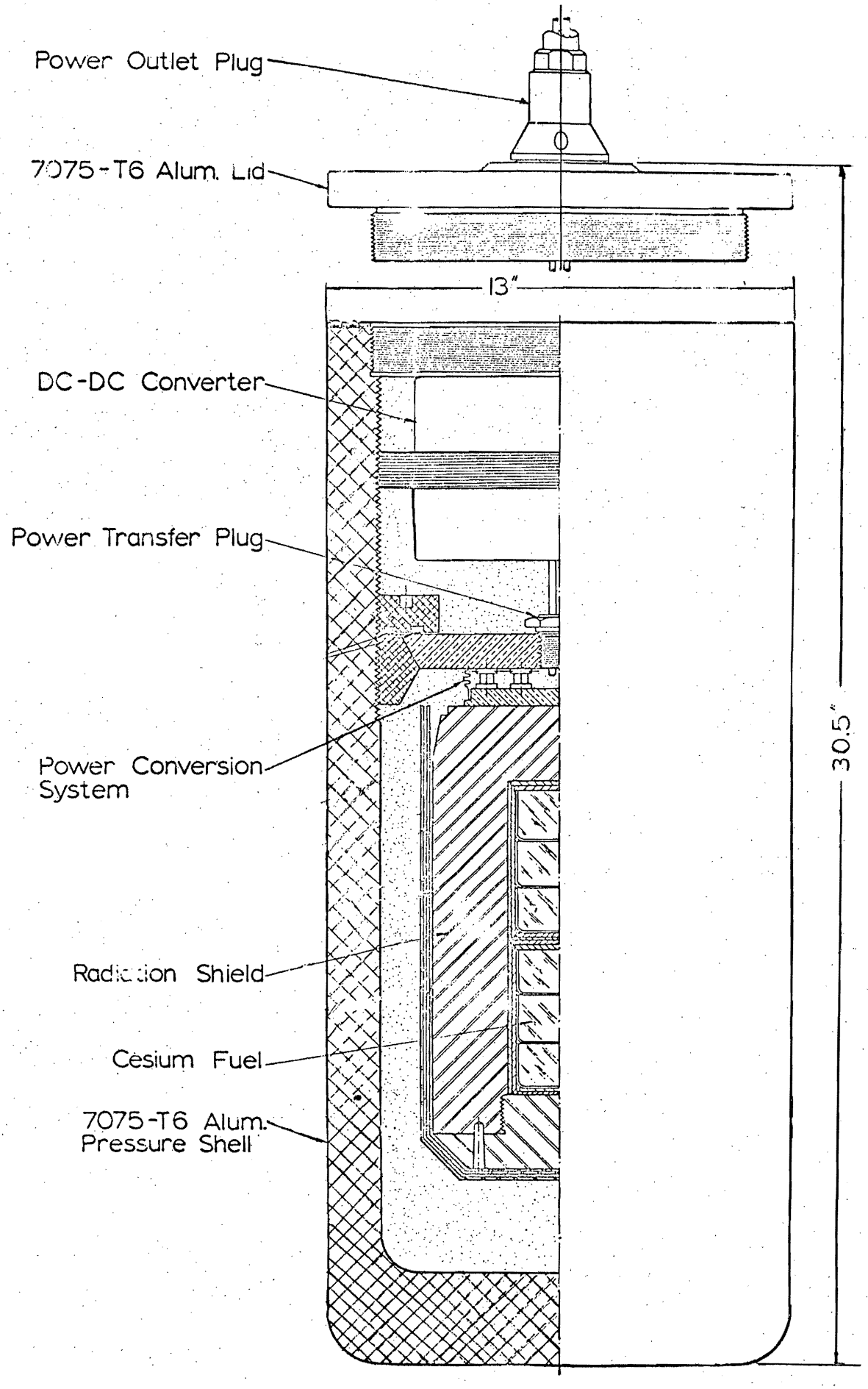


Fig. VI-3. CESIUM-137 POWERED 5-WATT THERMOELECTRIC GENERATOR

feasible method of measuring directly the radioactivity of large samples that produce only alpha or beta radiation. The dependability of calorimetric methods and their generally greater accuracy than competing counting procedures has led to their increasing use.

Calorimetry as applied to radioactive sources differs in one basic respect from conventional calorimetry: the quantity measured is a power or rate of heat production and is measured in such units as watts or calories per second while in conventional calorimetry a quantity of heat is measured in units such as watt-hours or calories. Because of this fact, most of the many commercial calorimeters cannot be used for the assay of radioactive sources.

A number of calorimeters for assaying the power of radioactive sources have been described in the literature,[13, 14] since the first was described by Rutherford and Barnes in 1904.[15] These calorimeters vary greatly in detail. However, the operation is in all cases based on one of the following measurements:

1. Rate of temperature increase of a mass of material of known heat capacity
2. Temperature gradient produced by the conduction of heat through a path of known properties
3. Temperature increase of a constant stream of cooling fluid
4. Rate of vaporization of a liquid

Most of the calorimetric work in the ORNL Isotopes Development Center has been done with simple calorimeters developed here. These are of the second type mentioned above. They occupy a minimum of hot cell space and are easy to operate, resistant to damage, and low in cost.

The basic parts of these calorimeters is shown in Fig. VI-4. The upper part is a metal cup mounted on a metal shaft, at the base of which is a heat sink cooled by a stream of water at constant temperature. The heat produced by radioactive decay of the source is absorbed by the cup and conducted down the shaft to the sink. At steady state, the temperature difference between the top and the base of the shaft is essentially proportional to the rate at which heat is produced by the radioactive source. This temperature difference is measured by a thermopile with junctions attached to the surface of the shaft. Copper lead wires are attached to the thermopile at the hot end of the shaft. Extraneous heat loss is minimized by enclosing the cup and the shaft in an inverted Dewar flask.

The materials of construction were selected to maintain nearly exact proportionality between the thermopile output and the thermal power of the source. Iron-constantan couples were chosen for the thermopile because their sensitivity (mv/°C) varies only slightly within the range of temperatures measured. Copper was selected for the shaft because its thermal conductivity is relatively constant. The departures from constancy of thermopile sensitivity and thermal conductivity that do occur are opposite in effect to the small loss of heat through the Dewar flask.

The calorimeters are calibrated before use by means of an electrical heater placed in the cup in substitution for the radioactive source. The voltage drop across the heater, the heater current, and the thermopile output are measured with precision electrical equipment of commercial manufacture. These electrical measurements during calibration and those during use are the only instances during which a high degree of precision

Table VI-1 CALIBRATION DATA OBTAINED WITH CALORIMETER SHOWN IN UPPER LEFT HAND CORNER OF FIG. IV-5

Thermal Power, watts	Thermopile Output, mv	Calibration Constant, watts/mv
24.18	6.594	3.667
24.32	626	3.671
18.38	005	3.673
18.08	029	3.669
12.27	339	3.674
12.32	354	.675
.17	682	676
		(avg.)

the thermal powers of gamma-emitting samples can be determined in approximately the same time (1.5-2 hr) as that required with calorimeters designed for use with alpha- and beta-emitting materials. Some uncertainty is added because of differing degrees of self-absorption of gamma radiation by the samples themselves.

The last calorimeter, was designed for use with small samples of alpha- or beta-emitting materials held in a standard capsule used for irradiation of samples in the hydraulic tubes or the Oak Ridge Research Reactor. Measurements are accurate to 0.005 watt in the range 1-10 watts. A similar calorimeter with thick uranium walls has been constructed for gamma-ray absorption. It has been used primarily for the assay of small Ir^{192} samples.

A calorimeter for assaying alpha-emitting samples of 30-300 watts is being constructed. It will extend the range of sample power that can be measured. Since the shaft has a much larger gross thermal conductivity than that of any of the calorimeters previously constructed, it will be a test of the degree to which the time required to reach steady state can be reduced by this means.

Flow Calorimeters

Assay of sources in the form of long thin rods has sometimes been necessary. Calorimeters of the preceding type are not well adapted for sources of this shape. Two calorimeters, one for alpha- or beta-emitting sources and one for gamma-emitting sources, have been made. These calorimeters use the increase in temperature of a constant and accurately measured stream of water as a measure of the rate of heat production by the source.

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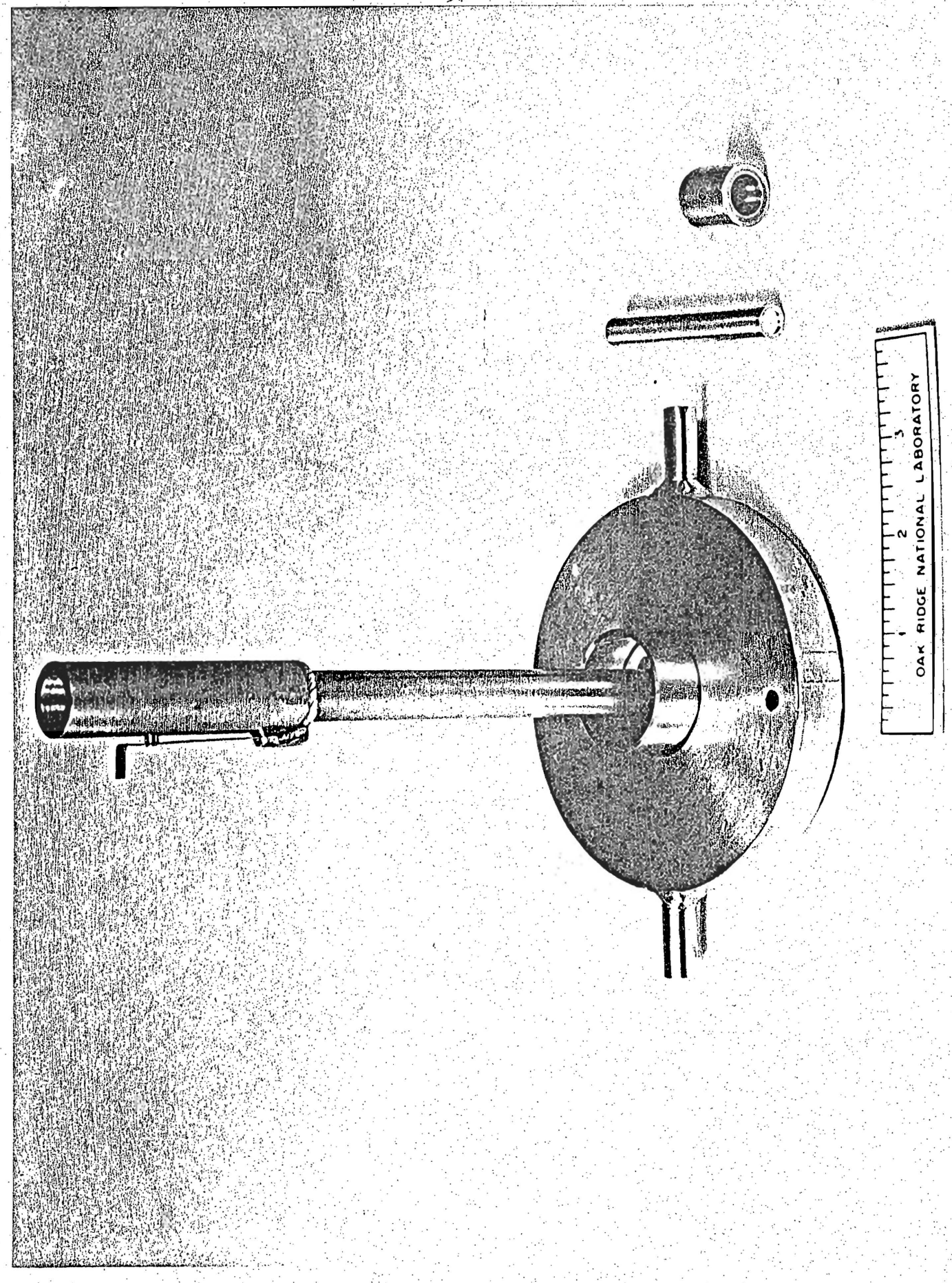

Fig. VI-4. Basic parts of calorimeter used for measuring rates of heat production by radioisotope sources.

must be attained. Comparatively loose tolerances are permissible in fabrication.

The design of a calorimeter of this type involves a suitable compromise between accuracy and speed of operation. The approach to steady state is described approximately by

$$\theta = \frac{C}{U} \ln \left(\frac{\Delta T_{ss}}{\Delta T_{ss} - \Delta T}\right)$$

where θ is the elapsed time, C is the heat capacity of the sample cup and upper part of the Dewar flask (The primary assumption is that these are at the same temperature), ΔT is the difference in temperature between the ends of the shaft, ΔT_{ss} is this temperature difference at steady state, and U is the gross thermal conductivity of the shaft. The time required for ΔT to reach a given fraction of ΔT_{22} is thus inversely proportional to the thermal conductivity of the shaft. However, ΔT_{ss} is inversely proportional to U. Consequently, since the error caused by fluctuations in cooling water temperature and by limits of precision of the measurement of the thermopile output are approximately inversely proportional to ΔT_{ss}, the increase in speed is accompanied by a loss of precision. Furthermore, if U is increased until the resistances to heat flow from the sample to the cup and from the cup to the Dewar flask are significant compared to the shaft resistance, the time required will not decline as rapidly as predicted by the equation.

Four of the calorimeters that have been constructed are shown in Fig. VI-5, minus the Dewar flasks and thermopiles. The first (upper left) was designed for measuring the thermal powers of Sr^{90} samples of 5-50 watts. The shaft ΔT is ~1.5°C per watt.

