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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

Editors:

P. S. Baker

A. F. Rupp

R. H. Lafferty

M. W. Gerrard

UNITED STATES
ATOMIC ENERGY COMMISSION
OAK RIDGE, TENNESSEE

This document has been reviewed and it has been determined that it does not contain Restricted Data or other classified defense information as defined in the Atomic Energy Act of 1954. W. F. Carroll
4-3-63 Chief, Declassification Branch

DATE ISSUED

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

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.....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

The recent report of the President's Science Advisory Committee, "Science, Government, and Information," stresses the importance of the interpretive review journal as a means of keeping our exploding scientific literature from engulfing us. To quote from this report, "Because there are so many technical reports, and most of them are unrefereed and of uneven quality, reviews of technical reports can be particularly useful as discriminating guides to the literature. The reviewer of the report literature must not only tell a coherent story; he must also serve as referee, weeding out what he believes to be wrong from what he believes to be right. Moreover, to make his review fully useful he must cover relevant open literature."

Photo
A. M. Weinberg

"Isotopes and Radiation Technology" appears at a time when we at once appreciate fully the size of the information crisis, and realize that the critical, interpretive review journal is an essential key to the resolution of the information dilemma. I believe that all those interested in isotopes and radiation technology owe a debt of gratitude to Dr. Baker and his staff for undertaking the onerous but useful task of making sense of the flood of literature in this field. To the staff of "Isotopes and Radiation Technology" I offer my best wishes for success in this most worthwhile new editorial venture.

Alvin M. Weinberg

March 4, 1963

P. C. Aebersold Comments and Photo

About the Editors

Philip S. Baker. A graduate of DePauw University with an AM from the University of Arkansas, and a Ph.D. in Inorganic Chemistry from the University of Illinois, Baker has had industrial experience with Dow Chemical Company, the Institute of Paper Chemistry, and Pan American Refining Corporation (now American Oil Co.). He taught chemistry at the University of Vermont and at Bradley University, Peoria, Illinois.

Photo
P. S. Baker

Since 1952, he has been on the staff of Oak Ridge National Laboratory, for five years as a Head of the Chemistry Department of the former Stable Isotopes Division and then successively as Superintendent of the Electromagnetic Separations Department, Superintendent of the Isotopes Sales Department, Superintendent of the Advanced Isotopes Development Department, and presently as Assistant Superintendent of the Isotopes Development Center. He has had considerable experience as writer and editor in the isotopes field and is well qualified to head up the new journal. He will specifically handle the section on Industrial Applications.

Arthur F. Rupp. The technical adviser of our journal needs no introduction to people in the isotopes field. After graduation from Purdue University as a chemical engineer, Rupp was employed in that capacity for several years, working in the fields of fermentation, industrial enzymes, and plastics. In 1943 he joined the staff of the

Manhattan Project's Metallurgical Laboratory at the University of Chicago, where his nuclear training started. Since then he has been closely associated with design and engineering of many systems and processes for handling radioisotopes. He has been a prime mover in the effort to produce and market radioisotopes -- both reactor-produced and from fission products -- for peacetime uses, having supervised and designed many procedures for processing these isotopes. His concepts for isotope processing, for waste management, and for control of contamination are in use throughout the world.

Photo
A. F. Rupp

For several years Rupp was Director of the Operations and Isotopes Divisions of ORNL; since 1957 the ORNL Laboratory Services Supervisor; and since 1961 Director of the ORNL Isotopes Development Center.

Robert H. Lafferty, Jr. A graduate of Davidson College, Davidson, N. C., Bob is an analytical chemist with a Ph.D. from Cornell University. After two years of teaching at Lehigh University, he joined the staff of the Kellex Corporation as a research chemist, and in 1945 was employed by Union Carbide Corporation's Oak Ridge Gaseous Diffusion Plant. After several years

Photo
R. H. Lafferty, Jr.

as Head of the Analytical and Fluorocarbon Section of the Chemical Research Department, he became technical assistant to the Superintendent of the Technical Division and began to assume duties which have become his assets in his present position. He was responsible for coordinating the preparation of the ORGDP quarterly report, and reviewed submissions for technical accuracy, format, and classification. He was secretary of a joint Carbide-Goodyear committee, of which A. F. Rupp was co-chairman, that was set up to study the prevention and handling of various kinds of radiation emergencies. As a Responsible Reviewer for the AEC, he has had considerable experience in surveying and evaluating technical literature; and as a member of the ORGDP Classification Committee, he assisted in the preparation of several classification guides. Lafferty will direct much of his attention toward covering the research applications of isotopes.

Martha W. Gerrard. The distaff member of the editorial group is a graduate of Agnes Scott College, Decatur, Ga., with an M.S. in Chemistry from the University of Georgia and additional graduate work at Cornell University. With a bent toward the biomedical applications, she rounds out the team.

Photo
Martha Gerrard

After three years as laboratory technician and teacher at the University of Georgia School of Medicine, Martha started specializing in technical writing and editing. She worked with McGraw-Hill Book Company as

editor for chemistry and physics books, particularly the National Nuclear Energy Series. Since 1950, she has been with Oak Ridge National Laboratory as a technical editor in the Chemical Technology Division. She took time off in 1958 to go to Geneva as an editor of the Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy. She is an avid linguist, abstracts for Chemical Abstracts in English, Russian, French, and Italian, and can manage Spanish and German in familiar fields. As you might guess, she will be helping us a great deal with foreign literature, as well as handling the sections on agriculture, biology, and medicine.

A new Quarterly Technical Progress Review, "Isotopes and Radiation Technology," 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 broadly cover the field of nuclear science and technology. The others are Nuclear Safety, Power Reactor Technology, Reactor Materials, and Reactor Fuel Processing.

In a period when there are already more publications than can possibly be assimilated, one can reasonably ask, "Why another one?" The answer is straightforward. The editors believe that they can enhance the value of existing publications by making reference to many articles, journals, and books; by correlating, summarizing, and evaluating the present efforts; and by presenting up-to-date critical review articles which include copious reference material as additional sources of information for those who desire it.

It is also felt that this new review can help fill the gaps in disseminating information relative to AEC-sponsored research and development work on isotope applications; 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 on

a worldwide basis, it can help prevent overlap of research effort and can aid in predicting isotope usage so as to point the direction for the 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.

The first few issues of I&RT will present brief status reviews of the isotope programs in industry, agriculture, biology, medicine, education, and research. A feature of a later issue will be the large-scale irradiation program being fostered by the AEC and other government agencies, with particular emphasis on Brookhaven National Laboratory's High Intensity Radiation Development Laboratory.

As time and available personnel allow, the editors hope to be able to include worldwide coverage through liaison with appropriate personnel in the IAEA and atomic energy establishments around the world. International cooperation in this respect is certainly highly desirable.

It is realized that during the early period of "growing pains" adjustments will be necessary. We shall leave undone many things which we ought to have done and will do some things we ought not to have done. Our references to relevant textbooks, articles, and authors, for example, will be incomplete for lack of time and manpower. In any

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

case, the ultimate success of the review will 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.

[redacted]

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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.

Photo

G. H. Cartledge

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.

INHIBITION OF CORROSION BY THE PERTECHNETATE ION

G. H. Cartledge
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} $\underline{f}$ 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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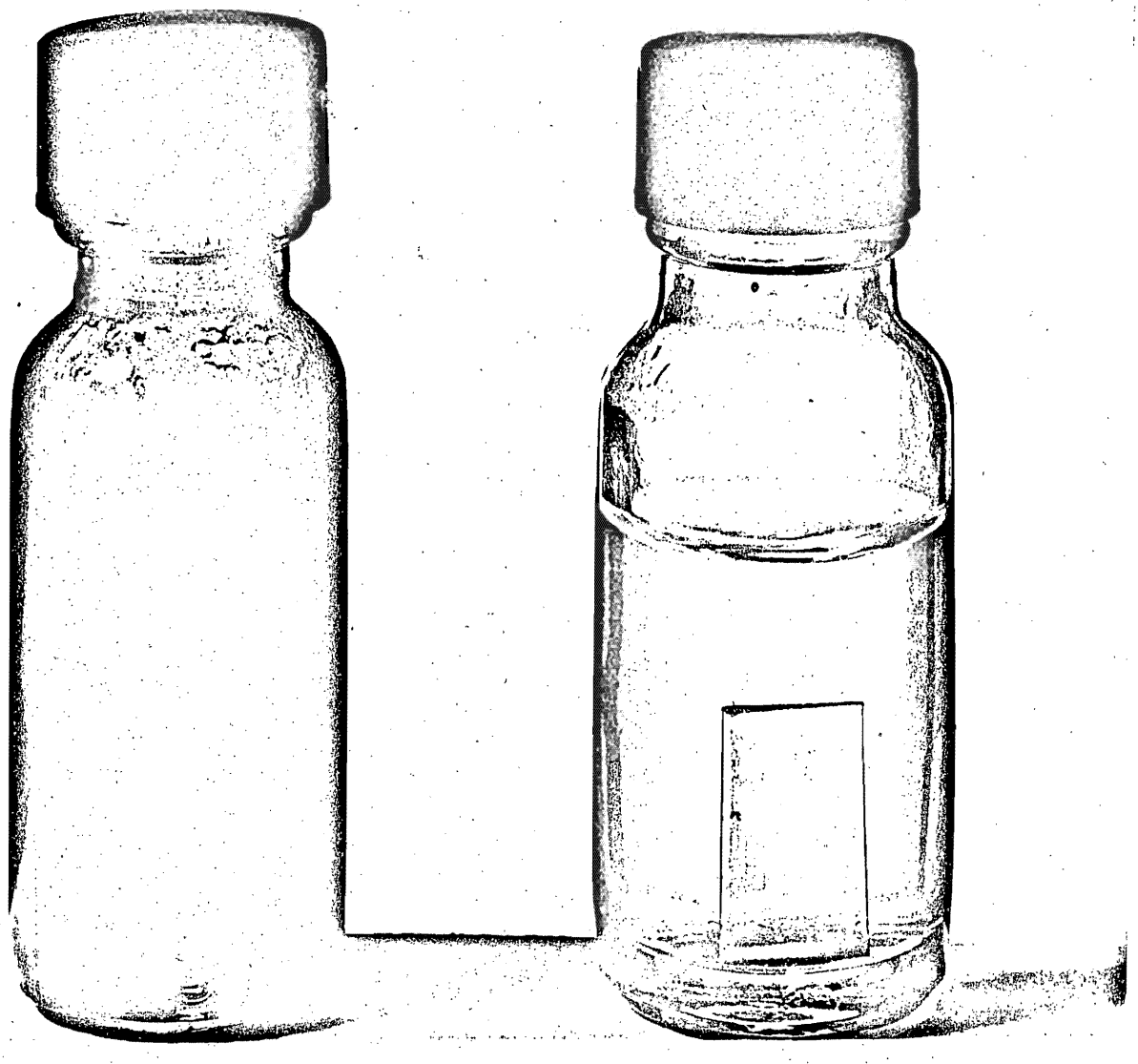
FIGURE LIST

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)].	



A B

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

10 ppm CHLORIDE ION PRESENT

Fig. 1

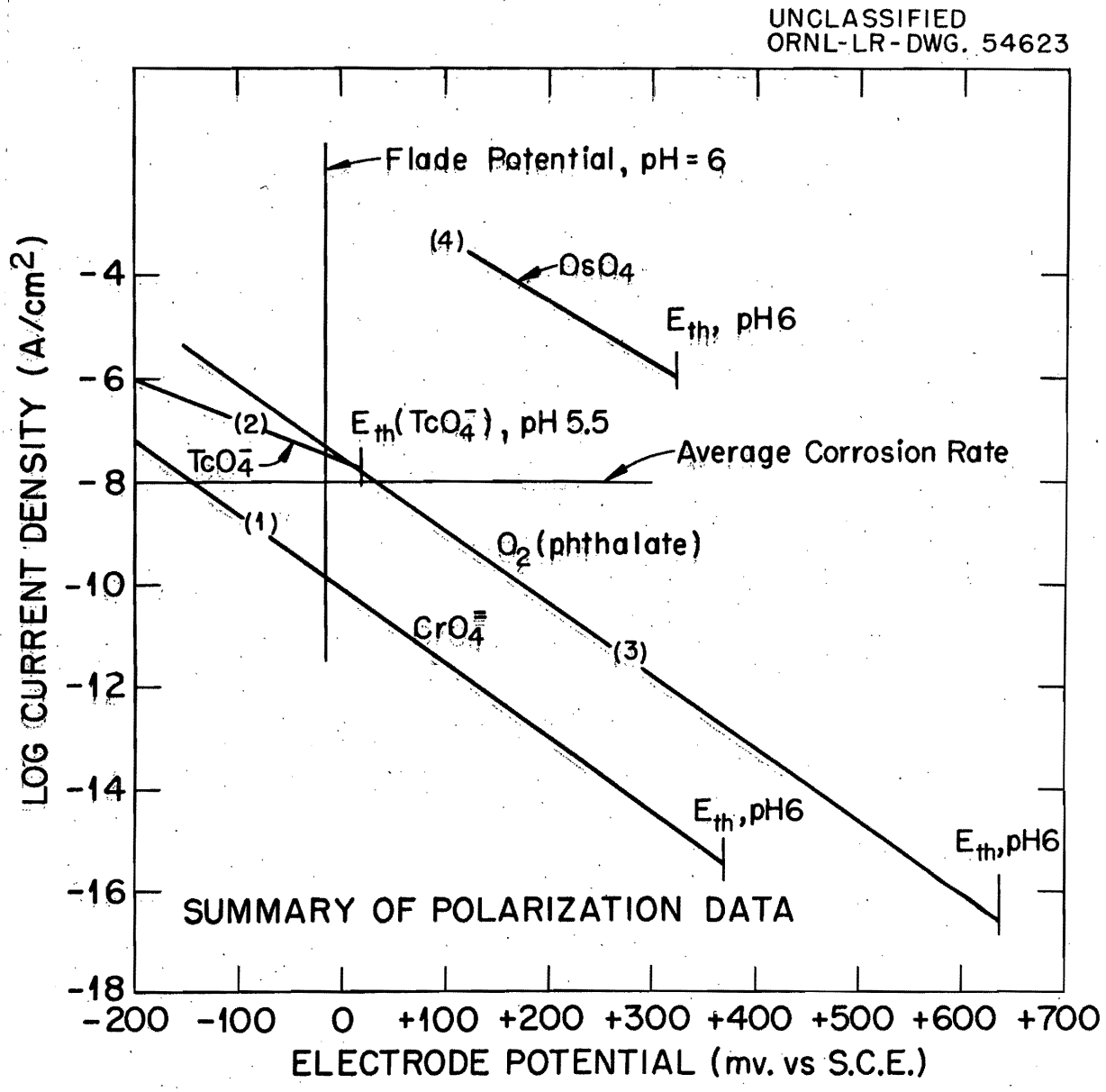

UNCLASSIFIED
ORNL-LR-DWG. 54623
Flade Potential, pH = 6
(4)
OsO4
Eth, pH6
(2)
TcO4-
Eth(TcO4-), pH 5.5
Average Corrosion Rate
(1)
O2 (phthalate)
CrO4=
(3)
Eth, pH6
Eth, pH6
SUMMARY OF POLARIZATION DATA
LOG CURRENT DENSITY (A/cm2)
-4
-6
-8
-10
-12
-14
-16
-18
-200
-100
0
+100
+200
+300
+400
+500
+600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

Fig. 2

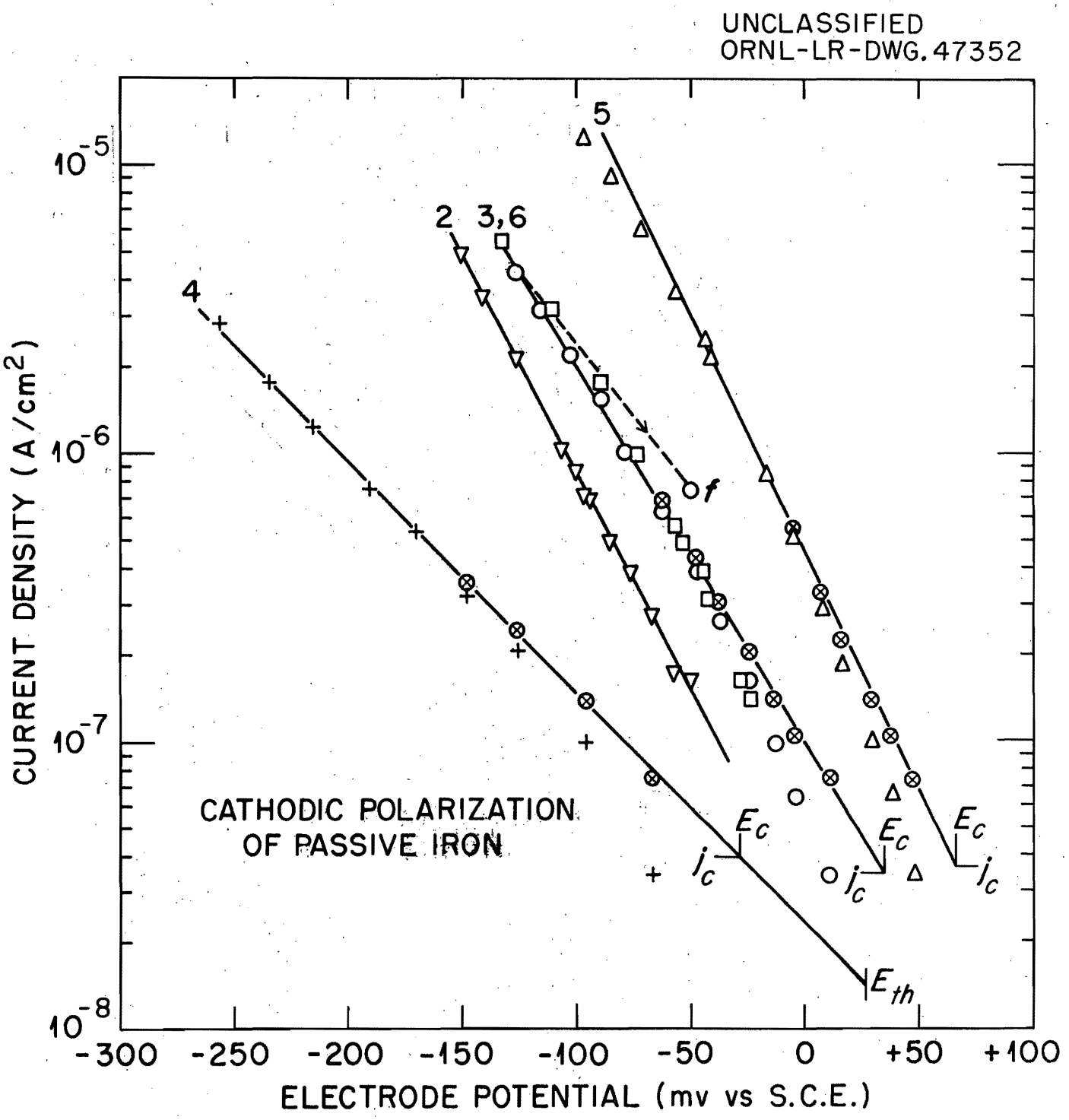

UNCLASSIFIED
ORNL-LR-DWG. 47352
CATHODIC POLARIZATION
OF PASSIVE IRON
CURRENT DENSITY (A/cm²)
ELECTRODE POTENTIAL (mv vs S.C.E.)
10⁻⁵
10⁻⁶
10⁻⁷
10⁻⁸
-300
-250
-200
-150
-100
-50
0
+50
+100
4
2
3,6
5
f
Ec
jc
Eth

Fig. 3

I. PRINCIPLES OF ISOTOPE TECHNOLOGY

As we embark on our new venture, we believe it is worthwhile to start by reviewing the present state of the art. 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]

In this issue, a brief general review of the principles of isotope production and utilization will be followed by more detailed consideration of various applications in industry, medicine, research, agriculture, and education. In subsequent issues, advances in technology and new applications will be covered but, in general, routine applications will be ignored. We plan to discuss various areas of applications. But, since there are only certain utilization principles, we also hope that novel disclosures in one area may suggest analogous techniques to users in other areas.