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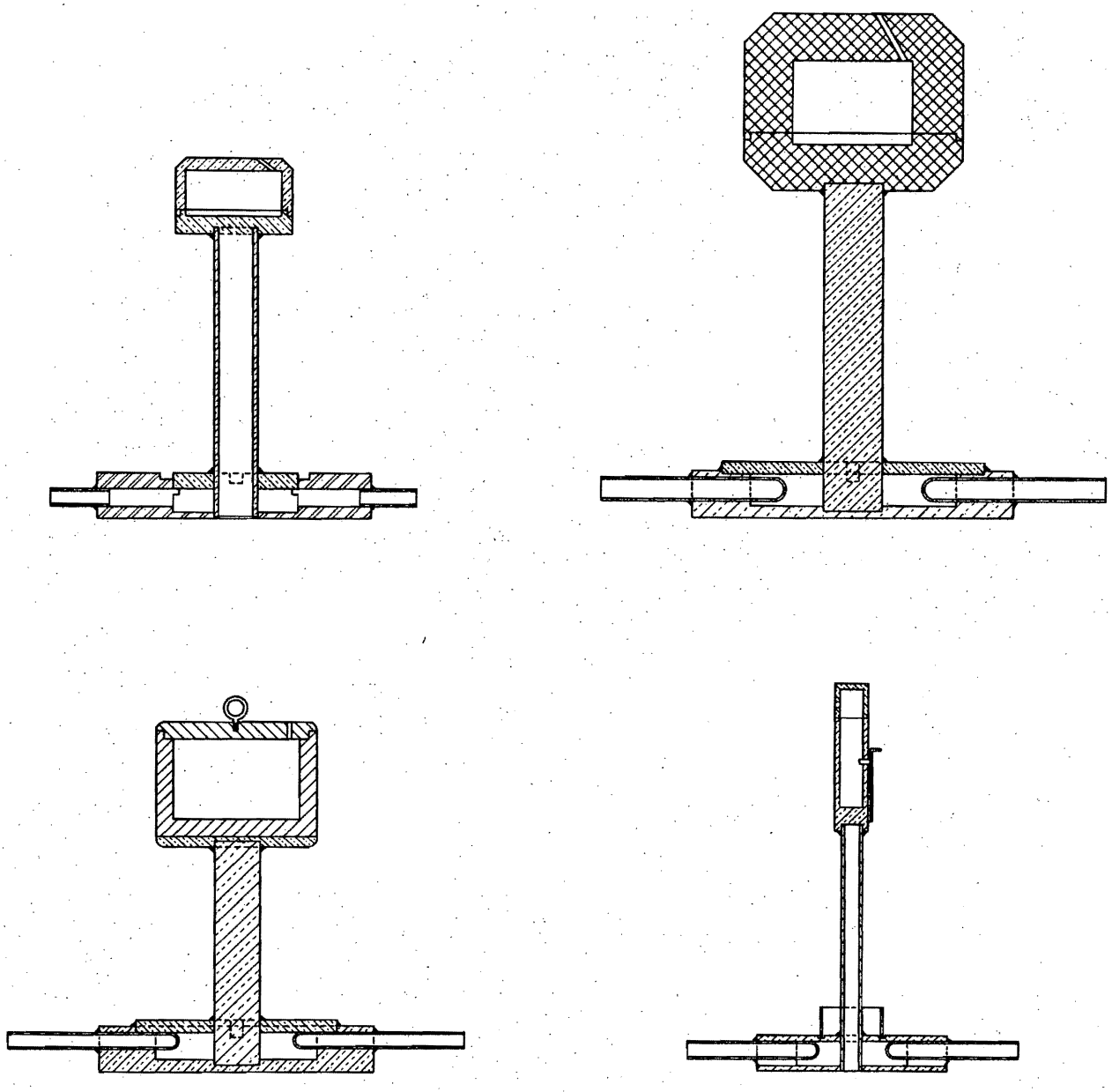

Fig. VI-[illegible]. Thermal Conductivity Calorimeters

The reproducibility of results obtained with this calorimeter are illustrated by the calibration data given in Table VI-1. No individual value of the calibration constant differs from the average by more than 0.13%. If deviations from the average are treated as random errors, the 95% confidence limit of a single determination is 0.2%.

The performance of the calorimeter is only moderately sensitive to the rate of flow or to the temperature of the cooling water provided both are constant during any given determination. Variation of the flow rate from 1.1 to 8 liters/min produced a change of only 0.4% in the calibration constant. A change in the cooling water temperature from 10 to 27°C lowered the constant by 1.4%. A lowering of 1.56% was calculated from the known change of the iron-constantan thermopile and the copper shaft with temperature. The agreement is within experimental error.

The second calorimeter (upper right) was designed for use with Cs^{137} samples of 5-50 watts. The cup is constructed of metallic uranium and has walls 1-1/16 in. thick. The gamma ray absorption is nearly complete. More than 99.8% of the total energy is absorbed.

Because of the heat capacity of the thick-walled cup a much longer time (7-8 hr) is required for this calorimeter to reach steady state than is required for calorimeters designed for use with only alpha- or beta-emitting materials. The results are no less accurate, however, and results are reproducible within 0.2%.

The third calorimeter (lower left) is designed to allow more rapid measurements of gamma-emitting sources. The thinner walls absorb a known fraction of the radiation (94.85% of the total energy from Cs^{137}). Thus,

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12.32	3.354	3.675
6.17	1.682	3.676
		3.672 (avg.)

the thermal powers of gamma-emitting samples can be determined in approximately the same time (1.5-2 hr) as that required with calorimeters designed for use with alpha- and beta-emitting materials. Some uncertainty is added because of differing degrees of self-absorption of gamma radiation by the samples themselves.

The last calorimeter, was designed for use with small samples of alpha- or beta-emitting materials held in a standard capsule used for irradiation of samples in the hydraulic tubes or the Oak Ridge Research Reactor. Measurements are accurate to 0.005 watt in the range 1-10 watts. A similar calorimeter with thick uranium walls has been constructed for gamma-ray absorption. It has been used primarily for the assay of small Ir^{192} samples.

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Applications of Calorimetry

Calorimetric methods are used for the final check of the magnitude of the heat output of the radioactive heat sources produced by the Isotopes Development Center and are for control analysis during manufacture of the materials of these sources.[16] Calorimetry also provides a convenient measurement of the degree of conversion, by (n,γ) reaction, of samples exposed in atomic reactors. The samples are removed from the reactor, and the thermal power is measured without removing the sample from its container. This method has been used notably in the curium development program.

One notable project carried out in support of the above work was a decay study of an irradiated Am^{241} sample. Although the thermal power of this sample was largely produced by the Cm^{242} product, significant fractions were produced by fission products and by long-lived transuranic impurities. The quantities of these other radioactive materials present in samples were commonly calculated, and it was desired to check the accuracy of the methods of calculation.

A decay study was carried out over a period of 65 days (Fig. VI-6). The total or measured powers were corrected by the normal method for the contribution of fission product decay. Since the decline of the corrected power with time could be attributed within experimental error entirely to the decay of Cm^{242} to Pu^{238}, these values could be correlated with time by a least mean squares equation, using literature values for the half-lives and energies of decay of Cm^{242} and Pu^{238}. Agreement of this derived equation with the data was excellent; the standard deviation of the points from the equation was only 0.0023 watt, or 0.06% of the average power of the sample.

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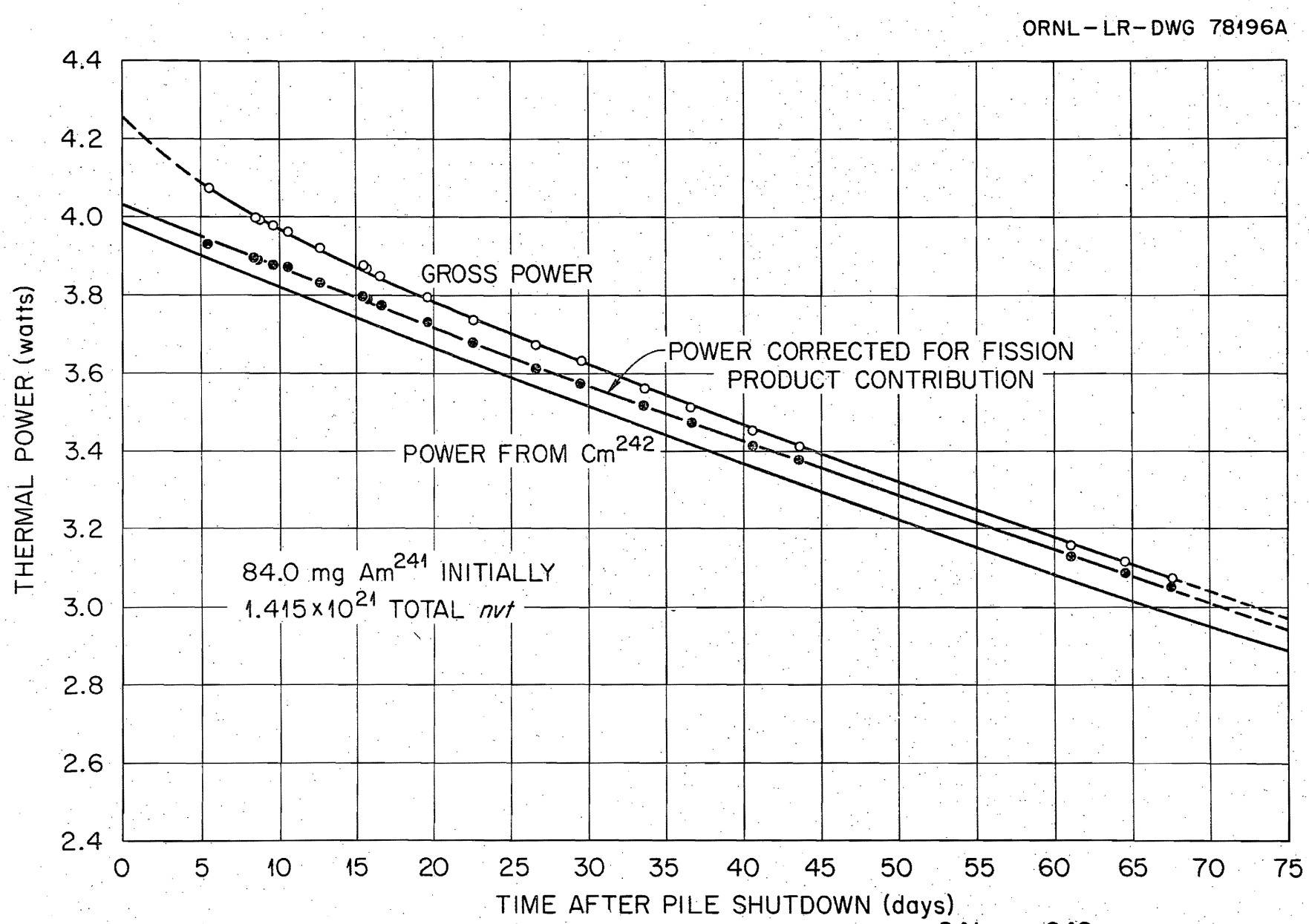

Fig. VI-6. Calorimetric Decay Study of Irradiated Am^{241} (Cm^{242}).

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VII. APPLICATIONS IN MEDICINE

The literature on medical uses of radioisotopes is briefly reviewed here.* Since developments in this rapidly expanding field have been summarized in various well-documented publications,[1] earlier original literature was not extensively consulted. Only a few of the many published papers are listed here, and emphasis is on recent, rather than older, work. Since many reports are preliminary and opinions on the effectiveness of some of the isotopes differ widely, in future issues of this quarterly various viewpoints on the medical use of radioisotopes will be presented, and some of the less well-known isotopes, both old and new, will be reviewed. Readers with publications in this field are invited to send in reprints to ensure coverage.

The medical use of artificial radioactive isotopes began in the late thirties.[2] The increasing interest in them is indicated by the numerous conferences,[3] journals,[4] bibliographies,[5] and reviews[6] partly or entirely devoted to this field. Radioisotopes are used in both diagnosis and therapy (Table VII-1[7]), in the former in extremely small amounts; in the latter, chiefly in malignant or malignancy-related conditions. Safety in their use is ensured (1) by requiring the licensing of physicians who use them -- and then only after special training; (2) by standardization of handling techniques and adoption of special safety regulations by hospitals.[8] Early fears of long-term adverse

*The editors of this review gratefully acknowledge the many helpful suggestions made by W. G. Myers, M.D., and M. Brucer, M.D., who reviewed a rough draft of the manuscript. Some of their suggestions that could not be readily incorporated will be used in future issues.

Table VII-1. Radioisotopes Used in Diagnosis and Therapy*

Diagnosis		
Test	Measurement	Usefulness
I^{131} studies of thyroid function; NaI, usually oral	Percentage of dose concentrated in thyroid, percentage of dose excreted in urine; plasma free and bound I^{131}	Valuable index of thyroid function; drugs cause interference
I^{131} for diagnosis of thyroid carcinoma; oral	Scanning and external procedures	Relatively little value while normal thyroid gland present; highly useful in locating well differentiated metastatic lesions after thyroidectomy
Fe^{59} iron metabolism; usually intravenous iron citrate; oral doses used to study absorption	Serial plasma assays for several hours; red cell assays for about 10 days; external counts over spleen, liver, sacrum; blood and fecal assays after oral dose	Helps explain mechanism of some anemias, importance of impaired erythropoiesis versus hemolysis; oral test indicates absorption defects.
Cr^{51} red cell survival; red cells labeled with Cr^{51} in vitro and injected intravenously	Serial assays of blood (red cells) for 2 or 3 weeks; external counts over spleen, liver, heart; fecal assays for gastrointestinal bleeding	Indicates rate of hemolysis and to some extent the role of the spleen in hemolysis
Co^{60}-vitamin B_{12}; oral	Fecal and urinary excretion; sometimes external counts over liver	Shows fundamental defect of pernicious anemia even if patient is not in hematologic relapse
I^{131}-labeled rose bengal; intravenous	Rates of removal from bloodstream and concentration in liver	An index of liver function and biliary patency

Table VII-1. Contd.

Test	Measurement	Usefulness
I^{131}-labeled fat; oral	Blood, urinary, and fecal levels	Information on intestinal absorption, pancreatic function
Cr^{51}, P^{32} red cell mass; I^{131} albumin-plasma volume, Na^{24}, K^{42} for Na and K spaces	All based on dilution principle; solution of isotope (or suspension of red cells tagged in vitro) given intravenously and after mixing in the body, blood concentration determined	These tests give data on red cell and plasma volumes, fluid balance and electrolyte status; may be helpful in presurgical evaluation, study of metabolic and endocrine disorders.

Therapy

	Patients Treated	Usefulness
I^{131} as NaI; usually oral	Uncomplicated Graves' disease patients over 40; complicated Graves' disease any age	General results excellent; multiple doses needed in some cases; significant incidence of early or delayed myxedema
	Toxic nodular goiter in selected patients	Eventual control of toxicity in most cases; may require repeated doses; concern over malignancy already present at onset, plus desire for rapid result, may recommend surgery for patients who can be well prepared

Table VII-1. Contd.

	Patients Treated	Usefulness
	Thyroid carcinoma in selected patients	For treatment of relatively small group of patients with nonresectable lesions showing ability to concentrate the isotope
	Certain patients with angina pectoris or intractable congestive heart failure	Clinical improvement in significant percentage
P^{32} as soluble phosphate; oral or intravenous	Chronic granulocytic leukemia	Generally good results
	Chronic lymphocytic leukemia	Useful, often in conjunction with local irradiation
	Polycythemia vera	Good results in patients not suitable for phlebotomy alone
	Lymphosarcoma, multiple myeloma, diffuse bone metastases	Occasionally some value
	Acute leukemia, Hodgkin's disease	Rarely useful
Colloidal Au^{198}, P^{32}, Y^{90}	Intracavitary in patients with effusions and ascites caused by malignant tumors	Symptomatic value; partially supplanted by nitrogen mustards
	Intravenous for granulocytic leukemia and diffuse lymphoma of liver	Very limited value compared with other forms of therapy

Table VII-1. Contd.

	Patients Treated	Usefulness
	Direct injection into tumor in certain cases of prostate carcinoma; attempt to radiate lymphatic channels	Believed useful if properly used in carefully selected cases
Co^{60}, Cs^{137}, radium-226, radon-222, Sr^{90} for implantation and brachytherapy	Certain localized tumors, especially carcinoma of cervix, head, and neck	Useful in skilled hands for properly selected patients
Co^{60}, Cs^{137} for teletherapy	For all lesions suitable for external radiation therapy	Highly useful and practical

*Reprinted with minor style changes with the kind permission of publishers, from G. A. Andrews, Medical students and radioisotopes, The New Physician, 9(12):23-7 (1960).

effects on patients have been tempered by the results of experience. According to Lawrence,[9] for example, cancer has not once been reported as a result of radioactive iodine therapy of thousands of hyperthyroid patients; and in 25 years, the survival rate of more than 500 polycythemia vera patients treated with P^{32} has been about the same as that of patients with diabetes or pernicious anemia treated by standard methods. There is wide usage of radioactive iodine for toxic goiter in patients over 45, of P^{32} for polycythemia vera, and of various isotopes for pituitary or thyroid ablations to relieve other conditions.[1]

Among the radioisotopes now used in internal medicine in the largest amounts are I^{131}, P^{32}, Au^{198}, Cr^{51}, Fe^{59}, and Hg^{203}, but many others are used in smaller amounts; large quantities of Co^{60} and Cs^{137} are used in teletherapy units. Many radioisotopes are being studied for potential use, and useful ones are introduced into the field as new production methods are developed and costs are lowered. Criteria for suitable isotopes include high specific activity, suitable half-life, physiological compatibility with body processes, and the ability to at least partially localize in the tissue being examined or treated. Radioiodine, for example, while not entirely thyroid-specific, is an obvious choice for thyroid conditions, as is calcium for bone. Specificity may sometimes be obtained by incorporation in compounds (e.g., radiocobalt in vitamin B_{12}, which localizes in the liver). An isotope may be localized physically as a wire, as a colloid, or as the recently introduced ceramic beads.[10] Both the physiological and biological half-life must be considered, and selection may be made on the basis of the need for particular radiation characteristics -- from a soft beta all

the way to a hard gamma radiation. In earlier work, isotopes with a short physical half-life were found inconvenient to use,[11] but they are finding application now with development of new instrumentation and sources of supply.

Iodine

I^{131} (8.1 d, 0.33- and 0.61-Mev β's, 0.36- and 0.64-Mev γ's). Iodine-131 is most commonly used for diagnosis of thyroid disorders.[1] Other uses include incorporation in human serum albumin for determination of the total circulating plasma volume, placentography,[12] estimation of mitral valve size,[13] and detection of right-to-left cardiac shunts. There has been partial success in localizing brain tumors with I^{131}-labeled compounds, and I^{131}-labeled methyl iodide has been used for detecting left-to-right cardiac shunts.[14] Rose Bengal containing I^{131} is useful for liver function tests and for blood flow tests[15] and has been suggested for detecting liver metastases that are not obvious from surgery.[16] Recent reports indicate the Rose Bengal-I^{131} hepatoscan to be a satisfactory technique.[17] Other I^{131}-labeled compounds reported useful are Hippuran for renal tests[18] and for determining cerebral circulation time;[19] Diodrast for evaluating venous diseases of the lower extremities;[20] sulfobromophthalein for liver function tests;[21] and compounds for studying fat metabolism.[22] An I^{131}-labeled compound demonstrated 200-ml pericardial effusions in a simulated system,[23] and an accurate and reproducible method for insulin assay with I^{131} has been reported.[24]

In therapy, I^{131} has been used for nontoxic[25] as well as toxic goiter.[1] It is used as a palliative in thyroid carcinoma, and an

I^{131}-tagged fat-soluble medium, which accumulates in regional lymph nodes, is potentially useful for irradiation of neoplasms.[26] Thyroid ablation by I^{131} in more than 1000 patients improved angina pectoris in 75% and congestive heart failure in 60% of those afflicted.[27] Fortunately, gamma irradiation of the ovaries from urinary excretion of administered I^{131} is considered negligible.[28]

I^{125} (57.4 d, no β, 0.035-Mev γ). Iodine-125*, which is easily produced by neutron bombardment of Xe^{124}, is becoming increasingly valuable in diagnosis. Irradiation of a patient by it is about half[29] that from I^{131}, assuming that the biological half time is equal to the physical half-life. Good thyroid and liver scans have been made with it,[30] and Hippuran labeled with I^{125} has been used for renograms.[31] As a tag for serum albumin and oleic acid and in the salt NaI^{125}, it has given good results in experimental blood volume, absorption, and thyroid tests,[32] and has been suggested for indicating the extent of arteriosclerosis.[33] In a promising new technique for lipid digestion and absorption studies, a compound labeled with I^{131} and another labeled with I^{125} are administered simultaneously.[34] The promising use of I^{125} in radiography[35] is indicated by a photograph of the bones of a hand made with this isotope (Fig. R3, R4). Exposure to both patient and technician is low because of the ease of shielding against the 27.2 and 35.4-kev photons. Portable x-ray equipment is being developed with I^{125} sources.[36]

I^{132} (2.3 hr, 1.5- to 2.1-Mev β's, 0.53- to 0.95-Mev γ's). The use of I^{132}, which is easily recovered from a tellurium source, would decrease patient exposure to 1-2% of that from I^{131} and would

*See also p. 245.

allow repeated administration.[37] Thyroid function after I^{131} therapy may be determined by observing the absorption of I^{132}, whose higher energy gammas are easily detected in the presence of residual I^{131}.[38]

I^{123} (13.3 hr, no β, 0.16-Mev γ). With I^{123}, exposure of a patient would be only 1-5% of that from I^{131}. Experiments on preparation of curie amounts are projected.[39]

I^{124} (4 d, 1.53- and 2.13-Mev β's, 0.6- and 1.72-Mev γ's). Iodine-124 has been suggested for greater uniformity of dose in thyroid carcinoma.[40]

Phosphorus

P^{32} (14.3 d, 1.71-Mev β, no γ). The largest use of P^{32} is in treatment of polycythemia vera and chronic leukemia.[1] It has not been very useful in acute leukemia. Blotting paper soaked with radioactive sodium phosphate was used in early applications for skin lesions, and activated red phosphorus incorporated in polyethylene in later work. Metastatic bone cancer has been treated with P^{32} as the phosphate.[41]

Colloidal chromic phosphate-P^{32} and Au^{198}, as well as nitrogen mustard, for control of malignant intracavity effusions are compared in a recent review[42] of 222 cases treated with P^{32} and 2110 with Au^{198}. Colloidal chromic phosphate-P^{32} has been used for palliation of cancer, and colloidal zirconyl phosphate-P^{32} administered intravenously to more than 250 patients with chronic leukemia and lymphomas is reported to have resulted in prolonged remissions and long treatment-free periods.[43]

In diagnostic applications, an in vivo test for human spermatogenesis using sodium phosphate-P^{32} has been described,[44] and the ability of newly proliferating cells to concentrate P^{32} has been used to

differentiate benign from malignant ocular lesions.[45]

Gold

Au^{198} (2.7 d, 0.97-Mev β, 0.41-Mev γ). Gold-198 is most commonly used as a colloid,[1] which has 0.003- to 0.007-μ-dia. particles. When this material is administered to a patient and a scan is performed, normal liver shows an even gradation of activity, but lesions result in activity concentrations.[46] Diagnostic uses include determination of liver blood flow,[15] and in regions where the parasitic Taenia *Echinococcus granulosus* occurs it has been used for localizing hydatid liver cysts and for following up healing after drug administration or surgery.[47]

Colloidal gold has been used in treating both primary carcinoma and metastases.[48] Transportation to the lymph nodes is hastened if the particles are coated with silver,[49] but radioactive colloidal gold has been injected directly into the lymphatics.[50] Other uses are in treating leukemia[51] and in controlling malignancy-induced intracavity fluids.[52]

Interstitial implants of Au^{198} seeds have been used for hypophysectomy and for palliation of metastatic carcinoma.[53] The hypophysis implantation requires about 30 min. Problems in surgical and radiotherapeutic pituitary ablation are discussed in recent reviews.[54]

Chromium

Cr^{51} (28 d, no β, 0.32-Mev γ). Hexapositive Cr^{51}, in $Na_2Cr^{51}O_4$, attaches to the red cells and may be used to determine such quantities as red cell survival time.[1] A new instrument for accurate blood volume measurement uses Cr^{51}-tagged red cells or I^{131}-albumin.[55] Such cells have been suggested for determination of cardiac output and for placental

localization (no activity is transferred to the fetal circulation), and are the basis for a simple method for quantitation of menstrual blood loss[56] and for spleen studies.[57] Tripositive chromium (as $Cr^{51}Cl_3$) in solution attaches to plasma proteins and is useful for determining plasma volume. Serum albumin labeled with Cr^{51} is easily prepared and has been introduced for demonstrating gastrointestinal protein loss.[58]

Implants of Cr^{51} have been recommended for cancer therapy.[59]

Iron

Fe^{59} (45 d, 0.26- and 0.46-Mev β, 1.1- and 1.3-Mev γ). Detailed methods for Fe^{59} tests are described in a recent brochure.[60] As the chloride, citrate, or sulfate, Fe^{59} has been used for iron absorption and utilization studies.[61] It has also been used for determining red cell survival time, but the necessity of labeling the cells in vitro makes it less convenient than Cr^{51}. The advantages and disadvantages of Fe^{59} and Cr^{51} are discussed in a recent review.[62]

Fe^{55} (2.94 y, 0.0059-Mev x ray following electron capture). Comparison of the percentage utilization of intravenously administered Fe^{59} to that of oral Fe^{55} has been used for iron absorption studies.[63]

Mercury

Hg^{203} (47 d, 0.21-Mev β, 0.28-Mev γ). Besides the use of mercury isotopes in tracing kidney drugs,[64] Hg^{203} has been used for localizing brain tumors and for determining cerebral circulation time.[65] Neohydrin and Chlormerodrin labeled with Hg^{203} have been used for kidney examinations.[66]

Boron

B^{10}. When the stable isotope B^{10} is deposited in brain tumor tissue and activated by neutron bombardment, the released alpha radiation is effective for control and palliation.[67] By this treatment, cells of transplantable sarcomas in mice have been killed and cancer has disappeared in 2-4 weeks.[68] This method of treatment is not new, but basic techniques are still being investigated.[69]

Fluorine

F^{18} (1.87 hr, 0.65-Mev positron emission). Fluorine-18 has been used as a replacement for Ca^{47} and Sr^{85} in bone scanning.[70] It has also found application in thyroid studies[71] and in locating brain tumors.[72]

Selenium

Se^{75} (120 d, no β, 0.14- to 0.28-Mev γ's). The high specificity of Se^{75}-selenomethionine for the pancreas suggests that this compound might be useful in early detection of pancreas carcinoma.[73]

Krypton

Kr^{85} (10.6 y, 0.67-Mev β). Because of its low solubility, injected or inhaled Kr^{85} is useful in determining cerebral and coronary blood flow and cardiac and pulmonary shunts.[74] For example, if a right-to-left cardiac shunt is present, activity in the arterial system rises rapidly when Kr^{85} is given intravenously.[75] If a left-to-right shunt is present, the Kr^{85} concentration in the pulmonary artery reaches a value 20 times that in a systemic artery following inhalation of this gas.[76] A simple technique for studying pulmonary function with Kr^{85} has been devised,[77] and a new technique for rapid and accurate

measurements of serial heart blood flow was reported recently.[78]

Strontium and Yttrium

Sr^{90} (28 y, 0.54-Mev β, 2.27-Mev β from Y^{90} daughter, no γ). In an external source containing Sr^{90} designed for use in ophthalmology, the Sr^{90} beta particles are filtered out so that only the Y^{90} betas actually reach the lesion. The lens is not irradiated.[1]

"Radiation curettage," destruction of the uterine mucosa by radiation from a Sr^{90} source placed in the uterine cavity, has been recommended for control of endometrial bleeding when no malignancy is indicated.[79]

Sr^{85} (65 d, no β, 0.5-Mev γ). Strontium-85 has been used for localizing bone tumors[80] and for studying bone metabolism. Because of its shorter half-life, irradiation of the body is less than with Sr^{90}. Its gamma radiation can be detected externally.

Sr^{87m} (2.8 hr, 0.39-Mev γ). This isotope, which can be obtained quickly, as needed, from 80-hr Y^{87}, has been mentioned[81] for possible use in obtaining information available in a short time; for example, circulation time, cardiac output, and various clearances, and in radiography.