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]Reference 1a, p. 68.

[b]Reference 7, p.

Isotope Production

Any applications involving radioisotopes presume availability of suitable nuclides. The second issue of Isotopes and Radiation Technology will be concerned largely with production problems. For the present, let us simply mention that 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). Although the fission products comprise a spectrum of about 100 isotopes, only about 30 have sufficiently long half lives to be worth considering for inventory materials and only a few of those are considered valuable enough to process. Of those possibilities listed in Table I-5, Sr^{90}, Ce^{144}, Cs^{137}, Pm^{147}, and Tc^{99} are currently being isolated because of their satisfactory combinations of half lives, radiations, and fission-product yields. Their uses will be mentioned later.

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,γ;EC)	$Xe^{124}(n,\gamma)Xe^{125} \xrightarrow{EC} I^{125}$
(n,γ;β^-)	$Pd^{110}(n,\gamma)Pd^{111} \xrightarrow{\beta^-} Ag^{111}$

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 (Figs. I-1,2) 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-3); a patient with a malignancy, receiving therapy (Fig. I-4); food being irradiated to retard decay (Fig. I-5); or a phosphor being excited to luminescence by an admixed isotope (Fig. I-6).

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-7); gaging thickness of moving sheets of paper, plastic, and metal (Fig. I-8); controlling cigarette densities (Fig. I-9); radiographing castings and internal structures of equipment (Fig. I-10); tracing oil flow in pipelines (Fig. I-11); 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-12).

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 (Fig. I-13); it may be incorporated in a compound added to water running through a leaking pipe (Fig. I-14); it may be an active atom in sugar (e.g., C^{14}) for studying body metabolism (Fig. I-15); or it may be a label in a fertilizer compound used to study the mechanism of fertilizer uptake (Fig. I-16).

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

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1. a. Isotopes: A Three-Year Summary of U. S. Distribution, U. S. Atomic Energy Commission, Washington, D. C., August 1949, 201 pp. (5-1/2 x 9 in.), 190 references. For sale by the Superintendent of Documents, $0.45.

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

   c. Ibid., Eight-Year Summary, March 1955, 357 pp. (8 x 10 in.), 7000 references, $2.00.

2. Isotope bibliographies compiled by J. A. McCormick, U. S. Atomic Energy Commission, Washington, D. C. For sale by Office of Technical Services, Washington.

   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.

f. Radioisotopes in Agriculture: Animal Husbandry, Bacteriology, Fertilizer Uptake, Plant Physiology, Photosynthesis, and Entomology, USAEC Report TID-3516, January 1959, 67 pp., 1335 references, $1.75.

g. Utilization of Radioisotopes in Physical and Biological Sciences, USAEC Report TID-3519, February 1959, 30 pp., 492 references, $1.00.

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4. For example, the report for July-December, 1958: Atomic Industrial Progress, January 1959, 386 pp. For sale by the Superintendent of Documents, Washington, $1.25.

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.

d. The Contribution of Atomic Energy to Medicine, Hearings before the Subcommittee on Research and Development of the Joint Committee on Atomic Energy, held on June 2, 3, and 4, 1954, 194 pp. U. S. Government Printing Office (1954).

e. Report of the Panel on the Impact of the Peaceful Uses of Atomic Energy to the Joint Committee on Atomic Energy, Vols. 1 and 2 155 and 749 pp., respectively. U. S. Government Printing Office, Washington, D. C. (1956).

f. Progress in Peaceful Uses of Atomic Energy, U. S. Government Printing Office, Washington, D. C., January 1958, 463 pp., $1.25.

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

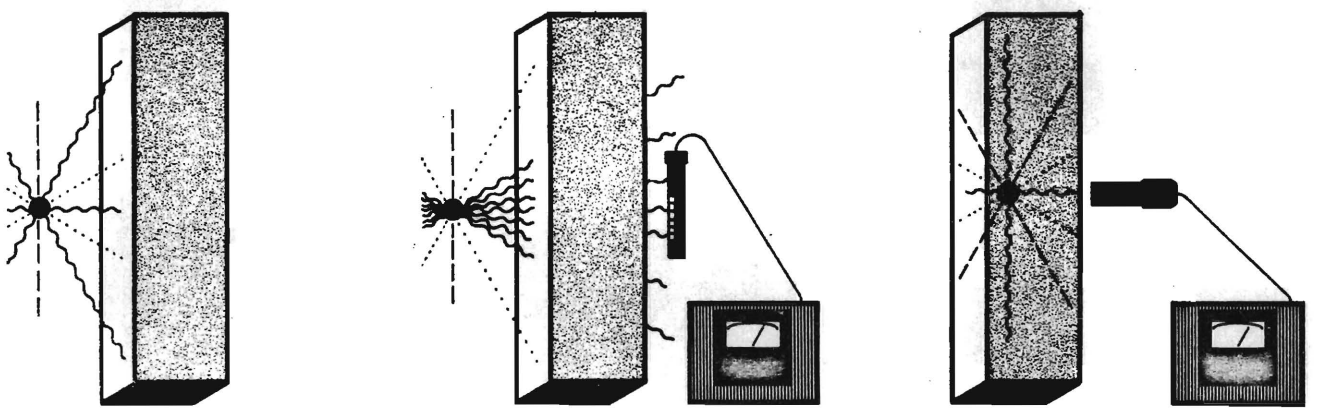
7. The Isotope Index, 6th Edition, J. L. Sommerville, Ed., Scientific Equipment Co., Indianapolis, Ind., 1962.

FIGURE LIST

Dwg. or Photo Number	Caption	Page
USAEC-ID-298A	Fig. I-1. Basic Principles of Radioisotope Utilization.	
USAEC-ID-48A	Fig. I-2. How Radioisotopes Can Be Used.	
USAEC-ID-267A	Fig. I-3. Activation of Chemical Reactions.	
USAEC-ID-200	Fig. I-4. Radioactive Strontium -- Sr^{90} for Treating Small Lesions.	
USAEC-ID-271A	Fig. I-5. Cold Sterilization.	
USAEC-ID-270A	Fig. I-6. Luminescence -- an Industrial Use of Radioisotopes.	
USAEC-ID-284	Fig. I-7. Radioactive Cobalt -- Co^{60} -- for Indicating Liquid Height.	
USAEC-ID-69A	Fig. I-8. Radioactive Source for Gaging Thickness.	
USAEC-ID-226A	Fig. I-9. Gauging Cigarette Firmness with Radioisotopes.	
USAEC-ID-59A	Fig. I-10. Measuring Thickness and Defects of Metal Castings with Radioisotopes.	
USAEC-ID-208	Fig. I-11. Radioactive Isotopes for Tracing Oil Flow in Pipelines.	
-	Fig. I-12. Weather Station Power Source.	
USAEC-ID-218A	Fig. I-13. Mechanism of Chromate-Induced Passivity.	
USAEC-ID-6A	Fig. I-14. Radioactive Iodine -- I^{131} -- for Detecting Leaks in Water Lines.	
ID-USAEC-276A	Fig. I-15. Continuous Metabolic Measurement Using C^{14}.	
USAEC-ID-179	Fig. I-16. Radioactive Phosphorus -- P^{32} -- for Study of Phosphate Fertilizer Uptake.	

BASIC PRINCIPLES OF RADIOISOTOPE UTILIZATION

RADIATION AFFECTS MATERIALS	MATERIALS AFFECT RADIATION	RADIATION TRACES MATERIALS



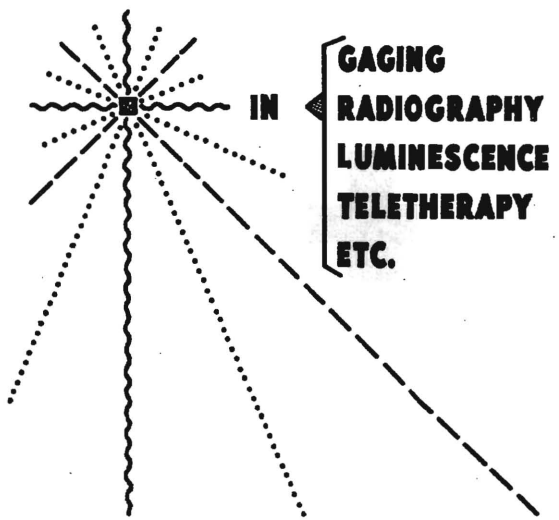
FOOD & DRUG STERILIZATION	CLINICAL RADIOGRAPHY	METABOLIC STUDIES
STATIC ELECTRICITY ELIMINATION	THICKNESS (DENSITY) GAGING	FLUID FLOW
TELETHERAPY	INDUSTRIAL RADIOGRAPHY	ABRASION & WEAR MEASUREMENT
RADIATION GENETICS	LIQUID LEVEL & HEIGHT INDICATION	STUDY OF REACTION MECHANISMS

Fig. I-1

USAEC-ID-298A

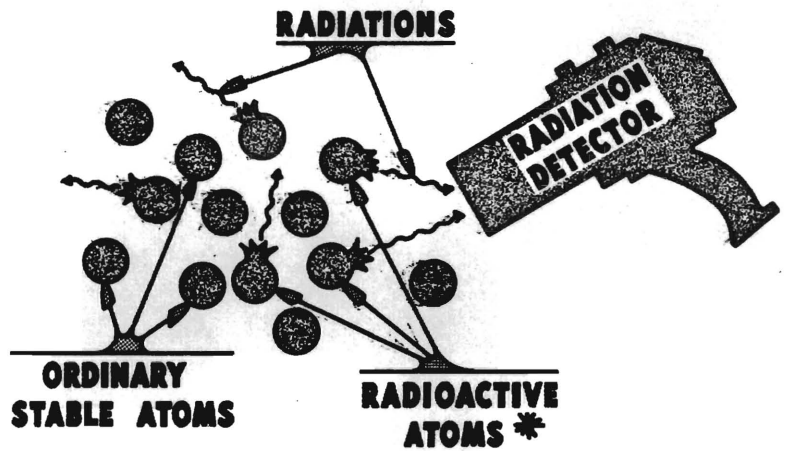
HOW RADIOISOTOPES CAN BE USED

AS SOURCES OF RADIATION



VARIETY OF RADIATIONS
LOW COST
COMPACT SOURCES OF HIGH ACTIVITY

AS TRACER ATOMS



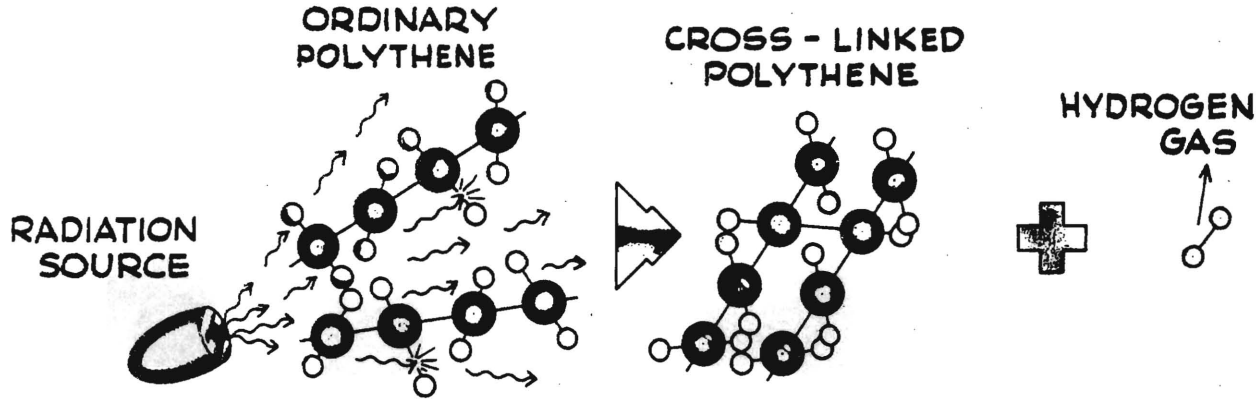
*RADIOACTIVE ATOMS BEHAVE LIKE STABLE ATOMS ——— ARE TRACED BY RADIATIONS

Fig. I-2

USAEC-ID-48A

Activation of Chemical Reactions

AN INDUSTRIAL USE OF RADIOISOTOPES



ADVANTAGES:

1 - CROSS-LINKED POLYTHENE IS:
- A - MORE HEAT RESISTANT
- B - MORE ELASTIC
- C - LESS SOLUBLE

2 - REQUIRES NO HEAT OR ADDITION OF FOREIGN ATOMS

3 - DEGREE OF CROSS-LINKING CAN BE CAREFULLY CONTROLLED

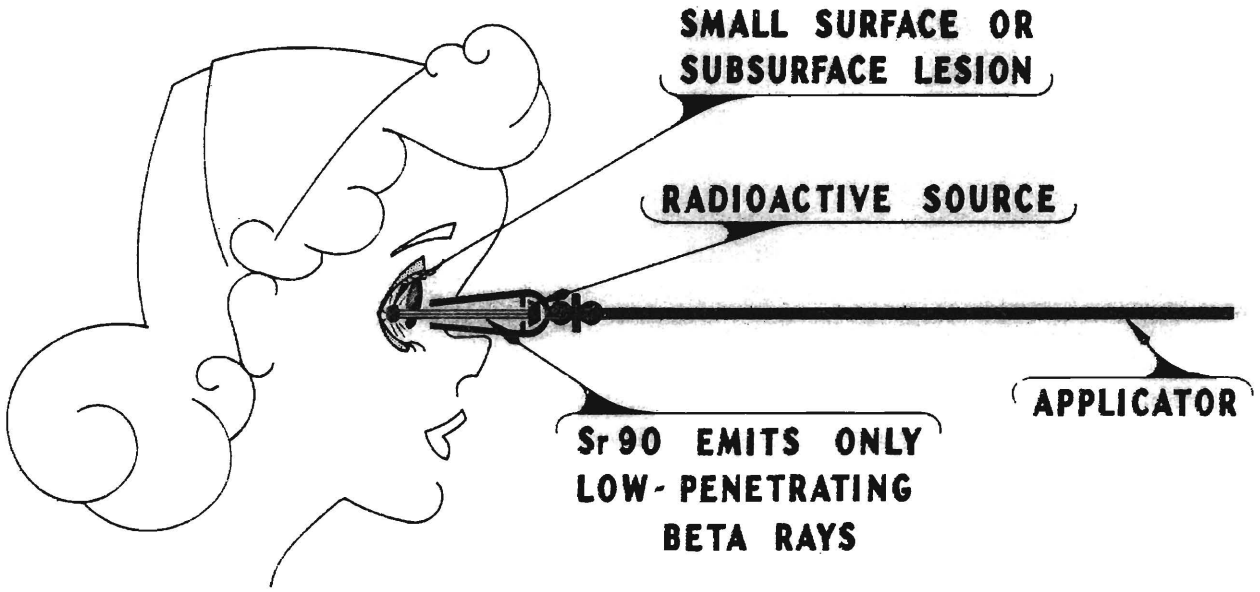
4 - MANY NEW PLASTICS CAN BE SYNTHESIZED

USAEC-ID-267A

Fig. I-3

RADIOACTIVE STRONTIUM – Sr 90

FOR TREATING SMALL LESIONS



ADVANTAGES:

1 - NO EXTRANEOUS GAMMA RADIATION

2 - REMOVAL OF BENIGN TUMORS WITHOUT SURGERY

3 - READILY ADAPTABLE TO THERAPY OF POSTOPERATIVE LESIONS

Fig. I-4

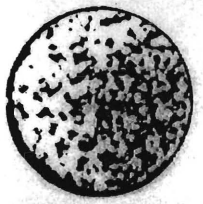
USAEC-ID-200

Cold Sterilization

AN

INDUSTRIAL USE OF RADIOISOTOPES

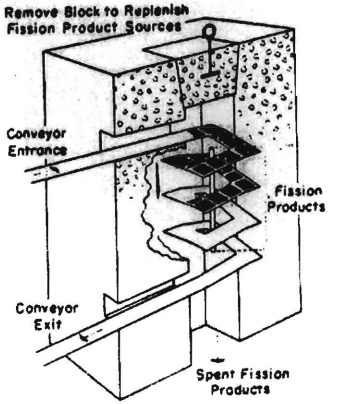
EFFECT OF RADIATION
ON MICROORGANISMS
(ASPERGILLUS NIGER)