Y^{90} (64.5 hr, 2.27-Mev β). Injected ceramic Y^{90} particles of ~60 μ dia., which are trapped in capillaries of the lung, liver, and other organs, have been used for cancer therapy.[82] Thirty-five patients with Parkinson's disease were improved by brain implants of ceramic Y^{90} beads.[83] Carrier-free Y^{90} may be eluted[84] with citrate from a cation exchange resin preparation of Sr^{90}. It may be infiltrated as a solution, implanted in a plastic filament, or applied in a bag or absorbed in

blotting paper.[85]

Because it is a pure beta emitter, colloidal Y^{90} results in less exposure of hospital personnel than Au^{198}, and it has been used for control of malignancy-induced effusions.[86] Complete pituitary destruction has been reported as a result of packing the fossa, following surgery, with powdered Y^{90} dispersed in wax.[87] For superficial radiation therapy, Y^{90} has been incorporated in small plates, which can be selected to cover an irregular area.[88] Transperineal injection of Y^{90} has been used for prostatic cancer.[89] Intravenous Y^{90} gave good results in eight polycythemia vera patients.[90]

Calcium

Ca^{45} (164 d, 0.25-Mev β, no γ). Calcium-45 has been used for studying calcium metabolism, Paget's disease of bone, and hypoparathyroidism.[91]

Ca^{47} (4.7 d, 0.66-and 1.94-Mev β's, 0.48- to 1.3-Mev γ's; accompanied by 3.5 d Sc^{47} daughter with 0.46- and 0.62-Mev β's and 0.175-Mev γ). Possible medical uses of the now-available Ca^{47}, which is of interest because of its high gamma energy, are being extensively investigated.[92]

Cobalt

Co^{60} (5.3 y, 0.31-Mev β, 1.17- and 1.33-Mev γ). By far the largest use of Co^{60} is in teletherapy units,[1b,c,93] descriptions of which may be obtained from various equipment manufacturers (see also book review section of this issue). Such units were widely distributed by 1959 (Table VI-2).[94] In 1962 they were reported to number 1400, of which 550 are in the U.S.A. and Latin America.[95] They are reported[96]

Footnotes to Table VI-2.

[a]Reprinted, with minor style changes, from reference 94.

[b]These figures represent the number of licenses issued as of August 1, 1959; they do not include such teletherapy installations as those at Oak Ridge Institute of Nuclear Studies and Argonne Cancer Research Hospital, for which no license was required.

[c]Estimated, mainly low curie units.

[d]Including only countries for which information is available.

Table VI-2. Radioisotope Teletherapy Source Distribution

Location	Unit		
	Ir^{192}	Cs^{137}	Co^{60}
Africa			
Algeria	-	-	1
Union of South Africa	-	-	1
America			
Argentina	-	-	3
Brazil	-	-	8
Canada	-	2	24
Chile	-	-	2
Cuba	-	-	2
El Salvador	-	-	1
Mexico	-	-	12
Peru	-	-	1
Puerto Rico	-	-	2
United States	-	6[b]	264[b]
Uruguay	-	-	2
Venezuela	-	-	5
Asia			
Burma	-	-	1
Ceylon	-	-	1
China (Taiwan)	-	-	1
Hong Kong	-	-	1
India	-	-	3
Indonesia	-	-	1
Iran	-	-	1
Israel	-	-	3
Japan	-	-	200[c]
Lebanon	-	-	1
Philippines	-	-	4
Thailand	-	-	1
Europe			
Austria	-	-	2
Belgium	-	-	2
Denmark	-	-	2
Finland	-	-	1
France	-	1	40
Germany (Fed. Republic)	-	-	4
Greece	-	-	3
Hungary	-	-	1
Italy	-	-	26
Monaco	-	-	1
Netherlands	-	-	4
Norway	-	1	-
Portugal	-	-	1
Spain	-	-	5
Sweden	-	-	7
Switzerland	-	-	4
United Kingdom	1	8	33
Yugoslavia	-	-	1
Oceania			
Australia	-	-	4
New Zealand	-	-	2
Total[d]	1	18	689

See following page for footnotes.

to be in use in Communist China, and have been referred to as the most important product of Atomic Energy of Canada Ltd.[97] A nonroutine use of Co^{60} teletherapy is a possible new treatment for leukemia in which blood, circulated outside the body through a tube, is exposed to Co^{60} radiation.[98]

Cobalt-60 needles have been implanted in tumors, and seeds enclosed in nylon tubing have been used in interstitial therapy and in wounds after surgery for a malignant condition.[99] Spheres of Co^{60}, gold-plated to prevent dusting, may be placed in hollow organs such as the uterus or bladder.[100] Cobalt-60 may be incorporated in plastic and molded to fit irregular areas such as the oral cavity.[101]

Vitamin B_{12} labeled with Co^{60} has been used in diagnosing pernicious anemia[102] and localizing liver tumors.[103]

Other Cobalt Isotopes. Co^{56} (77.3 d, 0.44- and 1.46-Mev β, 0.85-1.75-Mev γ); Co^{57} (270 d, no β, 0.014- and 0.12-Mev γ); Co^{58} (71 d, 0.48-Mev β, 0.81-Mev γ). Various other cobalt isotopes have been used[1] to label vitamin B_{12}. Cobalt-57 is reported[104] to give the lowest irradiation of a patient. Labeled vitamin B_{12} (Co^{57}) has been shown to concentrate in the parathyroid in dogs[105] and is therefore potentially useful in parathyroid conditions.

Cesium

Cs^{137} (30 y, 0.51- and 1.18-Mev β, 0.662-Mev γ from daughter Ba^{137}). This isotope, which is recovered as a by-product from the processing of nuclear reactor fuel, is a relatively new isotope, but many Cs^{137} teletherapy units are in use (Table VII-2). The long half-life and low cost of the activity are the principal advantages over Co^{60}. Typical

units have been described elsewhere.[1c,93a,106] (See also book review section of this issue.)

Iridium

Ir^{192} (74 d, 0.67-Mev β, 0.30- to 0.47-Mev γ's). Radioiridium teletherapy is an old idea.[107] A new 100-curie Ir^{192} irradiation unit weighs only 38 lb; a 10-curie source weighing 28 lb is also available.[108] A unit with interchangeable ends of different sizes and shapes, for interorificial therapy, has been described recently.[109]

The half-life of Ir^{192} is about 5% that of Co^{60}, and its less penetrating gamma rays allow greater local absorption and necessitate less protection for hospital personnel. Internally, it has been used as seeds embedded in nylon ribbon or as needles.[110] Experimentally, an Ir^{192}-labeled resin suspension has been used with external counting for determining gastric emptying rates.[111]

Arsenic and Copper

As^{74} and Cu^{64} (17.4 d and 12.8 hr, respectively; positron emitters). These isotopes have been used for localizing brain tumors.[112]

As^{76} (26.5 hr) and As^{72} (26 hr). Arsenic-76 has been used for brain tumor localization, as has As^{72}, which yields nearly three times as many positrons per average disintegration[113] as As^{74}.

Gallium

Ga^{72} (14.3 hr, 0.64- and 0.96-Mev β's, 0.63- to 2.20-Mev γ's). Gallium-72 has been used to localize malignant bone tumors by external measurements, but the short half-life results in a concentration of this chemically toxic material which may perhaps be higher than is warranted by the radiation delivered.[114]

Ga^{68} (68 min, 1.9-Mev β, 1.1-Mev γ). Gallium-68 has been suggested for such applications as brain tumor localization by annihiscopy. A source from which Ga^{68} can be easily and rapidly obtained has been designed.[115]

Palladium

Pd^{109} (13.5 hr, 1.03-Mev β) and Pd^{103} (17 d). A mixture of these two isotopes has been used for interstitial irradiation.[116]

Tantalum

Ta^{182} (115 d, 0.36- to 0.51-Mev β's, 1.12- and 1.22 Mev γ's). Tantalum-182 wires have been used as tissue implants in control of malignant conditions.[117]

Ruthenium-Rhodium

Intragastric beta irradiation with Ru^{106}-Rh^{106} has been tried.[118]

Lutetium

Lu^{177} (175 d; also 6.8 d, 0.50-Mev β, 0.18- and 0.38-Mev γ). Lutetium-177 has been suggested for infiltration to lymph nodes involved in malignant disease.[119]

Sulfur

S^{35} (87 d, 0.167-Mev β, no γ). Sulfur-35 has been used for determining extracellular water and as a therapeutic agent in chondrosarcoma and mycosis fungoides.[120] Labeling of Congo red dye with S^{35} decreases the amount of dye needed for diagnosis of amyloidosis, eliminating the side effects that occur in some patients.[121] Methionine labeled with S^{35} has been suggested for use in detecting pancreas tumors.[122]

Sodium

Na^{22} (2.6 y, 0.54-Mev β, 1.3-Mev γ) and Na^{24} (15 hr, 1.4-Mev β,

2.7- and 1.4-Mev γ's). These isotopes have been used for measuring extracellular water, sodium space, and exchangeable sodium; for determining when skin grafts are complete; and for cancer therapy.[1,123] Because of its shorter half-life, Na^{24} has advantages for some uses.

Potassium

K^{42} (12.4 hr, 3.6-Mev β, 1.5-Mev γ) and K^{43} (22 hr, 0.3- and 0.27-Mev β's, 0.37- to 0.62-Mev γ's). These isotopes have been used to localize brain and breast tumors, measure exchangeable potassium, and study potassium uptake by red blood cells.[1,124] Less shielding is required with K^{43} because of its lower energy gamma radiation.

Rubidium

Rb^{86} (19 d, 1.78- and 0.72-Mev β's, 1.1-Mev γ). Since rubidium acts similarly to potassium and has a more convenient half-life than K^{42}, it has been suggested for use in determining exchangeable potassium.[124c] However, it must be remembered that Rb^{86} is not a potassium isotope. Myocardial scanning tests have been made with Rb^{86} in dogs.[125]

Bromine and Chlorine

Br^{82} (35.7 hr, 0.44-Mev β, 0.55- to 0.78-Mev γ's). Bromine-82 ratios in blood and spinal fluid have been used to differentiate between tuberculous and nontuberculous meningitis.[126] For determining extracellular fluid,[127] it has advantages over the 37-min Cl^{38} or the 3×10^5-y Cl^{36} (ref. 1a, p. 233).

Solutions of Br^{82}, enclosed in a rubber balloon, were used in early work[1,123e] for treating carcinoma of the bladder.

Silver

Ag^{111} (7.5 d, 1.05- and 0.69-Mev β's, 0.34-Mev γ). Colloidal

Ag^{111} was one of the early radioactive colloids used.[128] Silver-111, injected into the bloodstream as a protein-silver complex, is removed less rapidly by a cirrhotic liver than by a normal one.[129]

Zinc

Zn^{63} (38 min, 1.38- to 2.34-Mev β's, 0.67- and 0.96-Mev γ's). Zinc-63 was used in early work for control of malignant effusions.[1] Its use in studying zinc metabolism was reported recently.[130]

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VIII. SAFETY

The U. S. Atomic Energy Commission is deeply concerned with the safety aspects of the isotopes program. Their contracts include studies of both shipping casks and radioactive sources and source capsules as evidence of their interest in reducing radiation hazards. Battelle Memorial Institute, for example, has embarked upon a program to develop safety performance criteria for sealed sources.[1] Information has been accumulated on source applications, designs, fabrication techniques, and testing procedures. Existing regulations, specifications, and performance criteria were also surveyed, compiled, and evaluated. This work and the work at ORNL on actual testing of commercial sources will be reported in more detail at a later date.

Exposure of Personnel Handling Radioisotopes

The results of a survey[2] of persons working with radioisotopes indicated that, in eight categories of use (Table VIII-1), the annual average external radiation exposure is probably not more than 1250 mrems. The attendant risks to personnel are concluded to be negligible compared to risks of everyday life (Table VIII-2).

The survey was conducted by sending out questionnaires to various establishments licensed to use radioisotopes in the manufacture of a commercial product and in medical diagnosis and treatment, and to universities. The persons included in the results were those actually working with or handling radioactive elements. Of 253 licensees replying, representing 3864 persons, 85% received whole-body doses of 600 mrem or less. The exposure data were taken from film-badge dosimeters.

Source Testing

The stainless steel capsule of a 9-year-old $Cs^{137}Cl$ source showed

Table VIII-1. Whole-body Radiation Exposures of Personnel Handling Radioisotopes

Field of Isotope Use	No. of Licensees	Exposure Range	mrem Avg	No. of Persons Exposed					% Exposed to 0-600 mrem
				0-600 mrem	601-4999 mrem	5-11.9 rem	12-27.5 rem	Total	
Medical diagnosis and treatment									
X rays or Ra only	12	0- 3552							
All sources	94	0-12,630	461	914	264	6	1	1185	77
Industrial radiography									
Radioisotopes only	17	0- 2130	253	38	6	0	0	44	86
All sources	47	0- 7400	472	192	43	2	0	237	81
Thickness, level, or density gaging	19	0- 4035	604	59	45	0	0	104	57
Universities	24	0- 3516	178	1259	58	0	0	1317	95
Isotopes* processing and resale	24	0-27,453	1726	187	37	21	6	251	75
Industrial tracers	6	0- 1280	244	89	5	0	0	94	95
Radiation effects studies	10	0- 2140	112	416	23	0	0	439	95
Static elimination or ionization sources	-	-	-	-	-	-	-	-	-
Total	253			3297	531	29	7	3864	85

*Beta doses were significant in this use category; consequently, the tabulated exposures overestimate the whole-body dose.

Table VIII-2. Comparison of Risk of Radiation Exposure with Everyday Risks of Life

Field of Isotope Use	RADIATION EXPOSURE 1960 Exposed Population*	No. of Persons Exposed 5-12 rems	No. of Persons Exposed 12-27.5 rems	Exposure Probability 5-12 rems	Exposure Probability 12-27.5 rems
Medical diagnosis & treatment	1378	6	1	4.3×10^{-3}	7.2×10^{-4}
Industrial radiography	281	2	0	7.1×10^{-3}	$<3.6 \times 10^{-3}$
Isotope processing & resale	251	21	6	8.4×10^{-2}	2.4×10^{-2}
All	3864	29	7	7.5×10^{-3}	1.8×10^{-3}

EVERYDAY RISKS OF LIFE

Occurrence	Age Group, years	1960 or 61 Exposed Population	No. of Occur-rences	Probability of Occurrence
Fatal accidents	15-64	1.1×10^{8}	5.2×10^{4}	4.9×10^{-4}
Fatal motor vehicle accident	15-64	1.1×10^{8}	2.8×10^{3}	2.4×10^{-4}
Drowning fatalities	15-64	1.1×10^{8}	3.9×10^{3}	3.6×10^{-5}
Cancer fatalities	15-64	1.1×10^{8}	1.2×10^{5}	1.1×10^{-3}
Home accidents resulting in insurance claims	All	6.0×10^{5}	1.6×10^{4}	2.7×10^{-2}
Sports and recreation accidents resulting in insurance claims	All	6.0×10^{5}	1.0×10^{4}	1.7×10^{-2}
Civilian unemployment	14 or older	7.3×10^{7}	4.0×10^{6}	5.5×10^{-2}

*Age group 18-65 years.

no corrosion or deterioration when examined visually and metallurgically.[3] This source, the first $Cs^{137}Cl$ source of greater than 1000 curies built by ORNL, was made in March 1954 and had been used continuously in a teletherapy machine at the Oak Ridge Institute of Nuclear Studies since 1955. It contained two 1.250-in.-dia pellets, of 0.531 and 0.504 in. height, 33.07 and 27.86 g weight, and 760 and 780 curies activity, respectively.

The source (Fig. VIII-1) was enclosed in two concentric type 316 stainless steel capsules, each with a 0.020-in.-thick window. The inner capsule was 2.575 in. long, 1.500 in. o.d., and 1.260 in. i.d. and was plugged with a type 316 stainless steel plug 1.5 in. long and sealed with Easy Flo silver solder. The outer capsule was 3.265 in. long, 1.750 in. o.d., and 1.520 in. i.d., and was plugged with a type 410 stainless steel threaded plug and sealed with All-State No. 430 silver bearing solder.

When the source was removed from the teletherapy machine head, the outer capsule appeared in excellent condition, with no signs of corrosion or discoloration. A smear of the outer surface showed an activity of 1920 dis/min. No bubbles appeared when the source was immersed in ethylene glycol in a glass vacuum vessel (sensitivity of method 5×10^{-5} cc/sec), and the pressure above the source was decreased to -25 in. Hg (about 5 in. Hg absolute).

During sawing off of the outer capsule, a corner of the inner capsule was inadvertently cut off, but no other leaks were observed in the vacuum leak test. A smear showed a high activity, 5×10^5 dis/min, because of this sawed-off corner. Visual examination of the exterior

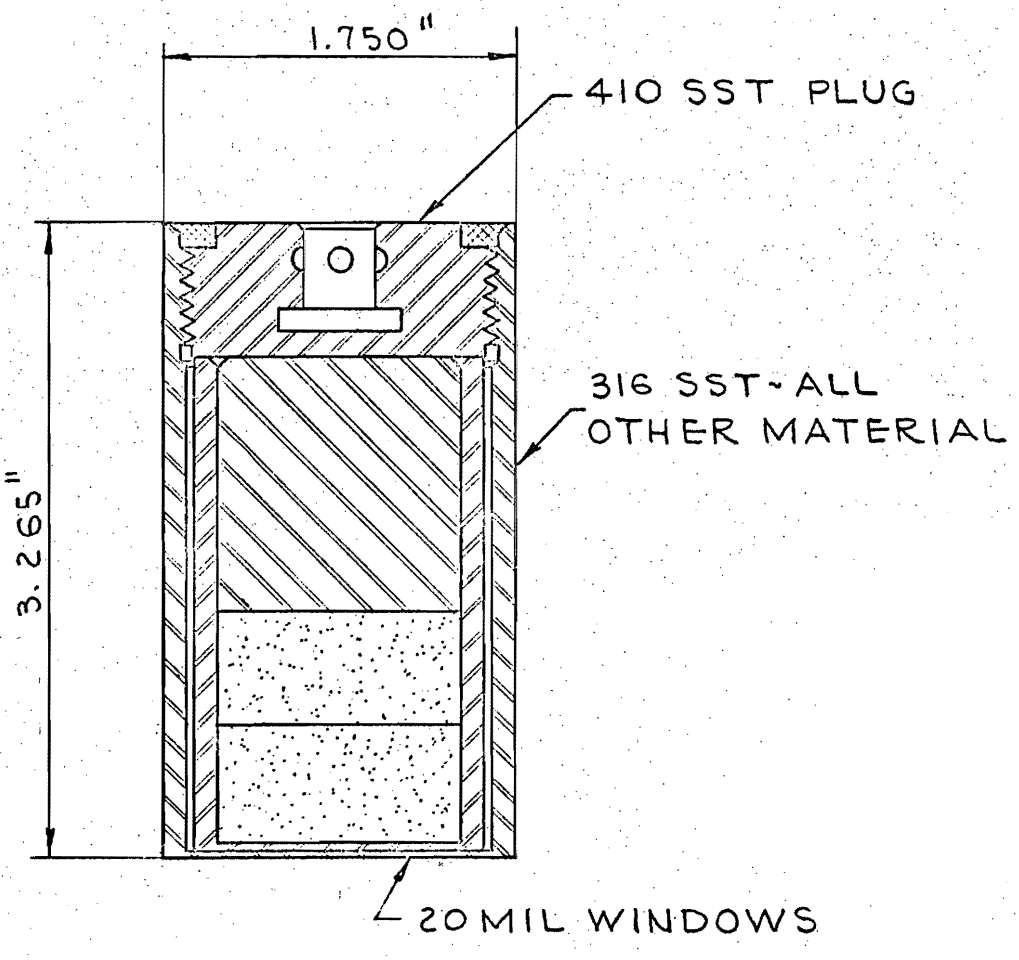


FIGURE 1

1540-CURIE ORINS Cs^{137} CL SOURCE

surface of the capsule showed no evidence of corrosion or deterioration.

When the inner capsule was opened, the source pellet surface appeared similar to that of a new pellet. It was white to gray in color, with a mottled brown streak near one edge, and the edges of the pellet and the inner wall of the capsule at the edge of the pellet were distincly brown. However, this discoloration was probably caused by heating during the original scaling of the capsule since, when the pellets were removed, the upper edge of the pellet and the adjacent area on the capsule were seen to be the only affected areas. The inner surface of the capsule and the window area were as bright and shiny as new material, and inspection under 80X magnification did not show any pitting or cracking.

The capsule was decontaminated by ultrasonic cleaning in water and photographed at 80X magnification (Fig. VIII-2). Metallurgical studies of sections (Fig. VIII-3-5) at 100X magnification showed no evidence of change in crystalline structure or deterioration of the capsule wall window.

Fig. 2. Source Capsule Before Being Sectioned for Metallurgical Studies

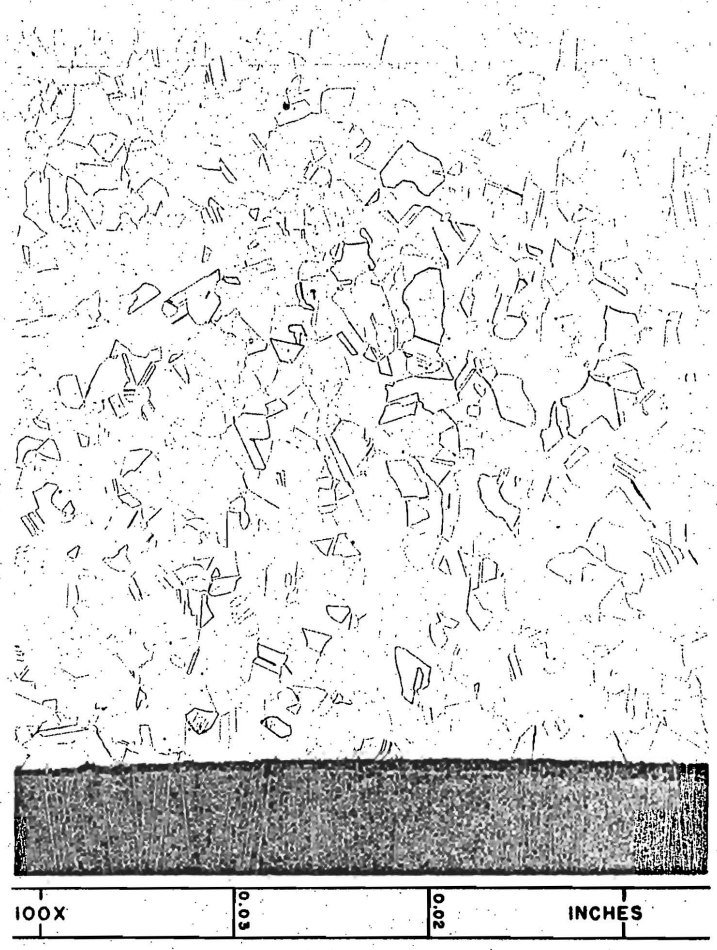


Fig. 3. Transverse Section of ORINS Source Capsule Wall, ID

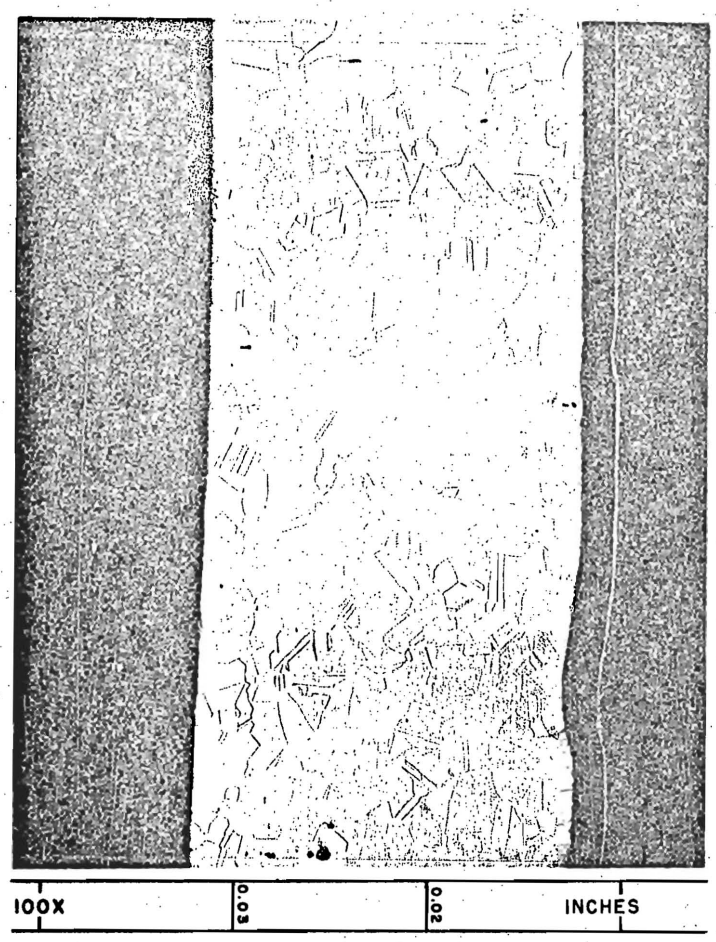


Fig. 4. 20-mil Window Sections (Scale on bottom inside of window is to left)

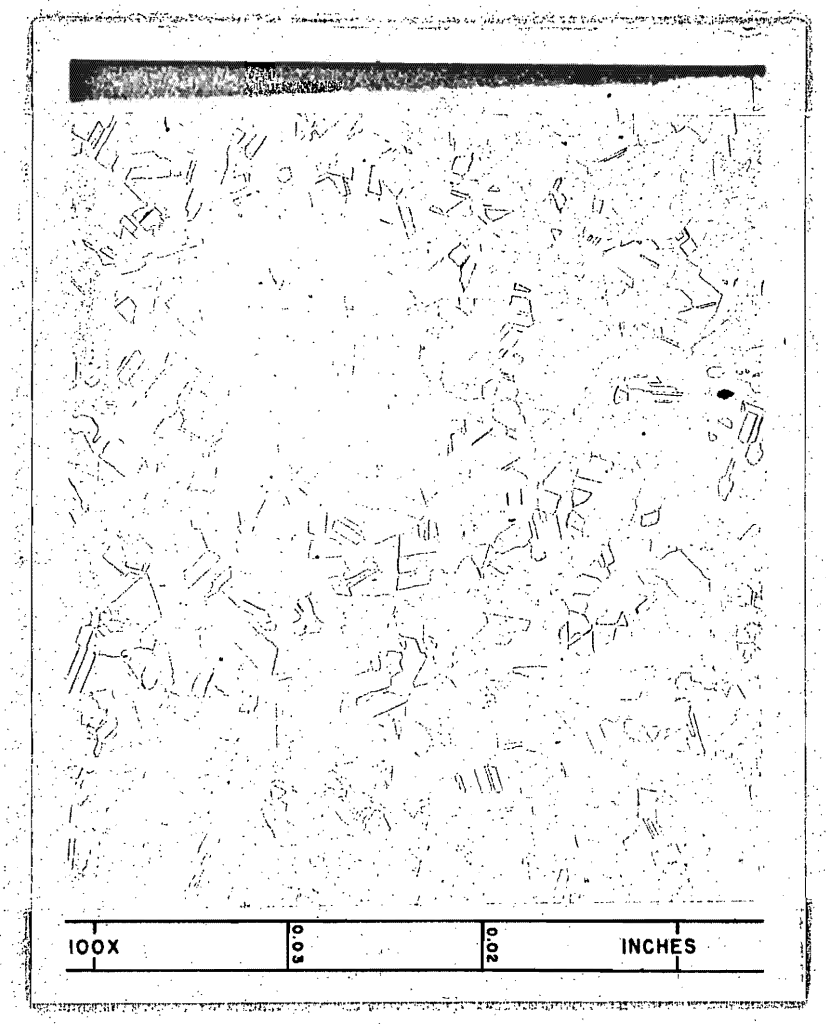


Fig. 5. Longitudinal Section of Wall of ORINS Source Capsule, ID

Shipping Cask Testing

The results of experiments performed at ORNL indicated that unprotected lead-filled steel casks weighing up to 6 tons will meet structural requirements of 10CFR72 with the exception of the 6-in.-dia puncture test. Since the results of a horizontal or piston drop can be estimated with a reasonable degree of accuracy, casks can probably be designed to meet the piston requirements. It is doubtful, however, that such casks would meet fire requirements. Small radioisotope shipping casks up to 1.5 tons and approximately 20 in. dia can be protected from both impact and fire with low-cost wooden shields.