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-5

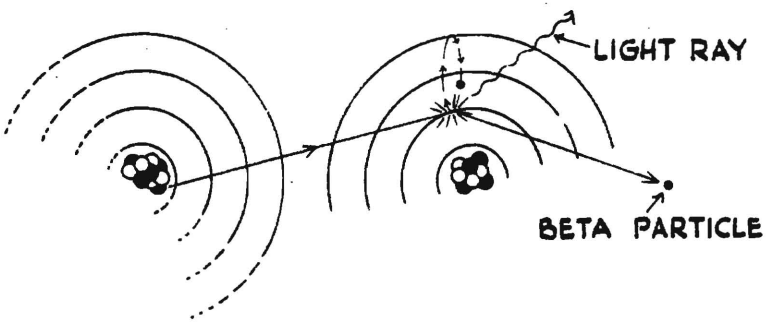
USAEC-ID-271A

Luminescence

AN

INDUSTRIAL USE OF RADIOISOTOPES

ONE MECHANISM



STRONTIUM-90 LUMINESCENT CENTER (Zn S : Cd)

RADIOISOTOPE ACTIVATED MARKERS

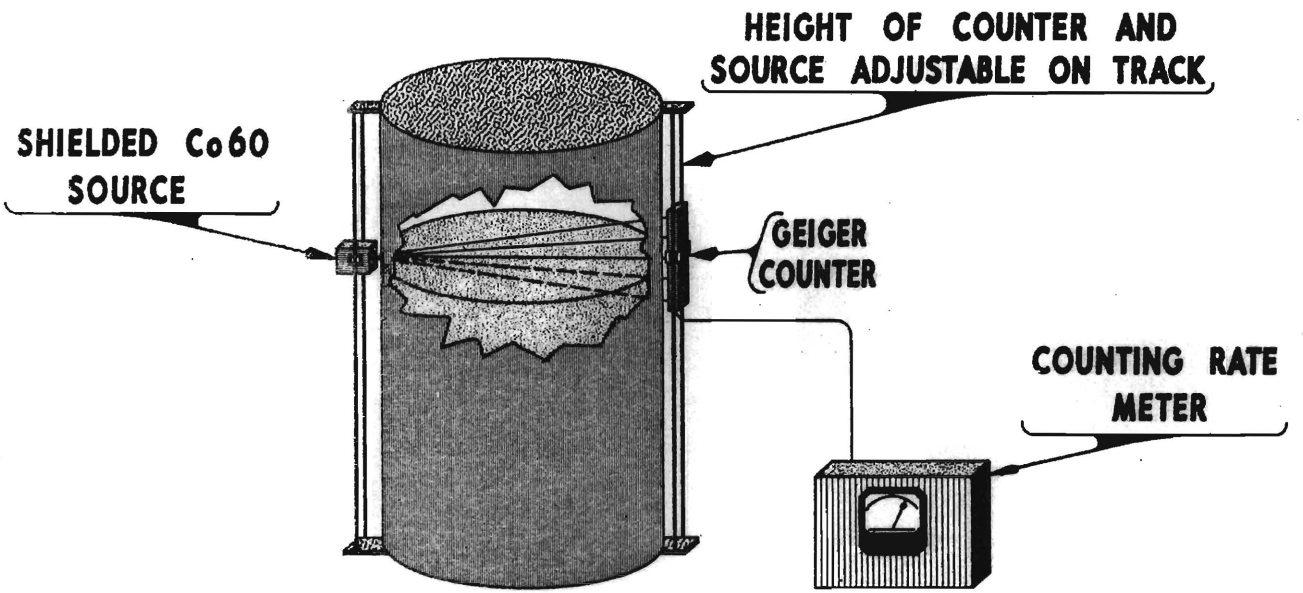
BETA SOURCES CAUSE LESS PHOSPHOR DETERIORATION THAN ALPHA — HAVE LONGER USEFUL LIFE AND LOWER EXTERNAL RADIATION LEVEL — MULTIPLE COMBINATIONS ARE POSSIBLE.

Fig. I-6

USAEC-ID-270A

RADIOACTIVE COBALT – Co60

FOR INDICATING LIQUID HEIGHT



ADVANTAGES:

1 - GAGE NOT AFFECTED BY CORROSION AND TEMPERATURE

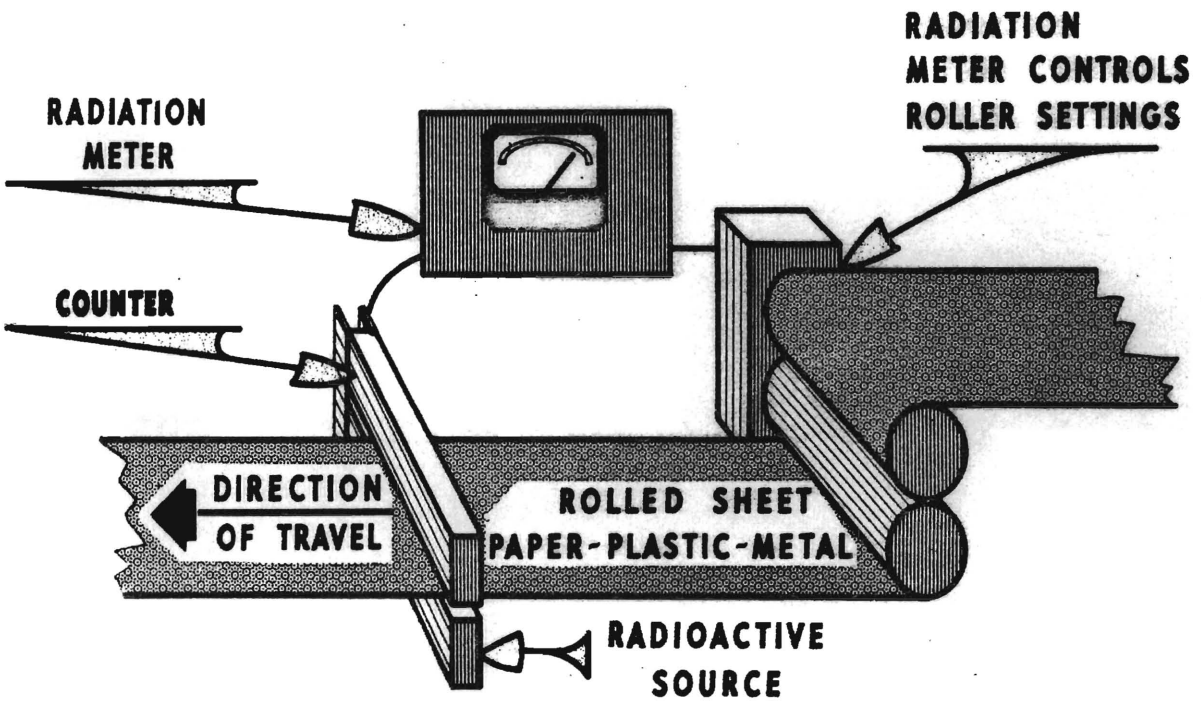
2 - CAN BE OPERATED BY NON-TECHNICAL PERSONNEL

3 - ADAPTABLE TO AUTOMATIC RECORDING AND CONTROL OF LIQUID LEVEL

Fig. I-7

USAEC-ID-284

RADIOACTIVE SOURCE
FOR GAGING THICKNESS



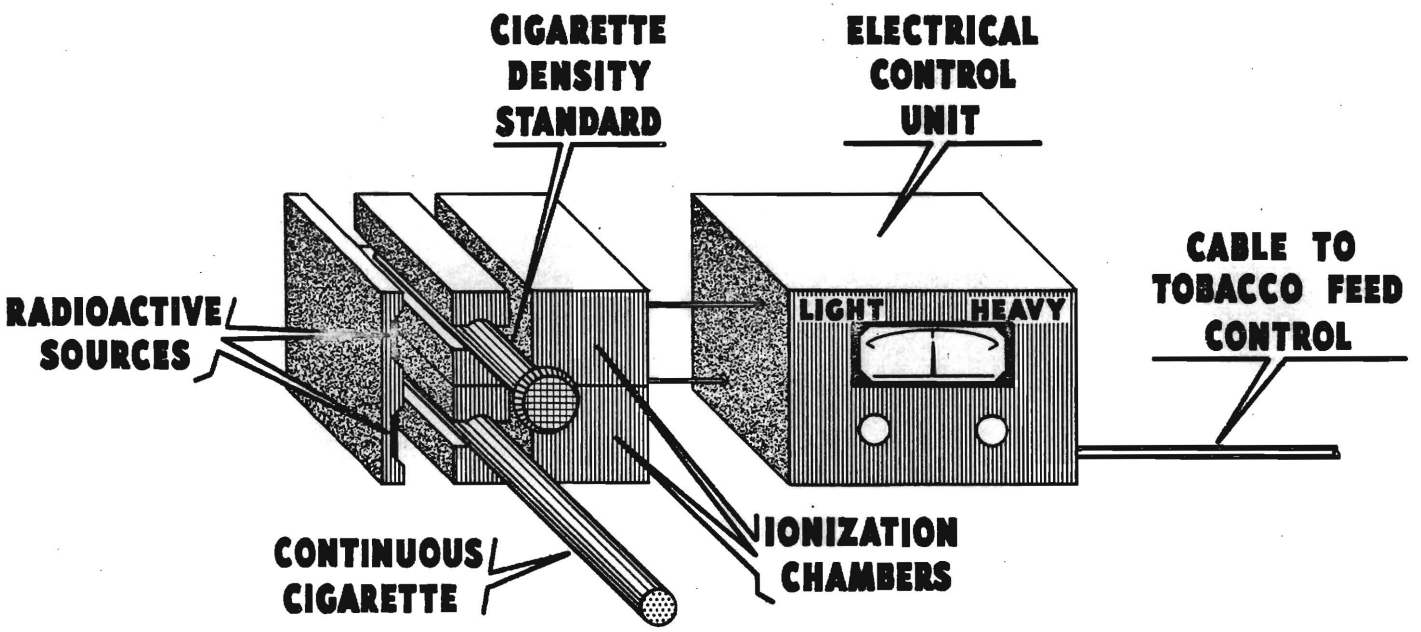
ADVANTAGES:

1- RADIATION SOURCE SELECTED TO SUIT MATERIAL
2- NO CONTACT - NO TEARING - NO MARKING MATERIAL
3- RAPID AND RELIABLE

Fig. I-8

USAEC-ID-69A

GAUGING CIGARETTE FIRMNESS
WITH RADIOISOTOPES

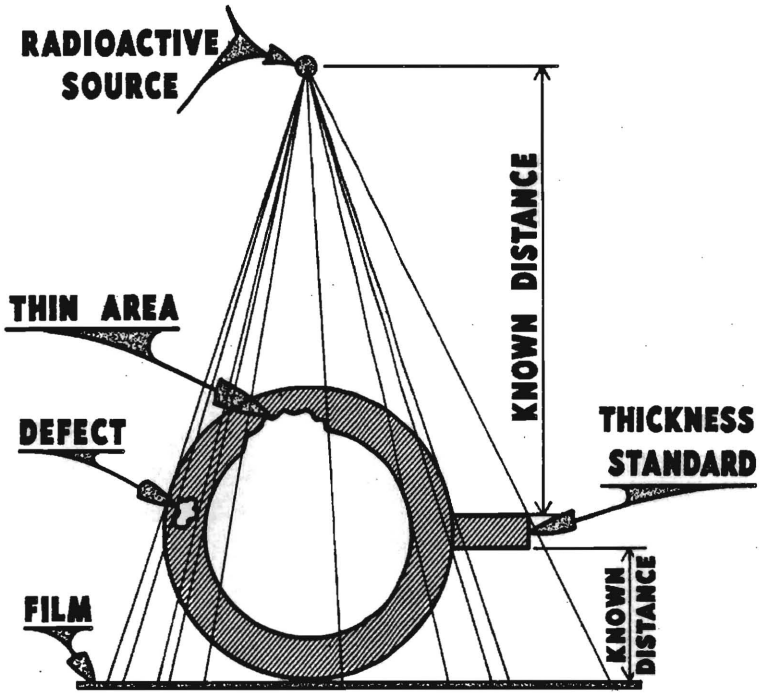


ADVANTAGES:
1 - MORE UNIFORM FIRMNESS
2 - SAVES TOBACCO
3 - ELIMINATES MANUAL CONTROL

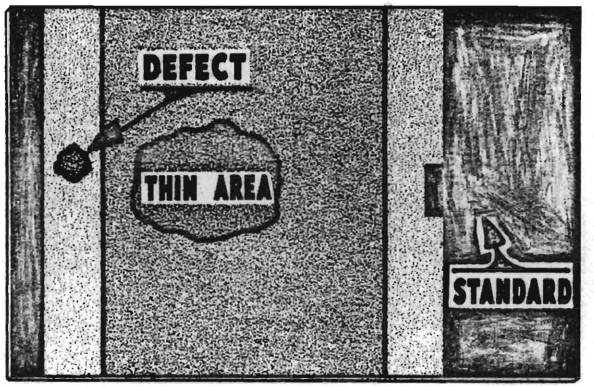
USAEC-ID-226A

Fig. I-9

MEASURING THICKNESS AND DEFECTS OF METAL CASTINGS WITH RADIOISOTOPES



RADIOGRAPH



DEVELOPED FILM

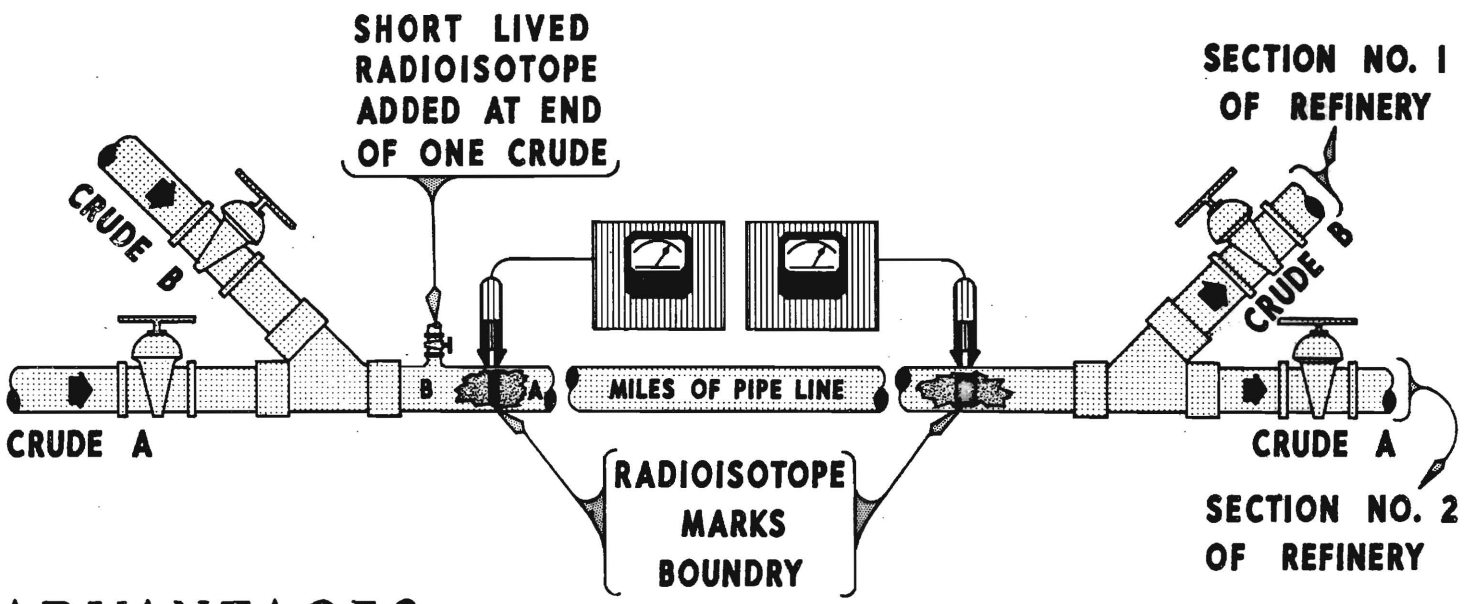
ADVANTAGES:
1 - VERSATILE AND RELIABLE INSPECTION
2 - INSPECTION MADE WITHOUT DISMANTLING
3 - VERY HIGH ACTIVITY SOURCES AVAILABLE AT LOW COST

USAEC-ID-59A

Fig. I-10

RADIOACTIVE ISOTOPES

FOR TRACING OIL FLOW IN PIPE LINES



ADVANTAGES:

1 - RADIOACTIVE "MARKER" CAN BE TRACED OVER LONG DISTANCE

2 - "MARKER" SPREADS TO ONLY SMALL OIL VOLUME

3 - PERMITS SEPARATION OF CRUDES WITH MINIMUM OF LOSS

4 - METHOD QUICK AND REQUIRES NO SAMPLING

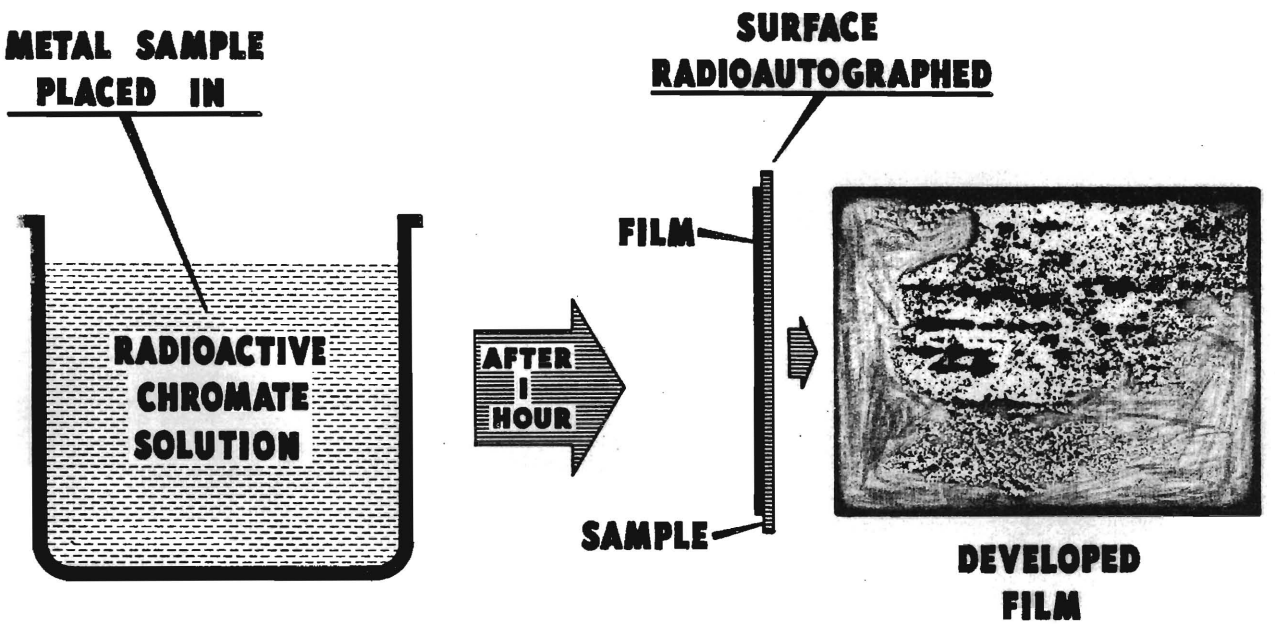
USAEC-ID-208

Fig. I-11

Cutaway of Weather Station Power Source

(See Nuclear News for August 1961)