Lead-filled steel casks and wooden fire and impact shields have been experimentally tested by the Isotopes Development Center at ORNL as part of a continuing program to improve radioactive material shipping procedures and packaging. The program has included both structural and fire resistance tests, and experience gained has set the standards for radioisotopes packaging and shipping throughout the U.S. and the world. Since 1946, the beginning of the radioisotopes program at ORNL, more than 176 thousand (as of May 31, 1963) shipments containing $\sim$ 2.2 million curies have been made without a single accident in which a release of radioactivity was involved. However, with the trend in radioactive material shipments to larger curie content per package, the need of greater safety in package design and shipping has become more apparent. Proposed and existing AEC and ICC regulations are given in references 1 and 2.

The testing program for fuel shipping casks was discussed in a recent issue of Nuclear News.[3]

A Lead-filled Cask Tests

Structural Tests. Forty-one drop tests* have been made with five lead-filled steel casks weighing 2725 lb each and two weighing 11,760 lb each. A lead-filled steel cask is shown diagrammatically in Fig. 6. Prior to a drop, each model cask was instrumented with two piezoelectric accelerometers, 12 dynamic strain gages, and compressometers to measure inner wall deflections. Some casks were covered with a brittle lacquer to determine the areas of high stress and thereby indicate where some of the instrumentation was to be placed. The casks were dropped horizontally, on end, on an edge (corner), and on pistons, and each cask was dropped more than once.

As a result of the drops, many areas of the outer and inner shell took a slight but permanent set. Except for the actual impact area, the inner shell was subject to higher stresses than was a comparable position on the outer shell. The maximum permanent change of the inner diameter of the casks was 1 in. for both the small and large casks when dropped 20-30 ft. In no drop was the inner shell penetrated. A water-filled cask continued to hold water after a 15-ft horizontal drop.

Figure 7 shows a 1.4-ton cask being dropped on a corner. The picture was made from selected frames taken by a high-speed motion picture camera operating at about 3600 frames per second. The sequence

* Made in conjunction with the Chemical Technology Division.

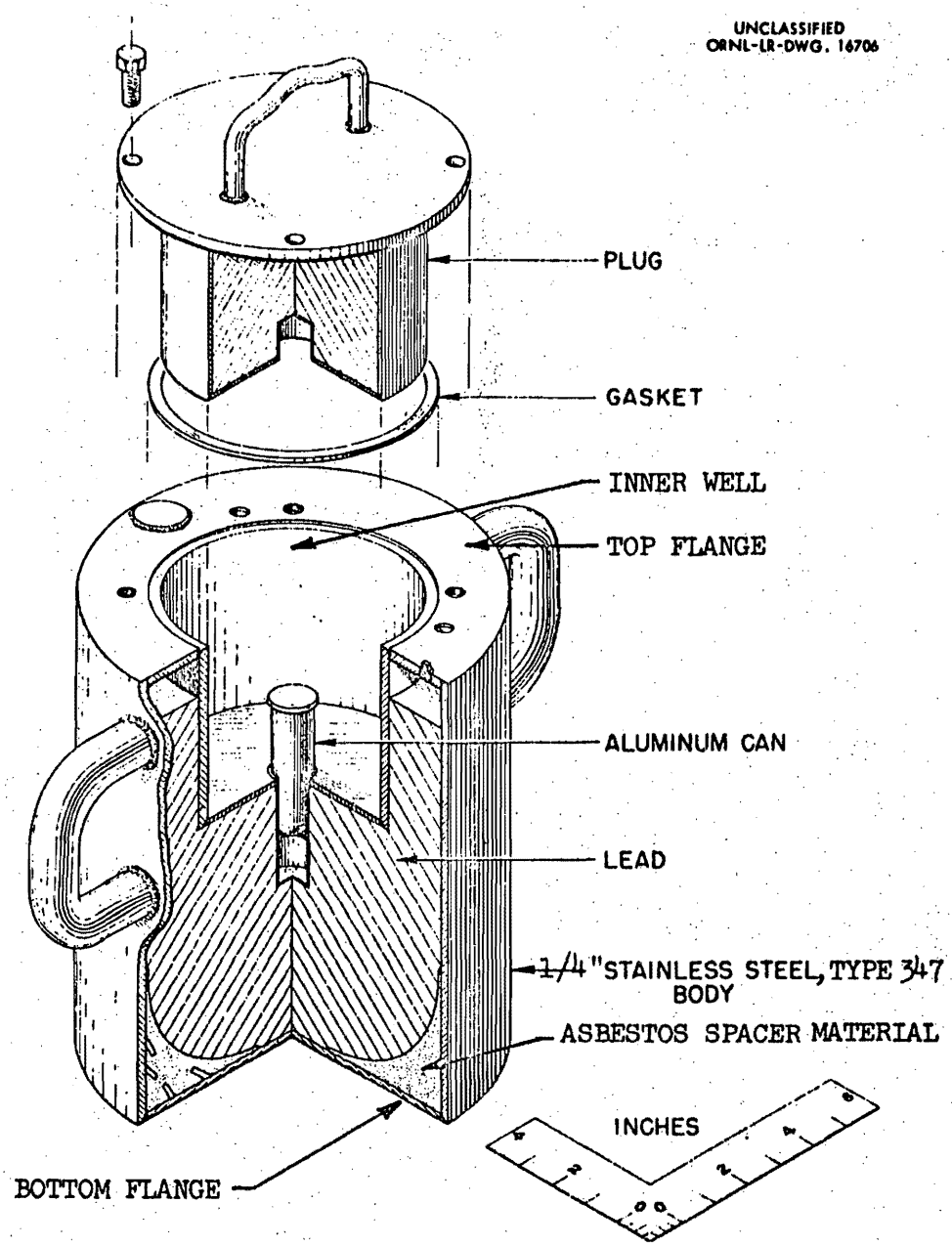
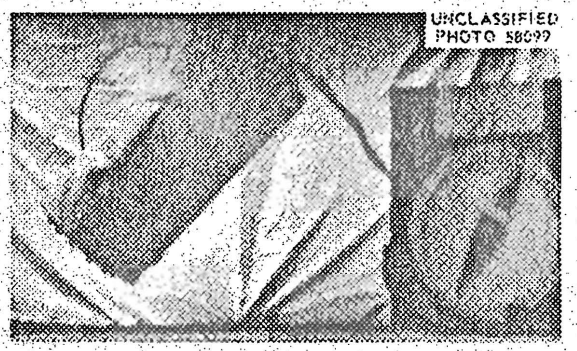
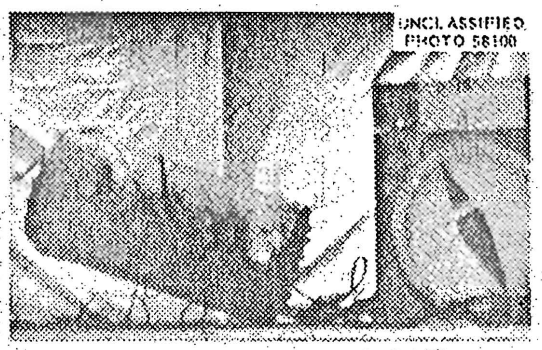


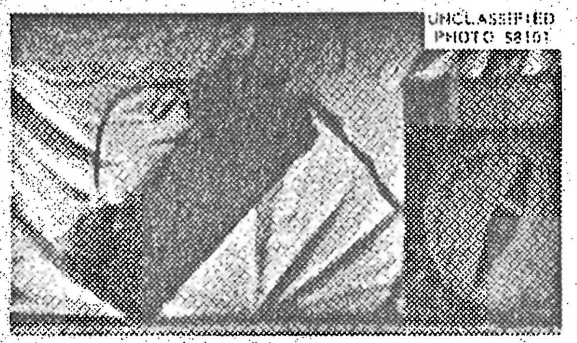
Fig. 6. Returnable Container for Solid Materials (Lead Shielding, 4 to 7½ in.).



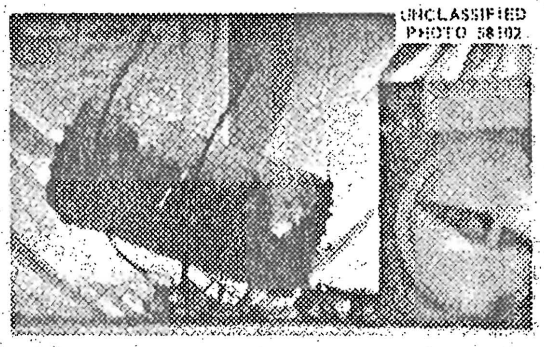
Prior to impact



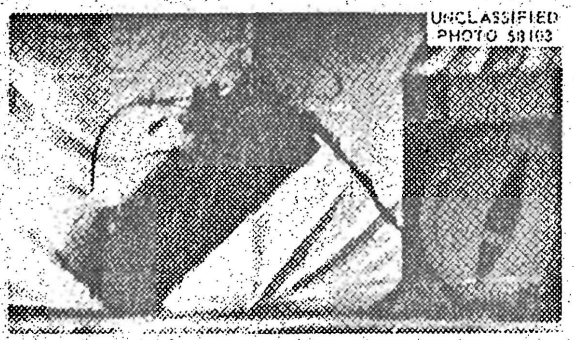
Second impact - time: 0.102 sec



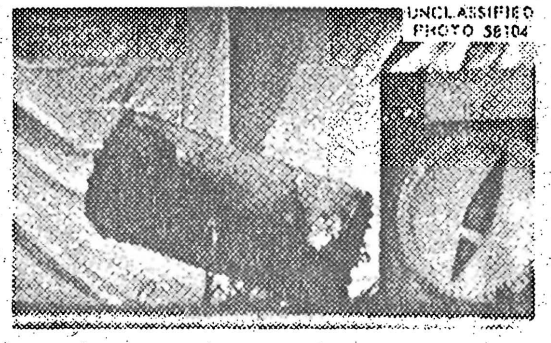
Initial impact - time: 0.000 sec



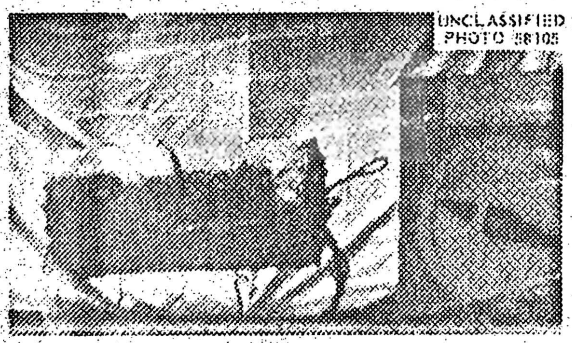
Second bounce - time: 0.196 sec



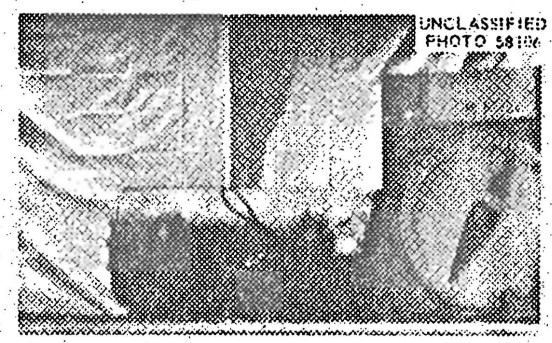
Peak deceleration - time: 0.0083 sec



Third impact - time: 0.391 sec



First bounce - time: 0.071 sec



Final position - time: 0.515 sec

Fig. ~~1.1.1.~~ 7 Corner Drop of a 1.4-Ton Model Shipping Cask; Drop Height 15 ft.

shows that the total impact, including bounces, occurs in about 0.5 sec. Following a 15-ft corner drop of a 1.4-ton cask coated with brittle lacquer, radial stresses were observed perpendicular to the cracking in the brittle lacquer material and were concentrated around the lead fill holes. After horizontal drops of a 1.4-ton cask from 15 and 28 ft, a small crack appeared in the weld where the outer shell and the end flange join. Cracks in the brittle lacquer indicating high stress around the lead fill hole were also seen. When a 1.4-ton cask was dropped 5 ft onto a 2-in.-dia piston, the piston penetrated about 1.5 in. into the cask.

Fire Tests. In a 15-min fire test at 1320°F, made by Underwriters Laboratories,[6] lead was ejected from the cask. In fire tests at ORNL,[7] a small amount of water resulted in spraying of molten lead through pinholes in the top flange of the cask. Therefore it is doubtful that unprotected lead-filled casks would meet fire requirements.[4]

Wooden Fire and Impact Shields

A typical shield is 3-5/8-in.-thick maple (Fig. 8) and has mitered corners of nailed construction. A mild steel frame holds the shield intact under impact conditions. The frame is made of 1-in. steel straps and 1.5-in. steel angle iron for the small size, and up to 2.5 in. and 3 in. angle iron for the largest shield. The top is hinged for removal of the container.

The smallest shield tested weighs 120 lb and holds a lead-filled steel container with 1.5-in. shielding and weighing about 60 lb. The largest shield weighs 570 lb and holds a 2600-lb container with 8-in-thick lead shielding.