MECHANISM OF CHROMATE INDUCED PASSIVITY



Cr 51 CONCENTRATES AT POINTS OF CORROSION—

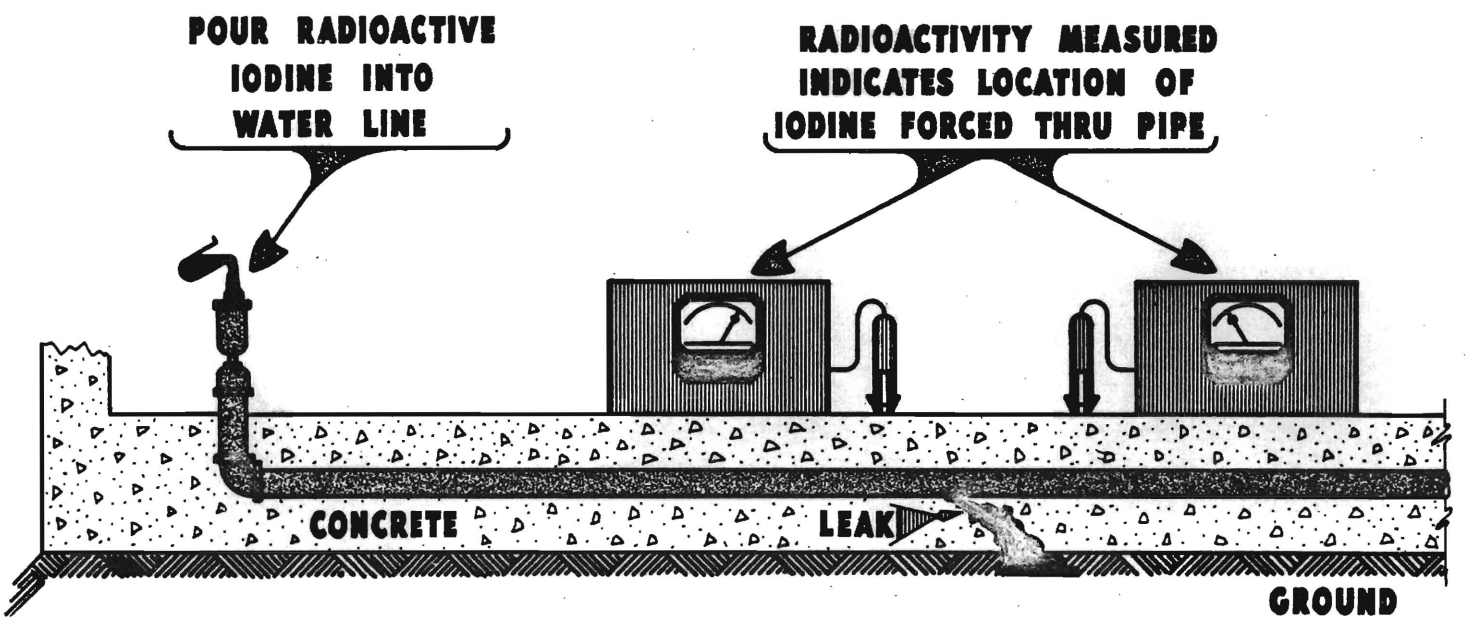
SUPPORTING OXIDE FILM THEORY

USAEC-ID-218A

Fig. I-13

RADIOACTIVE IODINE – I·131

FOR DETECTING LEAKS IN WATER LINES



ADVANTAGES:

1 - NOT NECESSARY TO REMOVE FLOORS

2 - LESS COSTLY AND MORE CONVENIENT

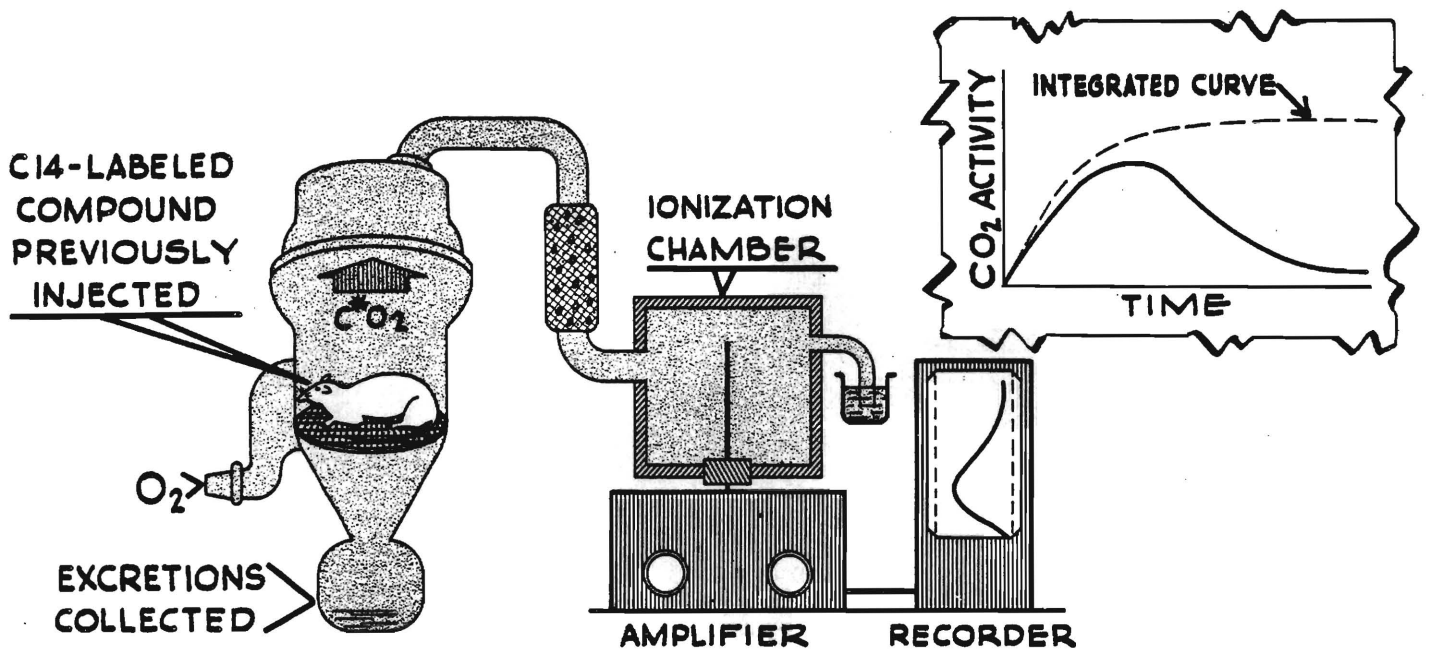
3 - SHORT HALF-LIFE --- NO RESIDUAL ACTIVITY

USAEC-ID·6A

Fig. I-14

Continuous Metabolic Measurement

USING CARBON 14



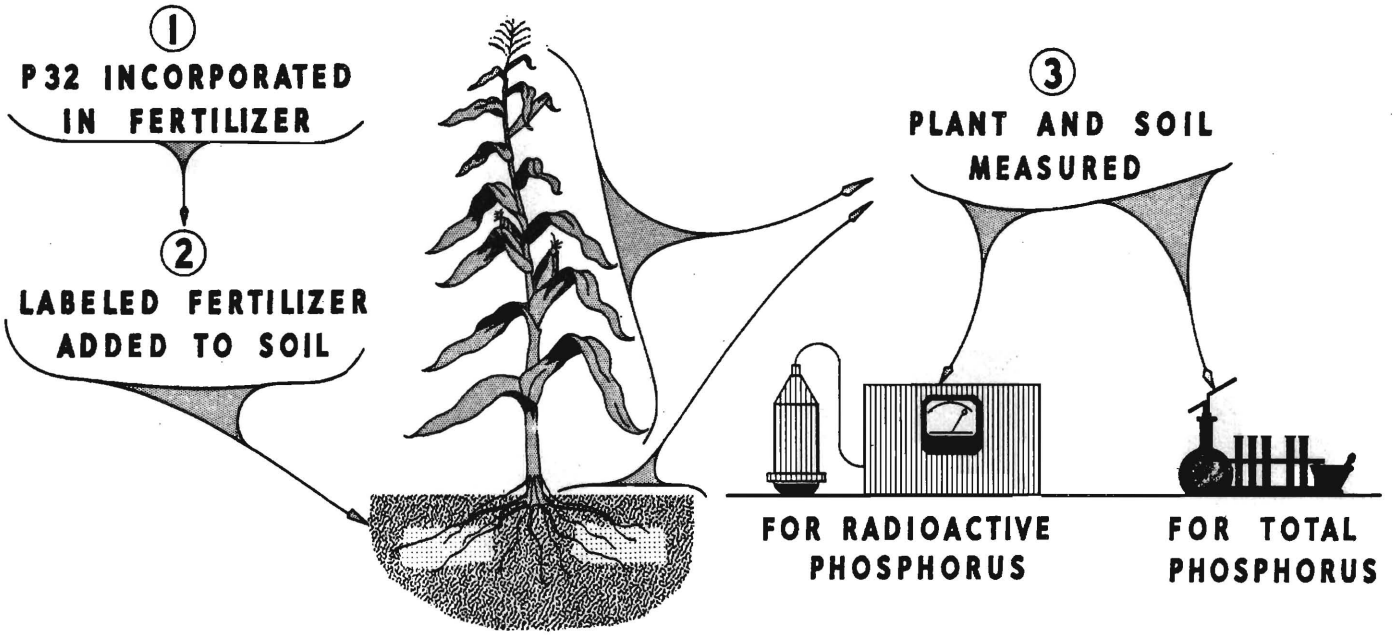
SHOWS:
1- CONTINUOUS RATE OF BREAKDOWN TO CO_2
2- RAPID METABOLISM OF MOST COMPOUNDS
3- DIFFERENCES DUE TO DRUG ACTION OR DISEASE

ID-USAEC-276A

Fig. I-15

RADIOACTIVE PHOSPHORUS – P32

FOR STUDY OF PHOSPHATE FERTILIZER UPTAKE



SHOWS:

1- FIXATION OF PHOSPHORUS BY SOIL
2- PHOSPHORUS UPTAKE BY PLANT
3- PROPER TYPE AND PLACEMENT OF FERTILIZER
4- EFFICIENCY OF FERTILIZER

USAEC-ID-179

Fig. I-16

II. APPLICATIONS IN INDUSTRY

The industrial uses of radioisotopes run the gamut from lighting up exit signs to gaging density of cigarettes, and from tracing leaks in pipelines to testing the efficiency of detergents. The point has been reached now where almost all large industries in some way or other use radioisotopes in their operation, as indicated in Table II-1.[1-6] One report[2] lists more than 800 specific industrial applications -- about half associated with production and half oriented toward research. Probably the best source of information relative to applications in the various industries is one published by the USAEC in 1962.[7]

History

The history of radioactivity as we know it goes back to the turn of the century and the work of Becquerel and the Curies in discovering the radioactivity of uranium and radium. The use of radium dials on watches and clocks was the first industrial use of isotopes and the forerunner of the present applications of such isotopes as Kr^{85} and H^{3} (tritium) in fluorescent signs. Radiography using x rays started with the work of Roentgen in 1895. But since x rays themselves are not appreciably different from the radiations given off by many radioisotopes, the ultimate use of the latter in radiography was inevitable.

It is revealing to read a section from the three-year summary of isotope distribution, published by the USAEC in 1949.[8]

> "To date, industrial utilization of radioisotopes as tracers has been largely confined to laboratory-scale investigations, by consulting laboratories or by companies that carry on active research programs. But industry already has found several ways to

Table II-1. INDUSTRIES CURRENTLY USING RADIOISOTOPES

Abrasives	Linoleum and allied trades
Agriculture	Mechanical engineering
Aircraft	Mining and quarrying
Beverages	Motor vehicles
Building	Nonferrous metals
Building materials	Oil prospecting and control
Canal, dock, and harbor	Oil refining and supply
Chemical industries	Paint and varnish
Civil engineering	Photography
Coal and coke	Plastics
Drugs and pharmaceuticals	Plating
Electrical engineering	Precious metals refining
Electrical generation	Radio engineering
Explosives	Refrigeration
Fertilizers	Rubber
Fire precautions	Sewage and effluent control
Food	Shipbuilding and servicing
Gas	Surgical instruments and requisites
Glass and glass containers	Textiles
Iron and steel	Timber and wood products
Jewelry	Tobacco
Laundering	Watches and clocks
Leather	Water

use the radiation from radioisotopes in industrial practices. These include unique devices for gauging liquid levels and for determining thicknesses of sheet material, and techniques for making radiographic inspections, all of which are generally adaptable to plant-scale use. In these uses the radioisotope is not incorporated into process material and there is no possibility of its remaining in the final market product. Furthermore, after their installation has been completed, these devices can usually be operated by people of limited training."

Obviously, some of these comments would not apply today at all. The gages used solely for determining densities of cigarettes, for example, number nearly 3000. Hundreds of other devices and applications have resulted in thousands of publications, many of them as proceedings of symposia and conferences, and large numbers of them already referenced in bibliographies.[2,9-14]

Most industrial applications of radioactivity have originated during the past fifteen years, after reactor-produced radionuclides became available. The first isotope from ORNL was C^{14}, distributed in August 1946. This happened to be for a medical application, but within a few months numerous other isotopes had been put in inventory and by 1947 industry had started using some of them on a small scale in measuring thickness of paint films,[15] in studying friction,[16] in radiography,[17] as oil field markers,[18] and in the metal-working industry.[19]

Choice of Isotope

The suitability of an isotope for any particular application depends upon several factors: (1) availability and cost; (2) appropriate

radiation characteristics, including half-life; and (3) satisfactory methods for detecting or evaluating the radiations. The fact that fewer than 10 percent of the known radioisotopes are being utilized on a routine basis is due to the cost and lack of availability of the other isotopes and not because of any undesirable characteristics.

Traditionally, the half-life of a radioisotope has been a controlling consideration in any use, since short-lived nuclides may easily become too weak too soon. However, as the sensitivity of detection methods has improved, it has become feasible to use smaller quantities of isotopes and, more important, to use radioisotopes with shorter half-lives. By combining our newer high-flux reactors with modern methods of high-speed transportation, it becomes possible to ship isotopes with half-lives of only a few days halfway around the world with sufficient residual activity to be useful. It behooves us, therefore, to examine carefully some of these hundreds of potentially available isotopes to determine whether or not many of them can be used. In many instances the availability of enriched stable isotopes as targets for radioisotope preparation will enhance the production of high-activity, relatively pure sources at rather reasonable cost.

In the past, the radiographic sources used to replace x-ray machines have consisted largely of Co^{60} because of its ready availability and comparatively low cost (as low as $1.00 per curie in large amounts), its satisfactory gamma radiations (1.17 and 1.33 Mev) and reasonable half-life (5.3 years), and the fact that its radiations can be readily detected by many different kinds of measuring instruments.

More recently, Cs^{137} has become important in industrial radiography. Although its gamma ray is lower than those of Co^{60} (0.662 Mev), its price is only 50 cents per curie in large quantities and it has a half-life of nearly 30 years. In the future it would not be surprising to see Cs^{137} replace Co^{60} almost completely because (1) Cs^{137} is a by-product from nuclear-reactor operations whereas Co^{60} has to be produced artificially; and (2) even at present prices, Cs^{137} because of its long half-life is only about one third as expensive on a comparable basis as Co^{60}, and should become even cheaper as the nuclear power program grows.

An illustration of a short-half-life isotope that is currently being used in industrial radiography is Sm^{153}, made by neutron irradiation of Sm^{152}. Although the half-life is only two days and the cost high when compared to cobalt and cesium, the radiations are particularly suitable for use in determination of turbine-blade wall thickness -- a problem for which every known measurement technique had been tried previously without success.[20]

Costs and Savings

That isotope costs are not necessarily prohibitive is borne out by a statement of Dr. Glenn Seaborg, Chairman of the USAEC, when he said that "an ounce of pure P^{32} would cost 10 billion dollars. Fortunately, important chemical and medical experiments can be conducted with a million-millionth of an ounce, costing only a penny."[21]

The savings effected by the use of isotopes are certainly very large, but also very difficult to pinpoint. The estimates vary all the way from approximately 40 million dollars per year[2] to more than

a half billion dollars per year.[5] In any case, the savings are increasing from year to year as the uses increase.

Applications

Some of the important specific isotope applications are listed in Table II-2, as well as, for a few of them, the estimated number of individual pieces of equipment licensed at the time the table was compiled.[5] As indicated in the table, by far the most widespread industrial use of isotopes is in gaging. This application usually depends upon the fact that as radiations penetrate material, the intensity of the radiation diminishes in proportion to the density and thickness of the material being penetrated. Level gaging, thickness gaging, and density gaging all fall into this category.

Level Gaging. To determine the level of the contents of a container, or to detect when the contents reach a certain level in a container being filled, the source is usually placed on one side of the vessel or container and the detector on the other (Fig. I-7, p. 38). The source and detector can be moved to locate a static level, or the source and detector can be set at a predetermined level and register the instant that the contents reach that level; in both cases, the change in density introduced by the contents is reflected in the radiation reaching the detector. Of course, in the case of the filling device, the detector can be incorporated into the filling-mechanism circuitry to shut off the flow. Likewise, the circuitry can be designed to operate a correct fill monitor (Fig. II-1).

Sometimes it is desirable to keep track of variations in liquid levels within a container. In this case, the source can be placed

Table II-2. SOME INDUSTRIAL APPLICATIONS OF RADIOISOTOPES

Application	Number
Cigarette density gages	2,700
Thickness gages	
a. Paper and allied products	370
b. Metal	160
c. Rubber and tire fabric	120
d. Plastic and adhesive	135
e. Others	150
Gages such as liquid level, moisture, H-C ratio, snow, etc.	420
Radiographic testing	520
Oil well stimulation (acidizing)	
Oil well logging	
Petroleum refining	
Pipeline oil flow	
Other applied industrial tracing	
Piston ring and similar wear studies	
Tool wear studies	
Corrosion studies	
Other industrial research	
Luminescent sources	
Miscellaneous industrial applications -- large-scale irradiation, sterilization, etc.	

vertically above or below the container with the detector at the opposite end and the radiation readings calibrated to show the liquid level. Or, the source can be placed on the surface of the liquid and the variations in intensity, based on the inverse square law, related to the level (Fig. II-2). In the petroleum industry liquid levels in refinery tankage and reaction vessels, as well as solid levels in storage bins and catalyst hoppers, are determined routinely, based on the movement of a Co^{60} "float."[22] A rather recent application of this type involves the detection of an interface in a propane storage reservoir.[23]

Levels can also be determined by a backscattering technique, using an isotope such as Cs^{134}, which depends upon the fact that the amount of backscattering from the container is greater below the level of the contents than it is above the level.[22]

Thickness Gaging. The use of radioisotopes for measuring and controlling thicknesses is well established. It makes little difference, in principle, whether the gaged material is metal, glass, paper, plastic, or cracker dough (Fig. II-3). Perhaps the unique advantage of this application is the fact that it lends itself so nicely to automation. As the detecting device measures the thickness, the instrument response can be incorporated in a servomechanism which automatically increases or decreases the spacings between the rollers to very close tolerances. It is not difficult to see how this saves material and decreases losses resulting from the product being "out of specs."

For measuring and controlling the thicknesses of coatings on paper, one technique is to combine two thickness gages -- one to measure

the uncoated feedstock, and the other to measure the coated product -- with a computer which calculates and then controls the actual difference between the two, which is the coating. This radioisotope procedure is obviously much faster than the traditional "sample and weigh" method and is also continuous and nondestructive. The close control of tolerances greatly reduces off-specification scrap.

It is also possible to determine thicknesses of thin layers by a "backscattering" procedure (Fig. II-4).[24-30] Recent development of bremsstrahlung and characteristic x-ray sources has added more variety to the possibilities for determining thicknesses.[31-33]

In general, the principal advantages to be gained from the use of radioisotopes in thickness gaging are:

1. Lower consumption of raw materials. Ordinarily, the manufacturer must set his thickness tolerances on the high side in order to be able to meet a certain minimum requirement at all times. Since the automatic radioisotope-actuated gages permit closer tolerances through better control, much material can be saved.
2. Reduced scrap. Since the isotope gage allows the equipment to get on stream rapidly, and it is not necessary to cut and weigh actual control samples, scrap is greatly reduced.
3. Increased productivity. Because of the shorter time required during start-up to get product within tolerances, actual productive time is increased.
4. Reduced manpower. The fact that the gages incorporate automatic adjustments with the quality-control measurements

eliminates the need of personnel to make manual adjustments and to carry out tedious analyses.

Density Gaging. Obviously, the measurement of thickness is not really different from the measurement of density. The first one assumes a fixed density, and the second a fixed thickness. Hence the equipment, at least in principle, is almost identical to that used for thickness gaging. By far the largest single use of any kind of gaging equipment is in the tobacco industry to control the "firmness" of cigarettes. Several thousand individual gages are in continual use in the U. S. alone. The principle is clearly illustrated in Fig. I-9, p. 40.[34] Density gages are also quite satisfactory for detecting interfaces in pipeline flow (Fig. II-5). As the second of two liquids of different density passes the source and detector, the change in absorption of radiation can be used to actuate a valve and send the second liquid in a new direction.

An unusual application of a density gage is to measure the degree of polymerization of butadiene and styrene (synthetic rubber). As the two reactants polymerize, the density changes, and the detector reading can be calibrated in terms of percent completion of the polymerization (Fig. II-6).

Without making specific reference to any individual application, we point out that density gaging has been used successfully in many different industries and in many ways. Included are sugar solutions, slurries (of lime, clay, and metal ores), suspensions, brines, bases, acids (e.g., battery acid), black liquor (in the paper industry), condensed milk, ground corn, starch, tomato paste, foamed latex,

synthetic rubber, rock wool, gasoline and other hydrocarbons, jet engine fuels, and rock wool blankets, to mention a few.

Radiography. Industrial radiography, like medical teletherapy, was well developed with X radiation long before the advent of radioisotopes. The literature on the subject is voluminous.[35] The use of radioisotopes to replace traditional x-ray sources and machines has resulted in a number of advantages, including low cost, portability, operability in small spaces, and greater effectiveness (Fig. I-10).[36-39]

The most-used isotope for radiography has been Co^{60},[6,36-39] with its 5.3-year half-life and 1.17- and 1.33-Mev gamma rays. Sources of Co^{60} of all sizes and shapes have been available from commercial suppliers for many years. A promising competitor, however, as suggested earlier, is fission-product Cs^{137},[40-42] with a 30-year half-life and 0.66-Mev gamma ray from its Ba^{137m} daughter.

Of particular interest are some of the more specialized and exotic radiographic sources that are continually being developed. Many of these have extremely high specific activities and can serve as the much-needed point sources which are so advantageous for good resolution and definition. The use of Sm^{153} for examining turbine blades has already been mentioned.[20] Considerable success is anticipated with the use of I^{125} as a source[43] (Fig. III-1, p.80). Iridium-192 ($T_{1/2}$ = 74 d, 0.1-0.6 Mev γ), because of its relatively soft gamma radiation, is particularly useful for radiography of light castings,[38,44-48] as is 127-day Tm^{170} with its 0.084-Mev gamma ray.[49-51] For measuring the thickness of concrete, Co^{60}, Cs^{137}, Ir^{192}, and Tm^{170} may all be used.[52]

An unexploited but potentially valuable type of radiographic source -- especially for thin materials -- involves secondary radiations[53,54] from such radioisotopes as fission-product Pm^{147} [55,56] and from Eu^{155} and Ce^{144}.[57] The energy of the secondary radiation can be controlled between very narrow limits by appropriate choice of matrix or backing.

In general, anything that an x-ray machine can do radiographically, some radioisotope can also do.

Tracing. As is the case in most fields of radioisotope utilization, industrial tracer techniques offer a wide variety of applications.[58] They permit nondestructive analyses, allow in-process studies by eliminating the necessity for shutdowns to obtain samples, and lend themselves to rapid detection so that such things as completeness of mixing, rates of precipitation, or mechanisms of reactions can be rapidly evaluated. The applications to wear studies are classic,[59-64] with piston rings, gears, and lathe bits being only a few of the many items that have been studied extensively. The effects of lubricants on various types of wear have been studied in great detail,[60,65] with resulting improvements to both lubricants and bearing surfaces. The wear characteristics of automobile tires have been evaluated with radiotracers.[66]

The petroleum industry has been particularly receptive to the use of radioisotope tracers in many of their operations.[22,67-72] Studies of catalyst flow,[73-75] coke formation,[76] and hydrogen exchange on catalysts,[77] made possible by the use of radioisotopes, have been very beneficial. Liquid flow rates can be controlled by the use of

added tracers.[78,79]

Interfaces in pipelines can be detected by use of a radiotracer such as Sb^{124} triphenylstibine added to one of the petroleum products being transported[80] (Fig. I-11, p. 42). As the tagged section of flow passes the detector, the fluid can be appropriately diverted.

King[72] has tabulated some of the isotopes most often used by refiners.

PROCESS	ISOTOPES USED
Radiography, mixing, inventory, trouble shooting, vapor line, sediment	Co^{60}, Cs^{133}, Ir^{192}
Catalyst mixing, inventory, stack losses	Ba^{140}, Ce^{144}, Cr^{51}, Rb^{86}, Sc^{46}, Co^{60}, Au^{198}
Liquid flow rates and mixing	I^{131}, I^{132}, Rb^{86}, Br^{82}
Heat exchanger leaks	Sb^{124}, Na^{24}, Br^{82}
Gas flow rates	H^{3}, Br^{82}, Kr^{85}
Entrainment	Ba^{140}, C^{14}, Sb^{124}, Co^{60}
Batch mixing	I^{132}

The study of laundering efficiencies[81,82] with various radiotracers has resulted in better detergents (Fig. II-7).

A specialized application of tracer technology is activation analysis.[58,83-87] The specific applications are varied, and run from checking backflows in water valves[88] to forensic use[89,90] and from dating archeological specimens[91] to studying zone melting.[92] (See also page).

Miscellaneous Applications. To try to cover all the industrial applications of radioisotopes would be completely beyond the scope of

this review. However, a few more typical uses to show the breadth of the possibilities is in order.

In many instances, the presence of static can be a very serious problem -- from the standpoint both of potential explosions, and of causing much scattering of fine particulates or interfering with the flow of fibers. Static eliminators[93-95] have almost completely solved this problem (Fig. II-8).

The use of radioisotopes in luminescent signs and markers has tremendous possibilities.[96,97] Tritium- and Kr^{85}-activated phosphors can be prepared to provide fluorescent signs that will continue to glow unattended for years. Small "lights" are contemplated to locate automobile and home door locks.

Two types of applications that are only mentioned here will be covered in greater detail in future issues of this review. One is the large-scale heat and thermoelectric sources using the decay energies from radioisotopes -- particularly fission products. The other is the use of radiations, usually gamma rays, for pasteurizing foods, sterilizing surgical instruments, effecting mutations in seeds and plants, and catalyzing chemical reactions. Perhaps the most interesting instance of the last use is the decision by Dow Chemical Company to use Co^{60} to catalyze the production of ethyl bromide -- the first application of radioactive energy to catalyze a commercial chemical reaction.[98]

FIGURE LIST

Dwg. or Photo Number	Caption	Page
AC-1-1-1059	Fig. II-1. Correct Fill Monitor	
AEC-ID-198	Fig. II-2. Floating Liquid Level Gage	
AC-1-2-1159	Fig. II-3. Automatic Thickness Regulator	
AEC-ID-220	Fig. II-4. Backscattering Thickness Gage	
AEC-ID-307A	Fig. II-5. Density Gage for Controlling Pipeline Flow	
MC-1-1-159	Fig. II-6. Density Gage for Controlling Polymerization of Synthetic Rubber	
AEC-ID-25A	Fig. II-7. Determination of Laundering Efficiency	
AEC-ID-269A	Fig. II-8. Static Elimination	

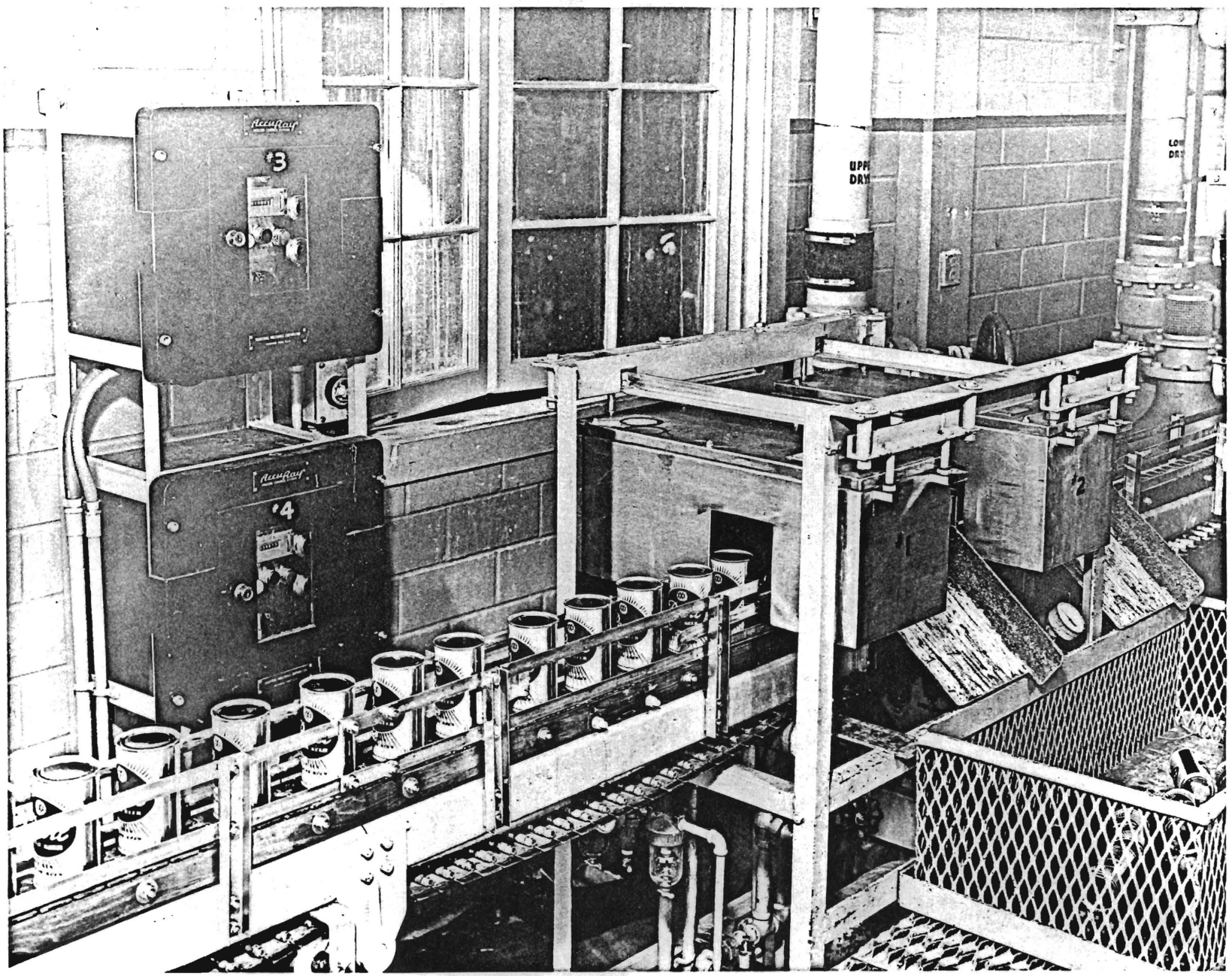
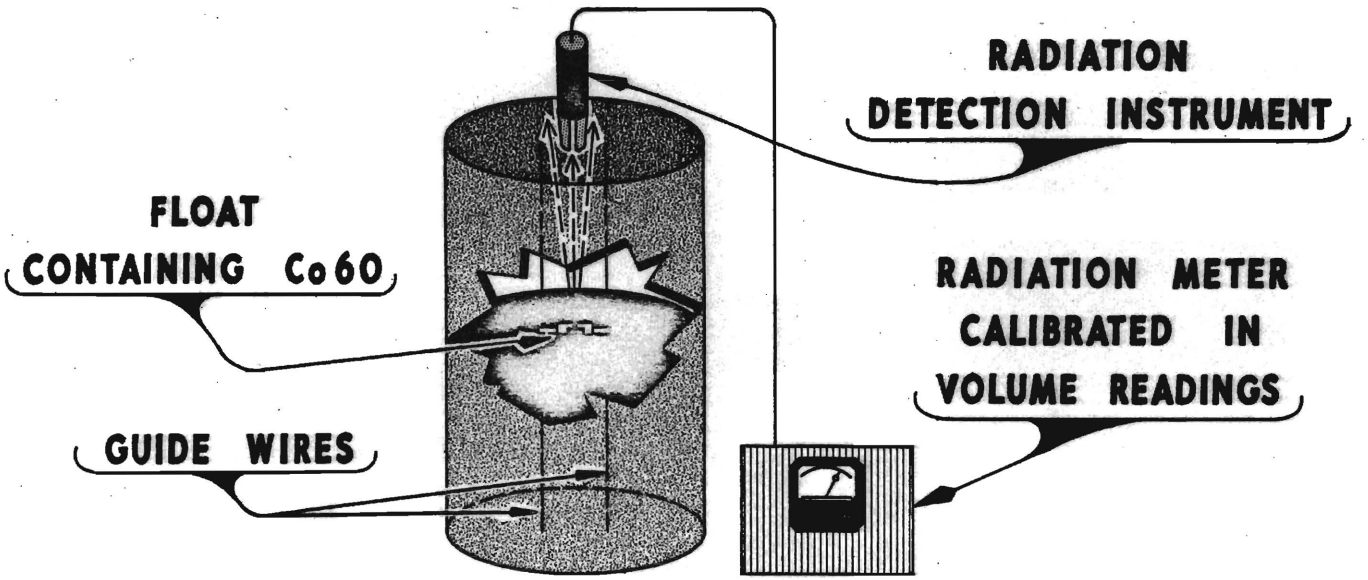


Fig. II-1

RADIOACTIVE COBALT – Co 60

FOR LIQUID LEVEL GAGE



ADVANTAGES:

1 - CONTINUOUS RECORDING

2 - MEASUREMENT MADE ON CLOSED SYSTEM

3 - ADAPTABLE TO AUTOMATIC CONTROL

Fig. II-2

USAEC-ID-198

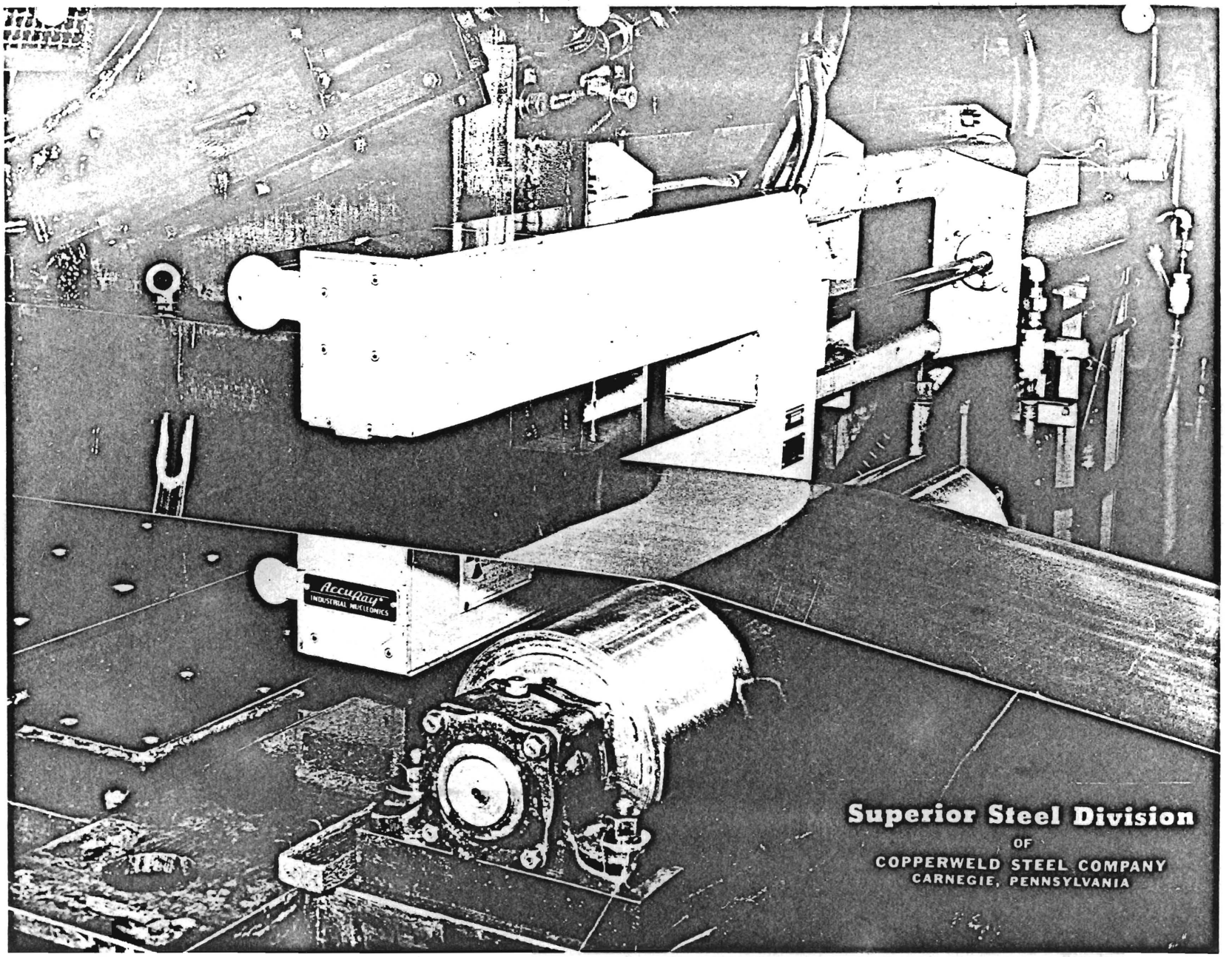
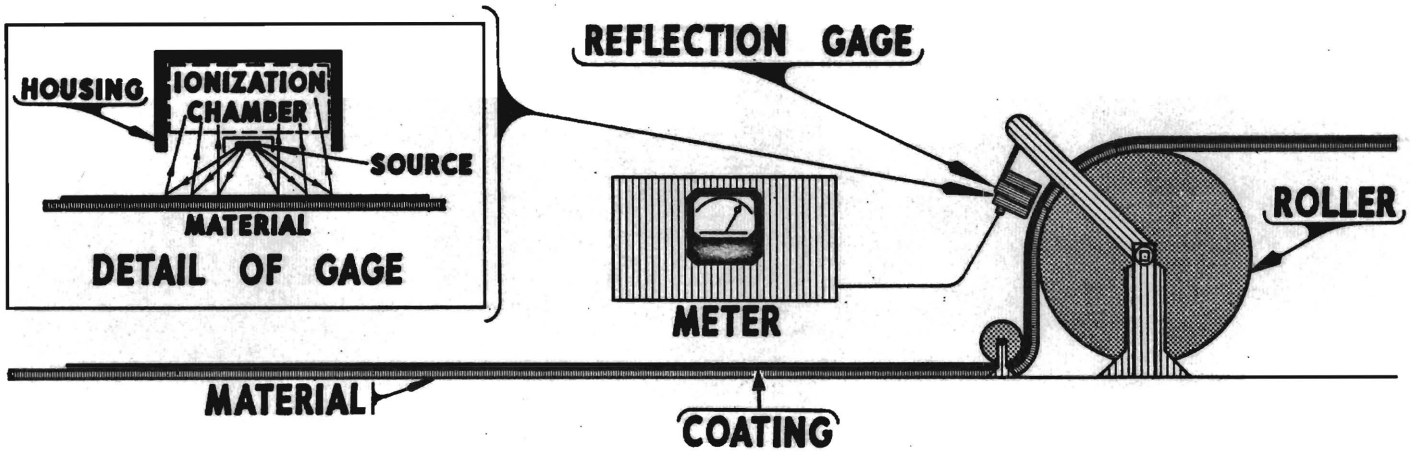

AccuRay
INDUSTRIAL NUCLEONICS
Superior Steel Division
OF
COPPERWELD STEEL COMPANY
CARNEGIE, PENNSYLVANIA

Fig. II-3

RADIOACTIVE SOURCE

FOR REFLECTION (BACKSCATTERING) THICKNESS GAGE



ADVANTAGES:

1- CAN MEASURE THICKNESS OF COATING AND/OR MATERIAL

2- MEASUREMENT MADE FROM ONE ACCESSIBLE SIDE

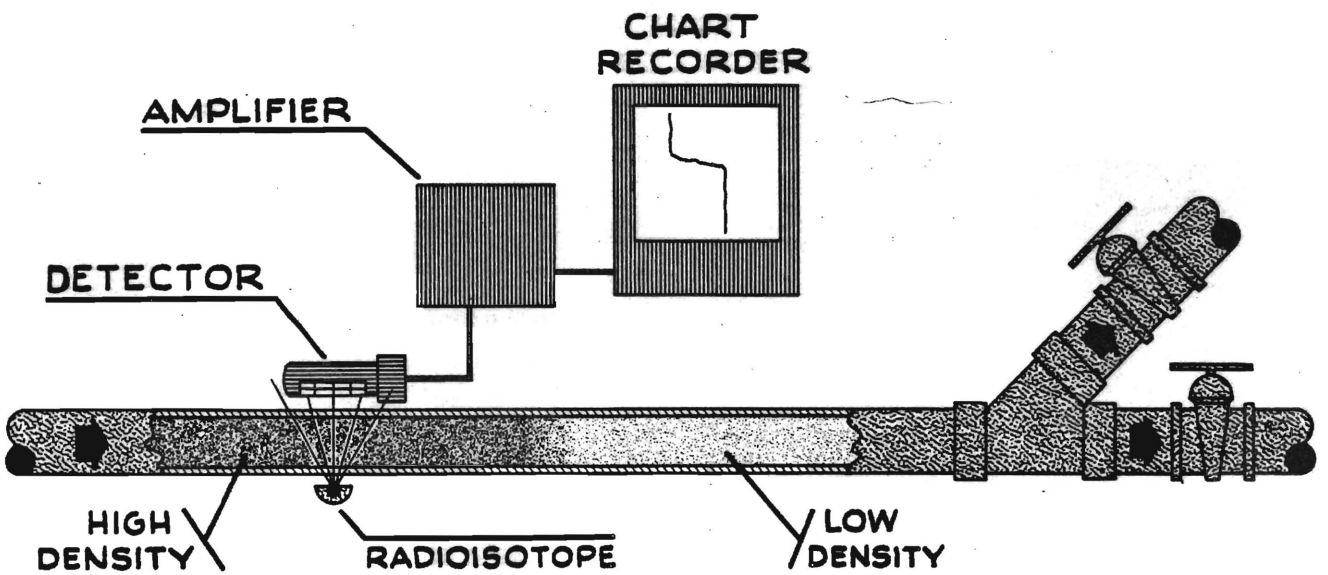
3- CAN MEASURE A VARIETY OF MATERIALS WITH ONE CALIBRATION

USAEC-ID-220

Fig. II-4

Detection of Interfaces IN PIPE-LINE FLOW

(DENSITY GAGE)



- PERMITS SEPARATION OF LIQUIDS WITH MINIMUM OF LOSS
- METHOD QUICK AND REQUIRES NO SAMPLING

USAEC-ID-307A

Fig. II-5

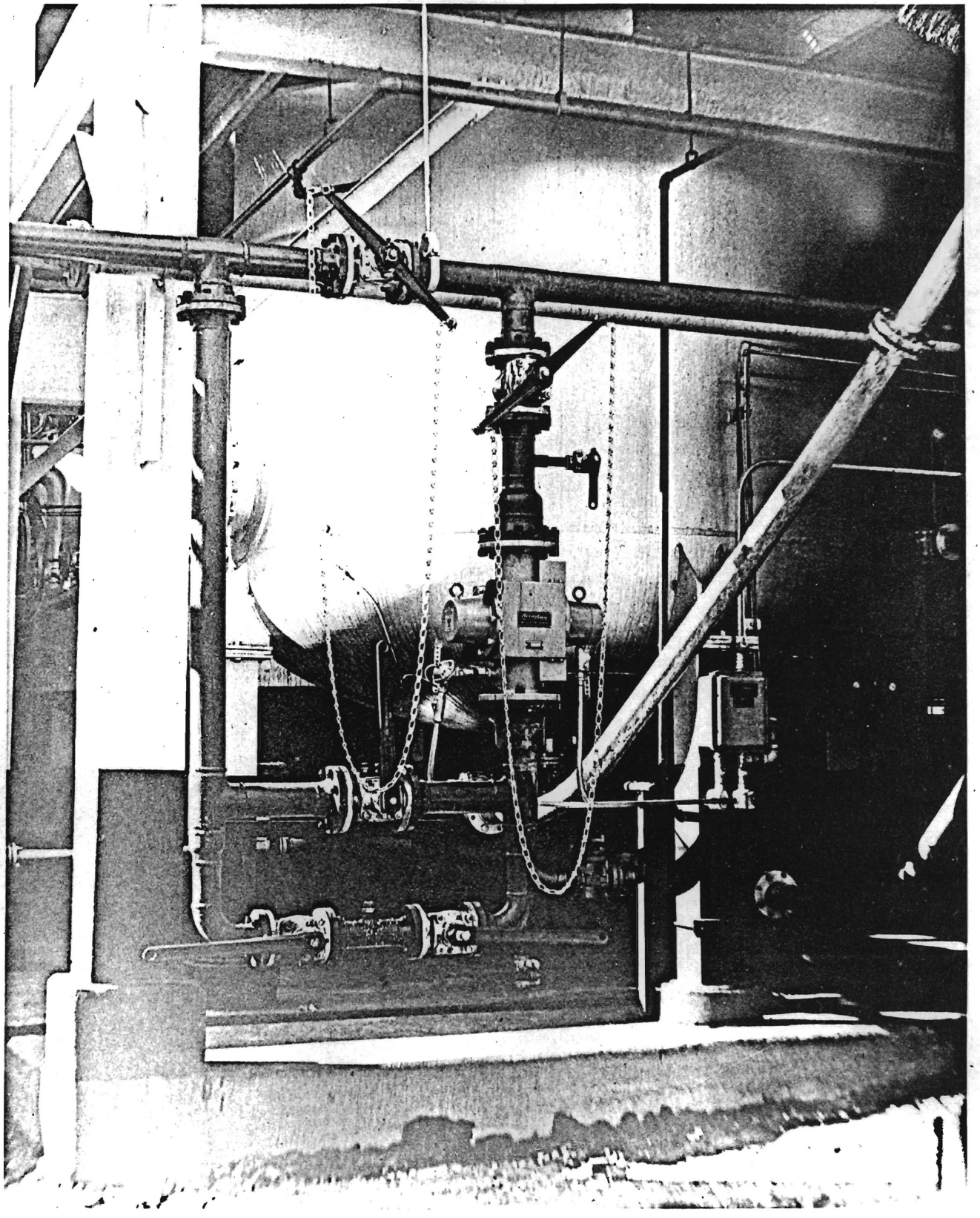
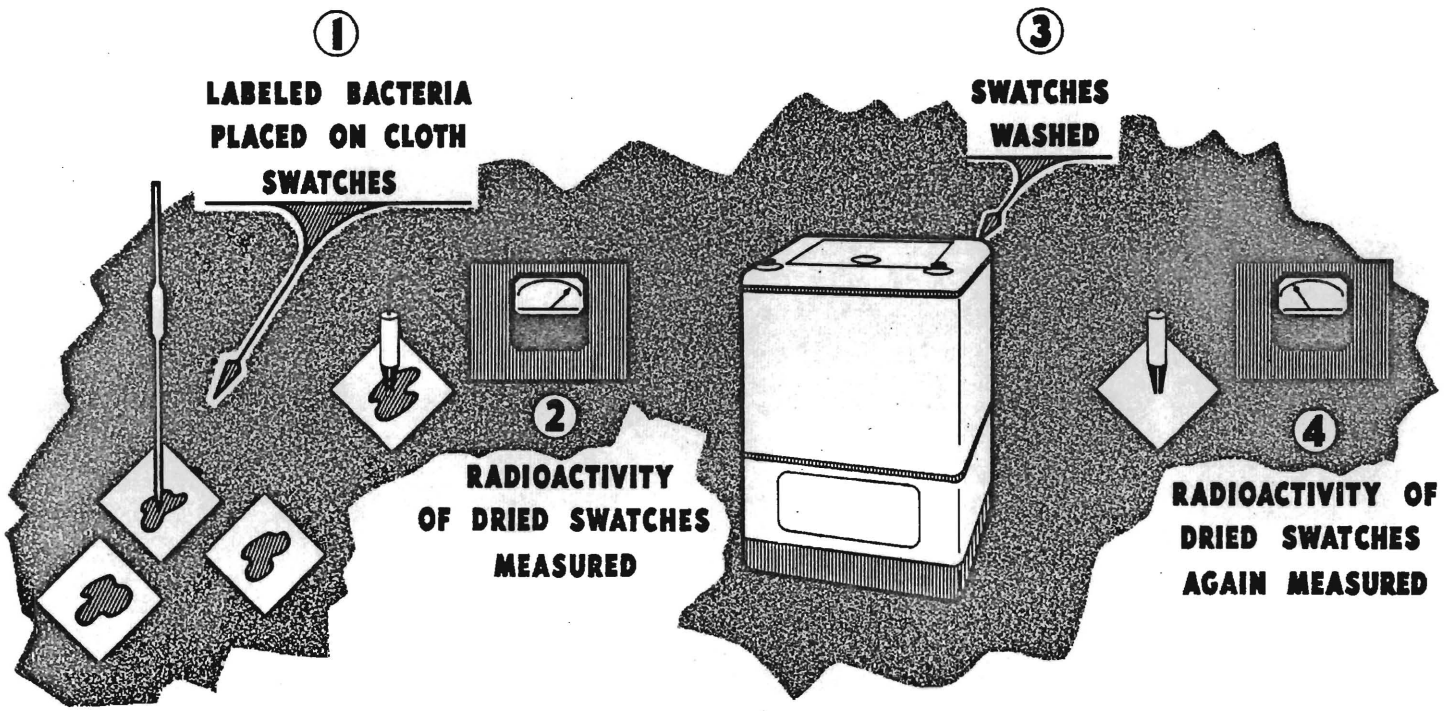


Fig. II-6

TEST FOR WASHING EFFICIENCY

USING RADIOACTIVE PHOSPHORUS - P32



SHOWS:

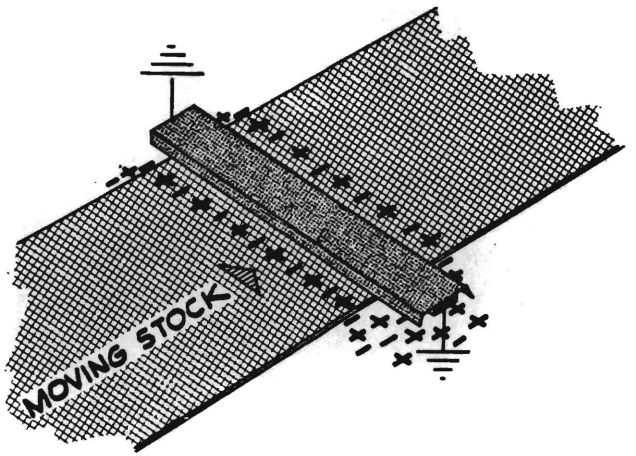
1 - EFFICIENCY OF WASHING PROCEDURES FOR REMOVING BACTERIA
2 - COMPARATIVE EFFICIENCIES OF VARIOUS DETERGENTS
3 - AMOUNT OF CROSS CONTAMINATION

USAEC-ID 25-A

Fig. II-7

Ionization

AN INDUSTRIAL USE OF RADIOISOTOPES



STATIC ELIMINATOR

RADIATION PRE-IONIZES
AIR PROVIDING EXIT
PATH FOR STATIC

USAEC-ID-269A

Fig. II-8

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III. 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 III-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

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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

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

	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

	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

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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. III-1). Exposure to both patient and technician is low because of the ease of shielding against the 27.2-35.4-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

TWO PHOTOGRAPHS OF HAND, TAKEN WITH IODINE-125

Fig. III-1 a
Fig. III-1 b

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 III-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]

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.

Footnotes to Table

[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.

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 III-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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IV. APPLICATIONS IN SCIENTIFIC RESEARCH

One of the biggest and most important uses of radioisotopes has been in the field of scientific research and development. Most of the techniques used in research with radioactive isotopes were developed prior to World War II;[1,2] these methods used naturally radioactive or cyclotron-produced radioisotopes, and counting equipment was scarce and not very sensitive. But, since the availability of reactor-produced isotopes, starting in 1946,[3] and of much better counting equipment as a result of the developments of Manhattan District research, these techniques and tools are now available to almost all research organizations. The result has been an "explosion" of publications describing instances in which radioisotopes have helped to solve problems that would be much more difficult or perhaps impossible with traditional methods.