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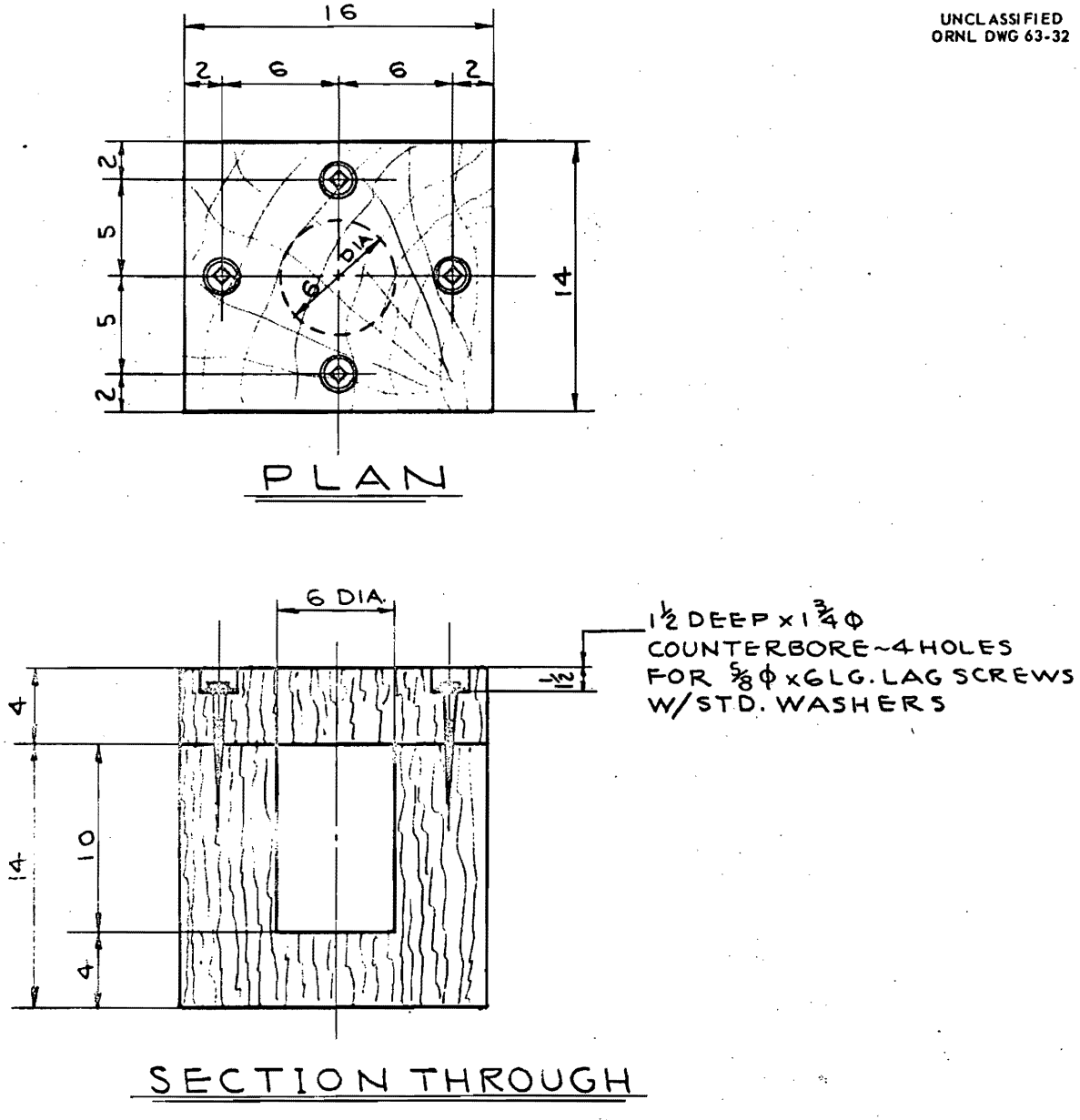

Fig. 8. Wooden Fire Shield. [redacted]

Methods under consideration for protecting larger shields (larger than 1.5 tons) include shock absorbing aluminum honeycomb.

Structural Tests. The lead-filled steel cask models and the wooden fire and impact shields were dropped from heights up to 30 ft onto a pad consisting of a 12- by 12-ft piece of steel armor plate 4.5 in. thick, backed up by a 5-ft-thick slab of reinforced concrete. Below the pad was a 3-ft-dia reinforced column reaching down approximately 7 ft to bedrock and then extending 3 ft into the bedrock.

The small wooden shield, 120 lb, which measures 14 by 14 in. by 17-3/8 in., was dropped from heights of 15 and 30 ft onto a 6-in.-dia piston. Very little damage was done to the shield, which was loaded with 4 lead bricks weighing a total of 112 lb to simulate the cask. There was some damage to the hinges of the top.

A second shield of the same size was dropped 30 ft onto a corner. There was no damage to the hinges and damage to the rest of the shield was minimal.

The larger shields did not perform quite so well as the smaller ones, but they did meet current AEC[1] and ICC[2] regulations on structural requirements. The larger shield is designed for a 3.5-ft drop onto a 6-in.-dia piston and a 30-ft free fall onto one corner. Testing on the larger shields is not yet complete.

Another design using a solid block of wood was tested at 15-ft free falls but because of cracking and splitting of the wood was abandoned.

Fire Resistance Tests. Fire resistance tests were made on pine, hickory, maple, and ebony. Maple was selected because of its resistance

to burning (2 in. penetration vs 2.5-3 in. for pine in 1 hr), availability, strength, insulating properties, and cost. Ebony, of course, was too expensive and was a rather poor insulator. Hickory was not available in the thicknesses and widths needed and was no less expensive than maple. Pine was the best insulator but burned more rapidly and did not have the strength of maple.

The shields were tested for 1 hr in the flame of a pressurized kerosene burner. The outside temperature was maintained at 900 ± 50°C for 1 hr to simulate the standard 1-hr fire test.

Pine shields burned 2.5-3 in. through. Maple burned about 2 in. Both appeared to be good insulators with the maximum temperature rising to 30°C in the pine shield and to 60°C in the maple shield after 1 hr exposure to the fire. Figure 9 shows the maple shield after 1 hr exposure to the fire.

The average cost of the shields will be approximately $250 each. The total cost represents 10% of the total cost of replacing all carriers now used and approximately 20% of the cost of modifying all the carriers now used in order to meet new safety criteria. A disadvantage is that freight costs will be increased from 20% to 200% on radioisotope shipments requiring this additional protection.

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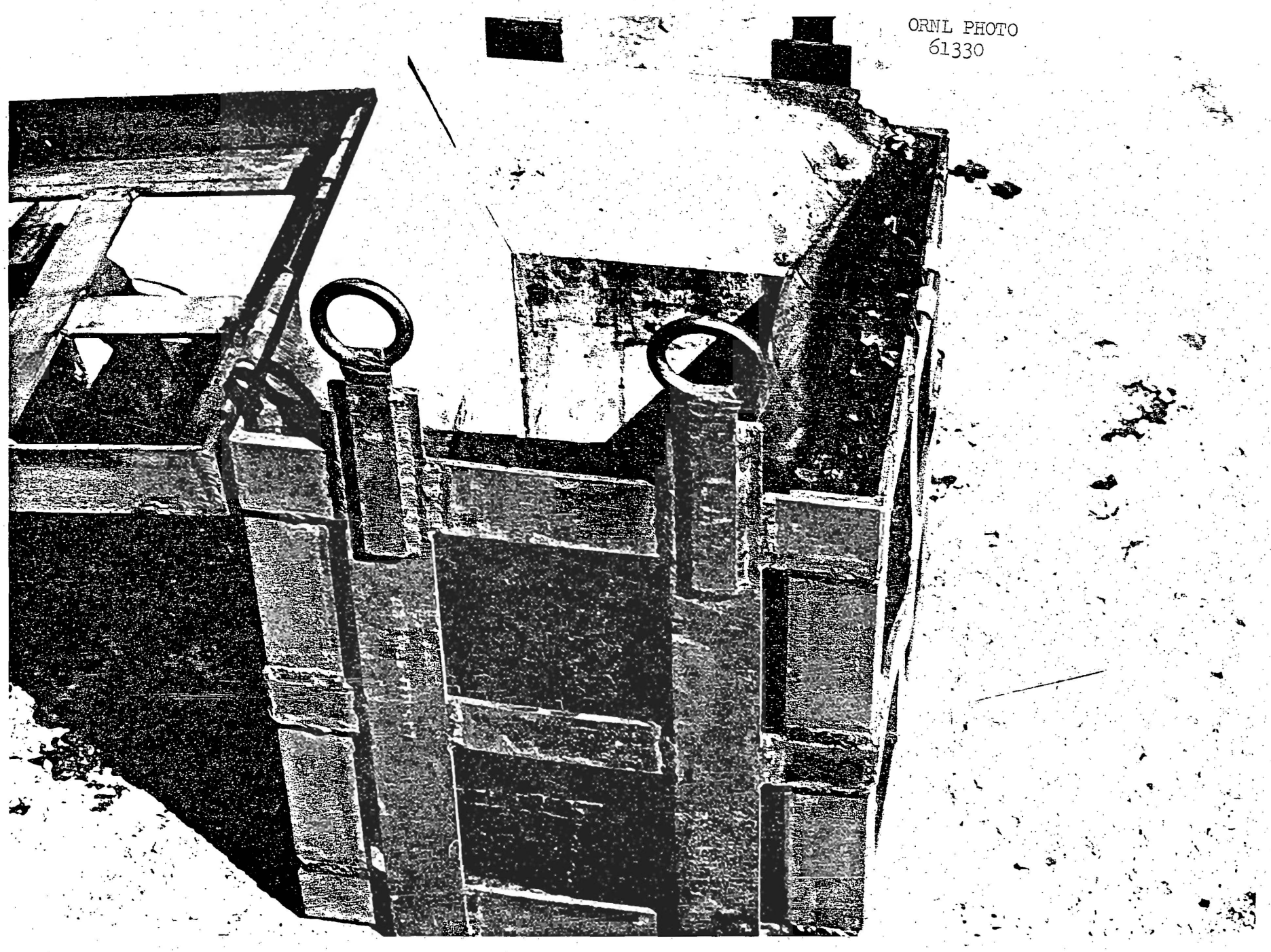

Fig. 9. Fire and impact shield after 1- hr fire test.

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3. K. W. Haff, Examination of Nine-Year Old $Cs^{137}Cl$ Source, USAEC Report ORNL-TM-584 (May 14, 1963).

4. Proposed CFR, Title 10, Part 72, Federal Register, September 23, 1961.

5. ICC Docket No. 3666, In the Matter of Regulations for Transportation of Explosives and Other Dangerous Articles, Proposed, April 12, 1963.

6. E. C. Lusk, J. E. Gates, New Developments in the Design of Shipping Casks for Irradiated Fire Elements, Nuclear News, 6(6): 11-14 (1963).

7. L. B. Shappert, Impact Testing of Shipping Containers, American Nuclear Society, Transactions 1963 Annual Meeting, Salt Lake City, Utah, June 17-19, 1963.

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References (continued)

10. K. W. Haff, ORNL Cask Testing Program; Tests of Radioisotope Shipping Cask-Type C, ORNL-TM-206, Apr. 23, 1962.

11. NFPA251, Methods of Fire Tests of Building Construction and Material.

FIGURE LIST

Dwg. or Photo Number	Caption	Page
-	Fig. VIII-1. 1540-Curie ORINS $Cs^{137}Cl$ Source.	
Photo 61322	Fig. VIII-2. Source Capsule Before Being Sectioned for Metallurgical Studies.	
Photo 4537	Fig. VIII-3. Transverse Section of ORINS Source Capsule Wall, ID.	
Photo 4541	Fig. VIII-4. 20-mil Window Sections (Scale on bottom inside of window is to left).	
Photo 4540	Fig. VIII-5. Longitudinal Section of Wall of ORINS Source Capsule, ID.	
ORNL-LR-DWG 16706	Fig. VIII-6. Returnable Container for Solid Materials (Lead Shielding, 4 to 7-1/2 in.).	
-	Fig. VIII-7. Corner Drop of a 1.4-Ton Model Shipping Cask; Drop Height 15 ft.	
ORNL DWG 63-32	Fig. VIII-8. Wooden Fire Shield.	
ORNL Photo 61330	Fig. VIII-9. Fire and Impact Shield after 1-hr Fire Test.	

IX. AVAILABLE MATERIALS AND SERVICES

General Electric has established a new Irradiation Services and Products Section in its Atomic Power Equipment Dept. to meet the growing demand for radioisotopes and irradiation services. With headquarters at Vallecitos Atomic Laboratory (Pleasanton, California), it will offer a complete range of nuclear irradiation services, including production of radioisotopes.

Union Carbide Nuclear Co. has announced that interest in isotopes is sufficient to encourage the company "to become a major producer of isotopes and to provide consultation and services in radiation application problems." They are now supplying numerous routinely used isotopes and have indicated that they can prepare others in bulk quantities or as special materials. They also plan to quadruple their neutron-activation analysis business.

In February, bacon became the first food to be approved by the U.S. Food and Drug Administration for radiation sterilization. The bacon, which is irradiated in cans with 4.5 megarads from a Co^{60} gamma source, will next be tested under arctic and tropical conditions. Some twenty other foods being studied for such processing include chicken, ham, potatoes, shrimp, and oranges.

In the meantime, the USAEC is negotiating with Stone & Webster to build the first seafood products irradiation pilot plant, at Gloucester, Massachusetts. The $600,000 plant, with a 300-kilocurie Co^{60} gamma source

and a capacity of 1 ton/hr, will be used to demonstrate the feasibility of radiation pasteurization of seafoods. The AEC's 1964 budget includes $350,000 for a transportable Co^{60} irradiator for field studies with fruits and $200,000 for a grain-disinfestation pilot plant.

A Co^{60} irradiation plant has been opened in England by Johnson's Ethical Plastics Ltd. for sterilizing plastic disposable syringes. The source initially is a 50-kilocurie unit, but the design is for 250 kilocuries.

Export of radioisotopes will be simplified by recent amendments to AEC regulations. A general license -- one for which no application to AEC is required -- has been established to replace the specific license formerly required for the export of certain forms of moderate quantities of tritium and polonium. Use of these isotopes, which is widespread and considered safe in the atomic energy industry, will be subject to the radiation safety laws and regulations of the countries to which they are exported.

The sale price of heavy water has been cut from $28 to $24.50 per pound f.o.b. Savannah River Plant, Aiken, S. C. The base charge for leased heavy water has also been cut to $24.50 for leases that do not have a fixed base charge. Further price cuts will be considered if commitments from customers require operation of the heavy water plant at increased capacity.

Oak Ridge National Laboratory has 99.99% Ne^{20} available for routine sales. In June 1963, concentrates of the mid-position isotopes, 20% Ne^{21} and 13% Ar^{38}, became available in very limited amounts.

The consultative committee for Eurisotop, the Euratom Information Bureau, held its first meeting in November 1962. A major topic of discussion was how to provide information on radioisotopes not only to the general public but to professional circles that may benefit from using them. Suggestions offered included traveling exhibits and television programs.