Sir John Cockcroft in his opening remarks at the Paris Conference on Radioisotopes in Scientific Research in 1957 said:

> "It is not often that a single tool or technique is the uniting factor to bring together scientists and specialists in such different fields as are here represented today. Radioisotopes have achieved this because they have unique properties as research tools and are applicable to every branch of experimental science."

This usefulness of isotopes in all branches of science has resulted in a series of meetings in which many scientific disciplines have been represented. In 1951, the first international conference on the subject of radioisotopes in scientific research and development was held at Oxford, England.[4] Other conferences were held at Oxford in 1954,[5] in Paris in 1957,[6] in Copenhagen in 1960,[7] and in Mexico City in 1961. Many papers were also presented at the two Geneva Conferences on Peaceful Uses of Atomic Energy in 1955[8] and 1958.[9] The increasing interest in

this subject is shown by the number of papers presented at the four isotope conferences:

	Biology & Medicine	Physical Sciences	Total
Oxford 1951	67	31	98
Oxford 1954	46	24	70
Paris 1957	111	118	229
Copenhagen 1960*	-	145	
Mexico City 1961	61	-	206

*Because of the number of papers, the meeting was split.

At the two Oxford conferences, the primary emphasis was on the development and refinement of techniques; but at Paris, Copenhagen, and Mexico City, most of the papers reported on the use of already developed techniques to explore a wide variety of subjects. The proceedings of these conferences provide an effective introduction to the use of radioisotopes in many fields of research and give a good survey of the large variety of available techniques using radioisotopes. A few selected uses of radioisotopes reported at these conferences are summarized below:

Physics: The first unequivocal proof of noninvariance of parity operation and charge conjugation in weak interactions was obtained from the asymmetric beta-ray angular distribution from polarized Co^{60} nuclei. (Paris, Vol. I, 737)

Botany: Up to 50% of the material assimilated by a plant leaf in the form of sugars labeled with C^{14} was shown to be directed from the leaves to the roots, where they are converted to organic acids and later to amino acids. (Paris, Vol. IV, 494)

Biology and Agriculture: The mammary gland of the cow was shown to use butyrate (sodium butyrate labeled with C^{14}) for fat synthesis. Some of the butyrate, however, is also incorporated into glycerol, lactose, phospholipids, and milk casein. (Paris, Vol. III, 155)

Biochemistry: It was shown that tetanus toxin in the rabbit is carried to the brain by the spinal cord rather than by the blood stream. The tetanus toxin was labeled with I^{131}-marked rabbit serum protein. (Oxford 1951, Vol. I, 219)

Inorganic Chemistry: The ability of an alkali fluoride to catalyze the oxidation of a fluorocarbon was measured by the exchangeability of F^{18} between the alkali fluoride and a fluorocarbon. (Geneva 1958, Vol. 20, 132)

Analytical Chemistry: The amount of silver in a developed photographic image was determined by reacting it with I^{131} and counting the film (Oxford 1951, Vol. II, 105). Silver in a film has also been determined by reaction with Cr^{51}, Se^{75}, Hg^{203}, or Ag^{110} and counting. (Paris, Vol. II, 284)

Organic Chemistry: It was proved that the iodide in methyl iodide and molecular iodine (I^{131}) exchange at room temperature (Oxford 1954, Vol. II, 33)

Physical Chemistry: The solubility of strontium sulfate (labeled with S^{35}) was shown to depend on the particle size. (Copenhagen, Vol. III, 531)

Physiology: By using blood cells labeled with both P^{32} and Cr^{51}, direct evidence was obtained in favor of the theory that in shock an appreciable fraction of the blood in the body ceases to circulate. (Oxford 1954, Vol. I, 193)

Metallurgy: The optimum method for the purification of aluminum by zone melting was determined with radioactive tracers and activation analysis. (Oxford 1954, Vol. II, 75)

Astronomy: Results of an analysis of a recently fallen meteorite for 35-day Ar^{37} and 325-year Ar^{39} showed that the cosmic ray flux is the same in the asteriod belt as it is in the earth's orbit. (Copenhagen, Vol. I, 3)

Archaeology: The gold and copper contents of 500 Greek silver coins from the fifth century B.C. were determined by activation analysis. From the results, the source of the silver was found; and from the amount of copper debasement, the economic health of the city issuing the coin could be judged. (Copenhagen, Vol. II, 261)

Meteorology: The water balance for the Mississippi basin was measured using tritium put into the atmosphere during bomb tests. It was concluded from this balance that the average residence time of water in the Upper Mississippi Valley is 15 years. (Paris, Vol. II, 634)

Geology: The age of the most ancient rocks of the Baltic shield of the Kola Peninsula in Russia was shown to be 2.5×10^9 years by means of the K^{40}-Ar method for age determination. (Paris, Vol. II, 550)

It is obviously impossible in a publication such as this to review all uses of radioisotopes in scientific research. In the sections which follow, some of the major fields in which radioisotopes are used will be reviewed and a few selected uses in other fields indicated. In this issue the major fields reviewed are physics, biology and agriculture, inorganic and analytical chemistry, organic chemistry, and geophysics. In future issues, other research fields will be reviewed.

PHYSICS

The relation between isotope technology and physics research has always been very close. The properties of enriched stable isotopes and radioisotopes that are prepared as part of the isotope program are investigated by physicists to further their fundamental understanding of the properties of matter, and new isotopes and better methods for the production of already known isotopes frequently result from these physical studies.

Isotopes now provide the physicist with much broader fields for study than they did previously; rather than 92 elements and about 280 stable or naturally occuring radioactive isotopes, they have close to 2,000 individual nuclides to be studied. The growth in the number of new isotopes is best demonstrated by the increasing size of the "Table of Isotopes," (the so-called "Seaborg Table") which has been published at intervals by the Reviews of Modern Physics.[10] In 1940, this table was 17 pages; in 1944, it had grown to 32 pages; by 1948, it was 82 pages; and in 1953, 182 pages. The latest edition, in 1958, was 320 pages.

Several groups already collect and distribute data on nuclear properties; we do not intend to duplicate their work. Since data such as half-life, type of decay, and decay energy are necessary in isotope work, however, we review briefly the available data compilations and publications in this issue, and in future issues will note new tabulations as they are published.

Data Tabulations

One of the most valuable sources of information on the location of

nuclear data is A Directory to Nuclear Data Tabulations,[11] by R. C. Gibbs and K. Way. This directory, which was issued in January 1958, included an annotated listing of about 200 tabulations of nuclear data published up to November 1957. In the 1959 Nuclear Data Tables,[12] this compilation was revised and updated to December 1958. Approximately 60 new tabulations were listed. It was updated to June 1961 in Part 4 of the 1960 Nuclear Data Tables[13] with 80 new references.

The principal group in the United States devoted to gathering nuclear data is the Nuclear Data Project, supported by the U. S. Atomic Energy Commission under contract with the National Academy of Sciences - National Research Council in Washington. The work of this group originated at Clinton Laboratories[14] in 1946 and was later formally organized under the National Bureau of Standards. In 1953, sponsorship was taken over by the NAS-NRC. The data compiled by this group have been published in a number of reports through the years,[15] and currently they are issuing data in loose-leaf form on 8-1/2 x 11 in. pages.[16] This new form, started in 1958, now supersedes almost all their earlier publications. The subscription rate for each year is $17 on paper, $20 on card stock, and the data now issued fill about eight large three-ring or post binders. This group reports all data no matter how peculiar unless it has clearly been superseded. It is their belief that unusual or odd values may sometimes be the clue to unsuspected anomalies. Since only a single value of each property can be shown on level schemes, the Nuclear Data Group selects a value which is often shown as "adopted values" in the data sheets. In the appendix to the sheets, selected values for nuclear moments, relative isotopic abundances, and thermal neutron cross sections are given.

For workers in fields other than physics, who do not need such complete data, this group has published a summary of "best" data on radioisotopes most frequently encountered. The data and references on which these best values are based are given in the appendix. This book, Radiations from Radioactive Atoms in Frequent Use,[17] covers the following elements: H^3, C^{14}, Na^{22}, Na^{24}, P^{32}, S^{35}, Cl^{36}, K^{42}, Ca^{45}, Cr^{51}, Fe^{59}, Co^{60}, Zn^{65}, Rb^{86}, Sr^{90}-Y^{90}, I^{131}, and Au^{198}. This book was intended especially for those in the biological and medical fields who are mainly concerned with the estimation of dosage. The Nuclear Data Group also issues more specialized tables which are published separately from the Nuclear Data Sheets. The 1959 and 1960 Nuclear Data Tables[15] are such publications.

A number of summaries of data are also available in more convenient ready-reference form. The General Electric Company,[18] the Harshaw Chemical Company,[19] and the Nuclear Research Center in Karlsruhe, Federal Republic of Germany[20] publish wall charts of the isotopes. These charts, which are quite similar in appearance, plot the atomic number of the nuclide against the neutron number (atomic mass minus atomic number), as shown in Figure IV-1. Each of them requires a wall space of about two feet by four feet for mounting, and the Harshaw and Karlsruhe charts are in color.

Another reference compilation intended for desk use is the Trilinear Chart of Nuclides,[21] by W. H Sullivan. This chart, rather than being made for mounting on the wall, is in a 10 x 12 in. loose-leaf binding and accordian-folded. The entire chart when stretched out is about 17 feet long. Using hexagonal shaped blocks, as shown in Figure IV-2, the

CHART OF THE NUCLIDES

KNOLLS ATOMIC POWER LABORATORY

Operated by the General Electric Company under direction of

NAVAL REACTORS, U.S. ATOMIC ENERGY COMMISSION

SIXTH EDITION—REVISED TO DECEMBER, 1961

LIST OF ATOMIC ELEMENTS

Actinium	Ac	89
Aluminum	Al	13
Americium	Am	95
Antimony	Sb	51
Argon	Ar	18
Arsenic	As	33
Astatine	At	85
Barium	Ba	56
Berkelium	Bk	97
Beryllium	Be	4
Bismuth	Bi	83
Boron	B	5
Bromine	Br	35
Cadmium	Cd	48
Calcium	Ca	[illegible]
Californium	Cf	[illegible]
Carbon	C	[illegible]
Cerium		
Cesium		
Chlorine		
Chromi[illegible]		
Cob[illegible]		
C[illegible]		
Mercury	Hg	
Molybdenum		
Neodymium		
Neon		
Neptunium		
Nickel		
Niobi[illegible]		
Ni[illegible]		

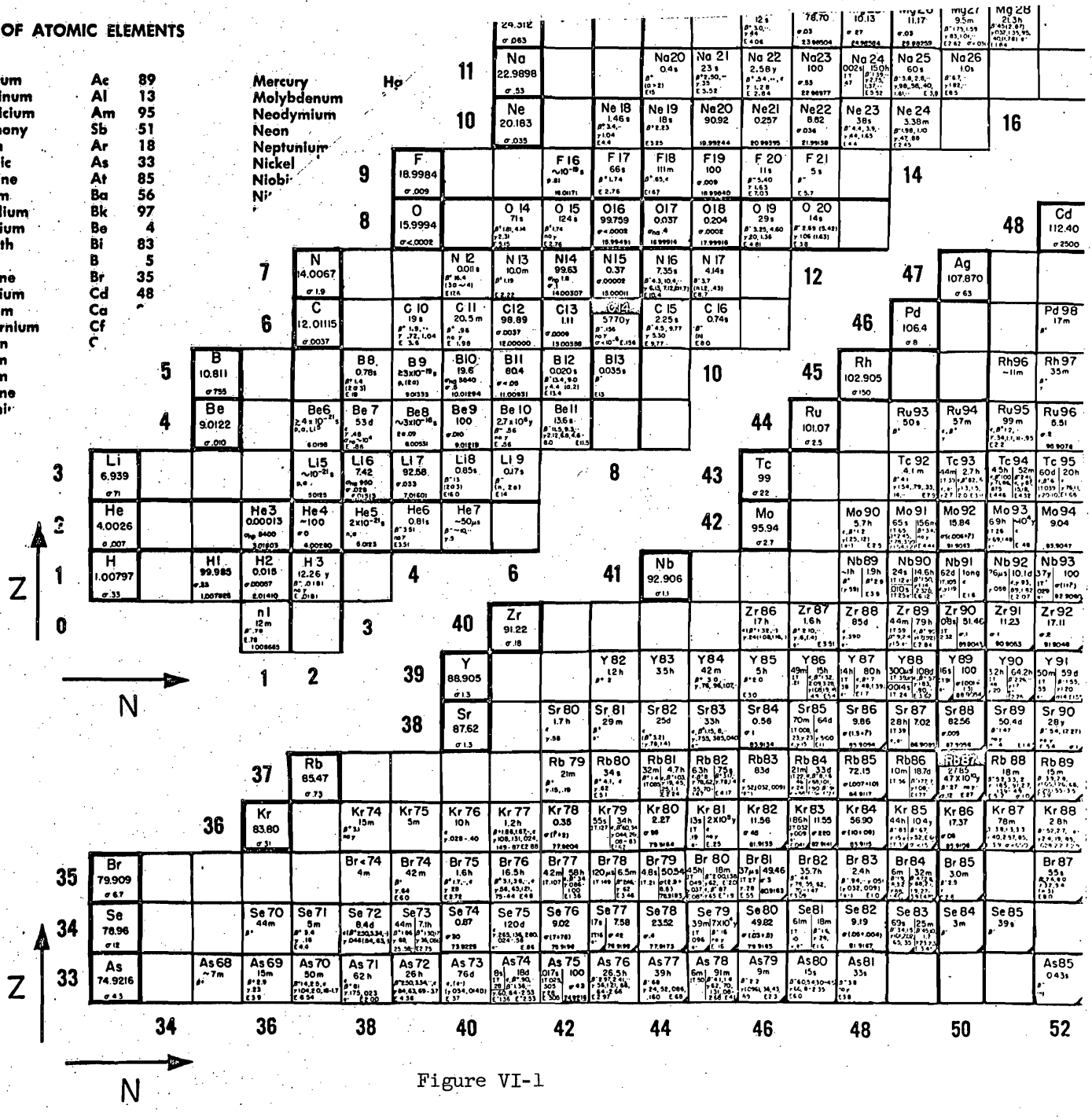


Figure VI-1

TRILINEAR CHART OF NUCLIDES

William H Sullivan

OAK RIDGE NATIONAL LABORATORY

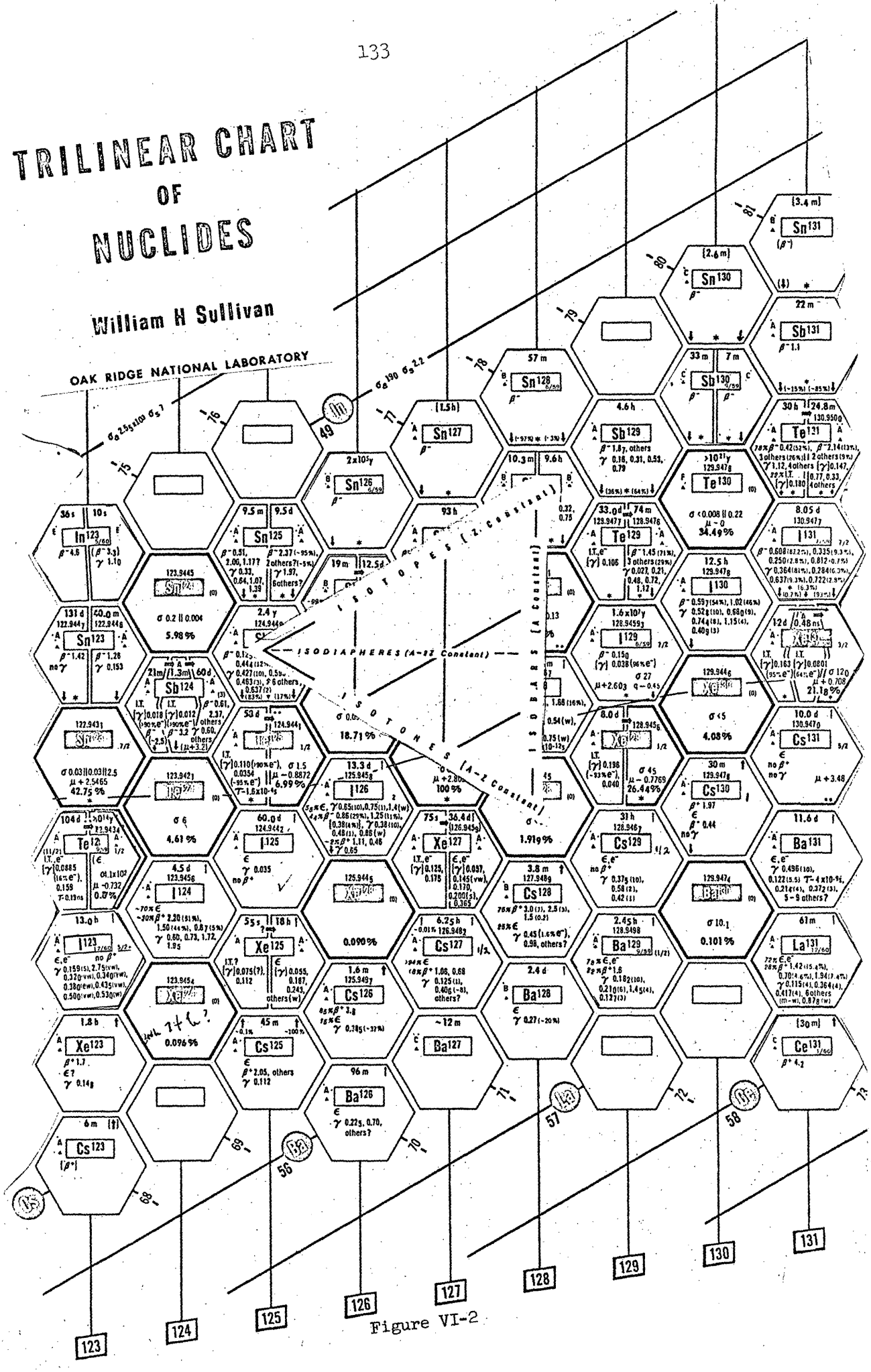


Figure VI-2

author plots atomic number against atomic weight, atomic number against neutron number, and atomic weight against neutron number, with equal emphasis. Revisions are made periodically by pasting gummed stamps over a nuclide for which values have changed.