The AEC has executed agreements with Kentucky, Mississippi, California, New York, Texas, and Arkansas for transferring to the states some of its regulatory responsibility for licensing, rule-making, and enforcement in the uses of radioisotopes.

ORNL's new edition of its Isotopes Catalog is now available. For the first time both stable and radioisotope offerings are included together. Worthy of note are the Commission's new low prices for bulk purchases of fission products, Sr^{90}--\$0.75/curie for >30 kcuries; Cs^{137}--\$0.50/curie for > 100 kcuries; Ce^{144}--\$1.00/curie for > 100 kcuries. Cesium-137 at this price should be quite competitive with Co^{60} for many applications.

As of August 12, 1963, the AEC (specifically ORNL and NRTS) will terminate land burial services for licensees in favor of privately owned facilities such as Nuclear Engineering Co., Flemingsburg, Ky., and Beatty, Nev., and Nuclear Fuel Services in upper New York State.

In early June the AEC Regulatory Staff moved to Bethesda, Md. Their address is: Phillips Bldg., Cor. St. Elmo and Norfolk Avenues.

X. BOOK REVIEWS

A Manual for Nuclear Medicine, E. R. King and T. G. Mitchell, Charles C. Thomas, Springfield, Ill., 1961, $13.50, 406 pp.

Due to its early stage of a rapid development, the field of nuclear medicine is not yet clearly defined. In the broadest sense, it should encompass those effects in man that are caused by nuclear radiation originating from both externally located sources and internally deposited emitters. Obviously, this definition would not only refer to the use of isotopes in the hospital but would also include such diverse areas as, for example, radiological civil defense, radiation exposure of high-altitude jet crews and astronauts, radiation-induced changes in human genetics, and many aspects of health physics. In the narrowest sense, nuclear medicine is taken as that field of medicine which deals with the administration of radioactive isotopes to patients, predominantly for diagnostic purposes. King and Mitchell's text treats the subject in this restricted manner.

The manual originates from courses given for several years at the National Naval Medical Center, Bethesda, Maryland. Therefore instructional material, laboratory exercises, and clinical procedures are well tested and perfectly adjusted to the requirements of physicians and medical students who intend to use radioisotopes as diagnostic tools. Because of this clear orientation toward the needs of medical practitioners, emphasis naturally is placed on present-day status rather than on future developments, and on immediate clinical application rather than on laboratory research.

The first four chapters contain radioisotope methods evaluating thyroid function, certain forms of anemia, blood volume, and blood forma-

tion. For each test, a brief background and a detailed laboratory procedure are given. Collectively, these tests represent that section of clinical isotope application which already has become an integral part of diagnostic medicine because the procedures are simple and economical and they yield information not otherwise obtainable. Two subsequent chapters are devoted to scintillation scanning, which currently represents the most rapidly growing branch of medical isotope use. As can be expected, the short space allows only a brief outline of the young but promising infant with its many developmental problems -- e.g., collimator shape and material, recording and print-out systems, and incorporation of isotopes with suitable gamma spectra into chemical compounds that will concentrate preferentially in cancerous tissue. The clinical part concludes with chapters on the use of isotopes in dilution studies, organ function tests, autoradiography, and therapy. The second half of the book contains basic information about physical processes and instrumentation necessary for sensible performance of the previously described clinical procedures.

In summary, King and Mitchell's manual is intended for the training of physicians and medical students who wish to use radioisotopes in diagnostic work. For that purpose the book is admirably suited and can be recommended very highly.

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Künstliche Radioaktive Isotope in Physiologie, Diagnostik, und Therapie (Artificial Radioactive Isotopes in Physiology, Diagnosis, and Therapy), ed. by H. Schwiegk and F. Turba, with 83 contributors. 2d ed. Springer-Verlag, Berlin, 1961. Vol. 1, 1327 pp.; vol. 2, 1248 pp. Set only, DM 398.

The two volumes of this publication are divided into three parts which cover, respectively, properties of isotopes and basic information needed in isotope work; the use of radioisotopes in physiology, pharmacology, and diagnosis (with an introduction by Von Hevesy); and radioisotope therapy. Isotopes of some 50 elements are considered, with about half the chapters in German, one in French, and the rest in English. A few of the radioisotope tracer studies discussed are metabolism of fats, proteins, sugars, and various derivatives; synthesis of biological compounds; metabolism in bones and teeth, mitochondria, and microorganisms; action of viruses, vitamins, hormones, and enzymes; and research in genetics, immunology, and photosynthesis. Clinical uses of isotopes discussed include diagnosis of tumors and of various conditions by labeled vitamin B_{12}; treatment of polycythemia vera and thyroid conditions; intracavitary irradiation; hypophysectomy; and methods of applying radioisotopes. Isotopes discussed for therapeutic uses include not only the well-known ones such as Au^{198}, I^{131}, and P^{32}, but also the less commonly used ones such as As^{76} and Ga^{72}. Each chapter is extensively documented. A 270-page index is included in vol. 2. (M.G.)

Diagnostica e Terapia con i Radioisotopi, P. Bastai, L. Antognetti, G. C. Dogliotti, G. Monasterio; coordinated by B. Bellion. 43 contributors. Edizioni Minerva Medica, Turin, Italy, 1962. Produced under the auspices of the Italian Society for Nuclear Biology and Medicine. 945 + ix pp. (in Italian). L 13,000.

The four parts of this beautifully printed one-volume book provide basic information on radioisotopes and describe diagnostic and therapeutic applications. In the first section are included the elements of nuclear physics and basic principles of activity measuring. Especially useful are tables such as those showing the radiation dose to a patient in various diagnostic tests and the penetration of Co^{60} and Cs^{137} radiation from teletherapy sources; the outline of methods for preparing isotopes; and the detailed instructions, with photographs, for handling radioisotopes. Chapters on lesions resulting from excessive irradiation, protection from radiation, and Italian legislation in this and related fields will be of interest to the industrial physician and health physicist. The mathematical discussion of tracer equilibrium and descriptions of detection methods are probably most applicable in testing new isotopes or finding better ones for specific purposes. The discussion of autoradiography is of general interest. Of special interest to the practicising physician are the last two sections, where diagnostic and therapeutic applications for most of the body organs and tissues and Co^{60} and Cs^{137} teletherapy are discussed in detail.

The book is extensively documented and is illustrated with both diagrams and photographs, including some in color. (M.G.)

Les isotopes radioactifs en médecine, M. de Visscher and C. Beckers. Albert de Visscher-Editeur, Rhode St Genese, Belgium (obtainable from Gauthier-Villars & Cie, Paris), 1961. 166 pp. NF 13.

In the preface, Dr. M. Tubiana, Professor of the University of Paris Medical School, says, "(The authors) have succeeded ... in writing a book where the specialist will find many useful data and the nonspecialist can become educated without becoming confused." The French is very easy reading for an English-speaking scientist who knows only a little French, and a non-nuclear physicist considering the use of isotopes will find the first two chapters a convenient compilation of information needed for understanding radioisotope applications. Most of the actual medical applications discussed are fairly well established ones. The basis for their adoption is explained, which should help a newcomer to the field, but they are not documented.

In the basic information section are elementary discussions of atomic structure, radioactivity, and half-lives and an explanation of terms. The principal isotopes used in biology and medicine are listed, with their half-lives and β and γ energies, and basic principles of such equipment as ionization chambers, Geiger counters, and scintillation media are explained. Medical applications discussed include the use of tracers for studying blood components, circulation, digestion, water, electrolytes, and thyroid function and in tumor diagnosis. Radioisotopic therapy in hyperthyroidism and thyroid cancer, in polycythemia vera, chronic leukemia, and lung cancer and in relieving heart conditions is described.

(M.G.)

Experimental Nuclear Chemistry, Gregory R. Choppin, Prentice-Hall, Inc., Englewood Cliffs, N. J., 1961, xi + 226 pp., $10 ($7.50 as a textbook).

The author, one of the co-discoverers of element 101, mendelevium, is now Associate Professor of Chemistry at Florida State University, Tallahassee, Florida. This book was prepared for use in a course which he teaches at that university. In the preface he thanks his students who helped him "debug" the experiments before they were published; therefore, the 40 experiments should be well tested.

Experimental Nuclear Chemistry can be used in many different teaching situations. It is suitable for a course in isotope techniques, or several experiments from it can be used to enrich traditional courses. In the preface the author states his aims:

"The non-nuclear chemist is often reluctant to use radioisotopes in his research, because of the idea that their use involves extensive revision of techniques, and extreme care in handling. There are special problems involved, but it is hoped that a discussion of their nature will help to reduce them. Some familiarity with the great advantages of radioisotope use, coupled with intelligent awareness of their unique safety requirements, can only promote a much more extensive employment of radioisotopes in all areas of science. For the student who is contemplating, or who has already decided to work in the area of nuclear science or the atomic energy industry, a thorough knowledge of the physical and chemical techniques involved in this field is essential. It is the author's hope that this book will encourage the use of

radioactive isotopes in scientific research."

There are several very attractive features of the book. (1) An entire chapter in a prominent place (Chapter II) is devoted to radiation safety; (2) the last five chapters provide a wide variety of material from which the student (or the instructor) can choose that which most nearly fits the need; (3) Appendix A suggests special projects that the student can do to further enrich the course; (4) most of the experiments can be done with license-exempt quantities of isotopes; (5) a good bibliography is provided.

Appendix B on licensing regulations and sources of isotopes could be misleading to a student or teacher inexperienced with isotopes. Although Oak Ridge National Laboratory sells license-exempt quantities of radioisotopes, it is not the most convenient or economical source for these small amounts. Also, The Isotope Index is probably a more complete source of information on commercial isotope vendors than the Nucleonics Buyer's Guide Edition suggested in the Appendix.

This book, which was also reviewed by J. W. Cobble of Purdue University in Journal of Chemical Education for January 1963, is an excellent addition to the textbooks designed to teach nuclear laboratory techniques.

(RHL)

Adventures in Radioisotope Technology, George Hevesy, Pergamon Press, New York, 1962, 1074 pp., $30 for the set.

If you had to choose one man as the father of isotope technology, there could be only one logical choice -- George Hevesy. To give his record of first accomplishments is to give a history of isotope uses:

... use of a radioisotope as a tracer in inorganic work (1913, solubility of lead sulfide and lead chromate)

... use of a radioisotope as a tracer in botany (1923, lead in the horse-bean)

... use of a radioisotope with an animal (1924, bismuth in the rabbit)

... use of a stable isotope as a tracer in biology (1934, deuterium in man)

... use of an artificial isotope in biology (1935, P^{32} in rats)

... making of a neutron activation analysis (1936, rare earths)

These two volumes contain 100 of Hevesy's papers (all translated into English) on the use of radioisotopes, and edited as they are, they make a fascinating scientific autobiography. By putting papers in subject categories and commenting on each group as a whole, the author is able to give a cohesive account of his scientific accomplishments.

The book opens with a short autobiographical paper which was originally published in Perspectives in Biology and Medicine. This paper helps to place the scientific papers in their proper historical framework. The book ends with three lectures: the lecture he gave on receiving the Nobel Prize in December 1944 and two lectures which he

gave some time later. These three lectures summarize his scientific accomplishments and round out the book.

This is not a book (or really books) that one can sit down and read through -- rather it can be browsed in, read one subject at a time, or used as a reference book. Because of Hevesy's tremendous breadth of interests there is something here for everyone -- biologist, physicist, chemist, or medical doctor.

It seems a pity to complain about anything in reviewing a book as valuable as this; however, I have two criticisms. First, the comments after each section, which contain some of the best human-interest-type information in the book, are too short. They whet your appetite for more. For example, in one comment Hevesy tells that the 600-mg radium-beryllium source he used to make his P^{32} was loaned to him by Professor Niels Bohr who had received it as a present on his fiftieth birthday. Second, while using this book for bibliography sources in writing other parts of this review, I found several incorrect references apparently resulting from careless proofreading. In spite of these two relatively minor faults, this is a worthwhile book and highly recommended for anyone interested in isotope technology.

(RHL)

Radioactive Isotopes in Biochemistry. By Engelbert Broda, Professor of Chemistry, University of Vienna (Austria). D. Van Nostrand Company, Inc., 120 Alexander Street, Princeton, New Jersey. 1960. x + 376 pp. $11.50.

This book is a remarkably concise survey of principles of methodology in the use of radioactive tracers and their application to selected problems in biochemistry. The author avowedly makes no pretense to completeness in any direction. The admitted deficiencies in detail are compensated for to a large extent by more than 3200 references to the original literature, including many of the latest pertinent papers.

The first third of the text is devoted to eight chapters dealing with radioelements in biochemistry, principles of radiochemistry, radiosynthesis, isotope effects, radiation chemistry, radiation biology, protection against radiation, and measurement of radioactivity. This is followed by two chapters on the application of radioactivity to analysis of living matter and absorption and excretion of elements.

The last half of the book entails six well organized chapters on the principal problems and special topics in intermediary metabolism.

The English translation reveals a sound grasp of and a feeling for both the original German and English idiom.

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(Reprinted with the kind permission of the editors of the Journal of the American Chemical Society.)

Radioisotopes and Bone. Edited by P. Lacroix and A. M. Budy under the direction of F. C. McLean. F. A. Davis Co., Philadelphia (Blackwell Scientific Publications Ltd.), 1962, 522 pp. $15.00.

The symposium, held in Princeton, N. J. in 1960, was organized by the Council for International Organizations of Medical Sciences, under the joint auspices of UNESCO and WHO. The 27 active participants represented six countries.

In his introduction, McLean states that nearly all the uses that may be made of radioisotopes in this field are illustrated in the 26 papers presented. Among the investigations reprinted are those on such widely diversified subjects as teeth, bone tumors, bone grafts, parathyroid action, rickets, and calcium metabolism, some in vivo and some in vitro. The radioisotopes considered are P^{32}, Sr^{85}, Sr^{90}, Ca^{45}, Ca^{47}, S^{35}, H^{3}, C^{14}, Na^{22}, Na^{23}, Na^{24}, I^{131}, K^{42}. Both formal papers and the discussion that followed are included. A bibliography at the end has some 450 entries. (M.G.)

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