The most up to date single-volume compilation of nuclear data is Energy Levels of Nuclei: A = 5 to A = 257,[22] published in 1961 as the first volume of the new Landolt-Börnstein series. The book, which contains three different tables, covers the literature to about June 1960. The first table is a compilation of nuclear data on elements of atomic weights 5 to 20 by Ajzenberg-Selove and Lauritsen. The second table, by Way, Gove, McGinnis, and Nakasima, covers elements of atomic weights 21 to 212. The third, by J. Scheer, covers elements from atomic weight 213 to 257. Because members of the Nuclear Data Group participated in the preparation of this book it can be expected to contain good up-to-date data.

Two other compilations cover nuclear data to about 1958. A Russian book, Decay Schemes of Radioactive Nuclei,[23] by Dzhelepov and Peker, has recently been published in a translation by D. L. Allan. An earlier edition of this book was translated by Atomic Energy of Canada, Ltd., Chalk River, in 1957.[24] Nuclear Tables, First Part, Nuclear Properties,[25] by Kunz and Schintlmeister, which also seems to have had its origin in Russia, covers data to about 1958. The introductory material is given in German, English, and Russian and the tables are in German. The second part, Nuclear Reactions, has not yet been published.

Two other tables, primarily of historical interest, are by Mattauch,[26] and by Grégoire, Joliot, and Curie.[27] The Mattauch tables were published

in 1949 and include data to the end of 1948. They are roughly equivalent in content to the "Seaborg Tables"[10] of that year. This was the last of the Isotopic Reports issued periodically by the Kaiser-Wilhelm Institut für Chemie since 1922. The Joliot-Curie report was issued in 1948 but only includes data through 1946.

The United Kingdom Radiochemical Centre issued a collection of physical properties of isotopes as Part 1 of their Radiochemical Manual[28] in January 1962. This 100-page book contains the following tables:

1. Physical Characteristics and Availability of Important Isotopes
2. Physical Data on Impurity Isotopes

3,4, and 5. The Natural Radioactive Series

6. Beta-Emitting Isotopes Grouped by Half-Life and Radiation Energy
8. Positron-Emitting Isotopes
9. Approximate Minimum Detectable Activities

It also includes sections on Production of Isotopes, Special Irradiations in Reactors, Physical Characteristics of Radioactive Sources, and Standardized Radioactive Solutions.

Neutron Cross-Sections

One of the most important nuclear properties in atomic energy work is the neutron cross-section. Several groups among the Atomic Energy Commission contractors are actively engaged in coordinating and collecting data on cross-sections. To correlate such activities, the Commission has set up the Neutron Cross Section Advisory Group. This

group is appointed on a yearly basis, makes a continuing review of the Commission's program of nuclear cross-section measurements, evaluates the need for cross-section information in the various activities of the Commission, and suggests appropriate research projects. Informal progress reports,[29] giving the status of neutron cross-section work at the various sites, are issued.

The neutron cross-section project at Brookhaven National Laboratory is divided into two parts: the Reactor Cross Section Evaluation Group which publishes periodic news letters[30] summarizing new cross-section data, and the Neutron Cross Section Compilation Group which has published a collection of neutron cross-section data.[31] It is an aim of the compilers of this work to pick the best data where this is possible. This group also published a compilation on Neutron Cross Section Angular Distributions[32] in June 1956; a new edition is due before the end of 1963. D. J. Hughes, who headed the Brookhaven project until his death in 1960, wrote Neutron Cross Sections,[33] which gives a brief explanation of neutron theory, explains how the data in the neutron tabulation[31] were obtained, and tells how to use it.

The other group in the AEC working with neutron cross-section data is under the direction of R. J. Howerton at the Livermore site of the Lawrence Radiation Laboratory of the University of California. Their compilation[34] is in three parts: (1) a tabulation of neutron cross-sections from 0.001 to 14.5 Mev; (2) a list of "best values" from Part I and a cross-section curve for each element; and (3) a tabulation of angular distributions of neutron cross-sections. The author has promised that there will be a companion volume to Part 3 in which "best data" are indicated.

BIOLOGY AND AGRICULTURE

No other field of science has felt the impact of isotopes as strongly as biology. Since isotopic tracer research techniques are very similar in biology and agriculture, this section also covers agricultural research.

History

The uses of isotopes in biology reflect the general history of the development of isotopic techniques progressing from naturally radioactive to stable to artificially radioactive isotopes. Hevesy (This is the Nobel Prize Winner who signed his early papers G. von Hevesy and later G. de Hevesy. Now he seems to prefer only Hevesy.), whose papers on radioactivity were recently published in Adventures in Radioisotope Research,[35] pioneered many of these uses. (See p.)

In 1923, Hevesy first used an isotope of lead (thorium-B) to study its distribution in the horse-bean (*Vicia faba*).[36] The following year, in cooperation with a dermatologist who was interested in the use of bismuth salts for the treatment of syphilis, he studied the distribution of bismuth (radium-E) in the rabbit.[37]

In 1934, after the discovery of deuterium by Urey, Hevesy received some heavy water from him and used it to measure the mean lifetime of water in the human body[38] and the speed of exchange of water between the body of a goldfish and its environment.[39]

To complete the picture, in 1935, shortly after the discovery of artificially radioactive isotopes, Hevesy used P^{32} that he had made to study the metabolism of phosphorus in rats.[40]

Despite these early uses of isotopes in biology and the power of the method, isotopes did not come into general use in biology until after World War II. There were two reasons for this slow growth. First, good tracers with long half-lives were not known for carbon and hydrogen, the two most important biological elements, until late 1939 when Alvarez and Cornog discovered tritium,[41] and early 1940 when Ruben and Kamen[42] discovered C^{14}; second, isotopes were not generally available at reasonable prices until production was started in nuclear reactors in 1946. The increased use of isotopes in biology since the war is reflected in the number of papers using isotopes published in the Journal of Biological Chemistry. In 1945, radioisotopes were used by authors in 1% of the reports and stable isotopes in 4%. By 1956, 39% of the papers reported the use of radioisotopes and 7% used stable ones.

In 1958 the Atomic Energy Commission published several bibliographies covering the open literature from 1948 to 1957 on uses of isotopes in a number of different fields. A brief examination of 5 of these bibliographies, summarized in Table IV-1, gives a good picture of the amount of literature on the use of isotopes in biology and agriculture.

Biochemistry Textbooks

To cover the uses of isotopes as tracers in biology and agriculture, even superficially, is too large a job to be undertaken in this review. Several good textbooks are available in this field; however, even the authors of these books emphasize that they can do no more than provide a general introduction to a very large subject.

Isotopic Tracers in Biology,[48] written by the co-discoverer of C^{14}, M. D. Kamen, was originally published in 1947. The 1957 revision of

Table IV-1 NUMBER OF REFERENCES IN AEC BIBLIOGRAPHIES ON USE OF ISOTOPES IN BIOLOGY

Bibliography	Number of References
Utilization of Radioisotopes in Physical and Biological Sciences, General Topics[43]	492
Isotope Techniques in Biological Sciences[44]	807
Isotopes in Biochemistry and Biosynthesis of Labeled Compounds[45]	2430
Radioisotopes in Animal Physiology[46]	2439
Radioisotopes in Agriculture, Animal Husbandry, Bacteriology, Fertilizer Uptake, Plant Physiology, Photosynthesis, and Entomology[47]	1335
TOTAL	7503

this book included for the first time the use of stable as well as radioactive tracers. (The original title was Radioactive Tracers in Biology.) This book has a brief review of nuclear chemistry and physics before discussing the general biochemical aspects of tracer methodology. Kamen then describes three applications of tracers in intermediary metabolism: CO_2 fixation in photosynthesis, biosynthesis of cholesterol, and biosynthesis of porphyrin. A survey of the physiological and medical aspects of tracers is then followed by chapters devoted to the biological uses of isotopes of specific elements: hydrogen; carbon; oxygen, nitrogen, phosphorus, and sulfur; and trace elements.

No review of books on the use of radioactive tracers in biology would be complete without a mention of Hevesy's book, Radioactive Indicators,[49] published in 1948. As mentioned above, Hevesy originated many of the techniques using radioactive isotopes; therefore, he is especially well fitted to write this book, which is a survey of uses of radioactive indicators in the fields of animal physiology, pathology, and biochemistry. In the preface, Hevesy states that he was "contemplating" a companion volume on the use of tracers in plant physiology. This book has not yet appeared. Hevesy gave the probable reason for this and for the fact that his book has never been revised in the preface when he said:

> "In view of the increasing use of tracer methods it is very possible that not many surveys will appear in the future, but the results obtained by means of radioactive indicators will be incorporated into textbooks dealing with the over-all aspects of the specific topics concerned. At present, however, it seems justified to offer a collective presentation of the results."

Hevesy's forecast that the isotope literature would soon become too voluminous for surveying in a single volume was accurate, and all of the later texts on the use of tracers in biology and agriculture have made no attempt to achieve complete coverage. They have given only selected examples.

Radioactive Isotopes in Biochemistry,[50] by E. Broda of the University of Vienna, was originally written in German in 1958 but was translated into English in 1960. This book starts with a review of the principles of work with radioactive substances and of radiochemistry. Slightly over half of the book, however, describes methods for the use of isotopes in intermediary metabolism. The following items are discussed: phosphorylation and photosynthesis, carbohydrates, fats, proteins, nucleic acids, and special topics such as immunological problems and biosynthesis of heme, cholesterol, and lignin.

Three other books concerned with the use of radioisotopes in biology and agriculture are briefly described in Section VI, Education with Radioisotopes: Radioisotopes in Biology and Agriculture,[51] Isotopic Tracers,[52] and Techniques of Radiobiochemistry.[53] Of these, Radioisotopes in Biology and Agriculture deserves special mention since it is the only one that is devoted primarily to agricultural research. Comar's[51] chapters on techniques for handling isotopic tracer experiments with animals and plants is particularly valuable.

Specialized Books of Biological Importance

Often in biological work autoradiographic techniques are used to find where a particular element has been deposited. Yagoda, in Radioactive Measurements with Nuclear Emulsions,[54] describes this method

with both biological and physical applications. Boyd's book Autoradiography in Biology and Medicine,[55] intended for the biologist or medical doctor, is written with the assumption that the reader knows nothing about the subject.

Now that tritium is readily available, it has become more and more important in biological work. The proceedings of a symposium held in Vienna by the International Atomic Energy Agency in May 1961, Tritium in the Physical and Biological Sciences,[56] gives a good summary of the current status of work using this isotope. The first volume of the proceedings discusses the physics and chemistry of tritium, and the second volume covers biological applications:

TRITIUM IN THE PHYSICAL AND BIOLOGICAL SCIENCES
VOLUME II BIOLOGY

Preparation of Tritiated Compounds

General Aspects of Tritium in Biological Studies

Synthesis of Tritiated Biological Compounds

Radiation Effects of Tritium

Distribution and Metabolism of Tritiated Thymidine and Related Compounds in Studying Cell Metabolism

Use of Tritiated Thymidine and Related Compounds in Radiobiology

Use of Other Tritiated Compounds for Metabolic Studies

Advances in Tracer Methodology,[57] is a collection of papers presented at an Annual Symposium on Tracer Methodology sponsored by New England Nuclear Corporation and other industrial organizations. This book contains the proceedings of the fifth symposium held in 1961, and selected papers from earlier meetings. The major emphasis is bio-

logical uses of tritium, although several other subjects are discussed briefly. It has several good papers on the Wilzbach[58] method of tritium labeling (exposure of the organic or biological compound to tritium gas).

Photosynthesis

Probably the most spectacular success of isotopes in biology and agriculture is the help they have given in solving the problem of the chemical mechanisms of photosynthesis (Figure IV-3). Calvin and Bassham have written two books, The Path of Carbon in Photosynthesis[59] and The Photosynthesis of Carbon Compounds,[60] which describe the problems which were encountered in studying photosynthesis before the introduction of isotopes and the success that has been achieved with them. The story is summarized in the following quotations from their Introduction and Prologue:

> "The separation and identification of metabolites as intermediate compounds in photosynthetic carbon reduction proved to be an almost impossible task, despite the efforts of many able investigators, as long as only classical methods of analysis were available. However, in 1939, Rubin, Hassid, and Kamen reported a method of using the tracer-element technique in following the mechanism of carbon assimilation.[59]"

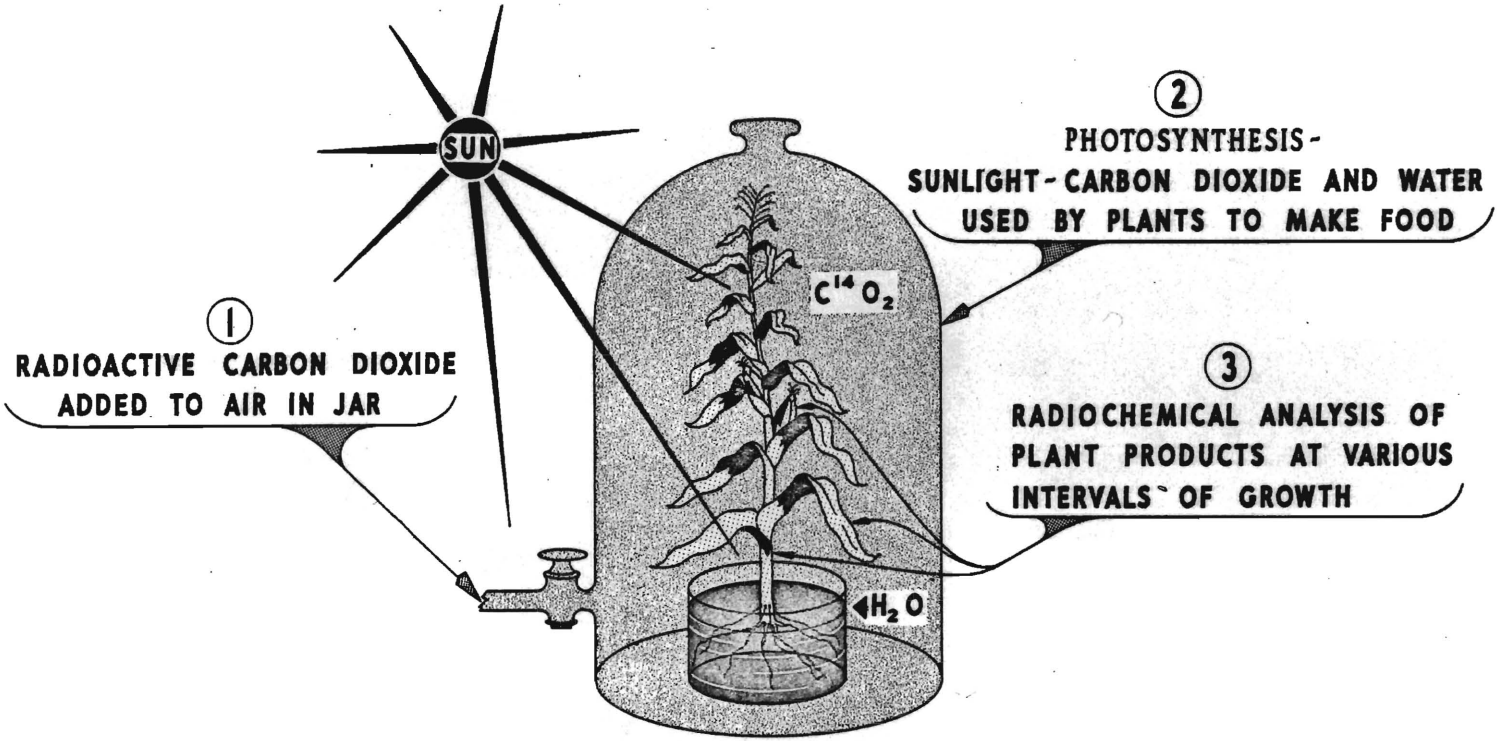
The technique of Rubin, Hassid, and Kamen,[61] mentioned in this quotation, used C^{11}, which was a great advance over classical methods; but the extremely short half-life of C^{11} ($t_{1/2}$ = 20.5 min) still left much to be desired. The story continues:

> "Fortunately, the discovery of the long-lived isotopic carbons (carbon-14) by Samuel Rubin and Martin Kamen in 1940 provided the ideal tool for tracing these synthetic routes.

Figure IV-3

RADIOACTIVE CARBON – C14

FOR STUDYING FOOD PRODUCTION BY PLANTS - PHOTOSYNTHESIS



SHOWS:

1 - RAPIDITY OF LIFE PROCESSES

2 - INTERMEDIATE STEPS IN PRODUCING FOODS

3 - ROLE OF CHLOROPHYLL (GREEN PIGMENT)

USAEC-ID-190

"In 1945, carbon-14, radiocarbon, became available in large amounts as a product from nuclear reactors. With the encouragement and support of Professor Ernest O. Lawrence, the Director of the Radiation Laboratory in Berkeley, Professor Calvin began to study the pathway of carbon reduction during photosynthesis using carbon-14 as his principal tool.

"This year (1961) Professor Calvin was awarded the Nobel Prize for his work on the assimilation of carbon dioxide during photosynthesis."[60]

This gives some idea of the importance of C^{14} in the study of photosynthesis; however, it is beyond our scope to discuss details of this work. Another interesting commentary on the impact that isotopes have had on the study of photosynthesis is given by Rabinowitch in the introduction to the last volume of Photosynthesis and Related Processes:[62]

"This volume brings to completion the review of Photosynthesis and Related Phenomena which the author rashly undertook to prepare for his own orientation in the summer of 1938. ..... Seventeen years and 2,000 printed pages later it is time to stop--even if this closure has come in the midst of rapid and promising developments in several areas covered in the narrative.

"A mistake, which proved almost fatal to the completion of the task, was to postpone the publication of the whole manuscript in 1943 when it was first finished in draft form, at a time when war had imposed a hiatus upon the progress of non-applied research. Instead only half of the manuscript was prepared for publication at that time (and actually published, as Vol. I in 1945); while the second half was set aside for final revision after the war. When in 1946, the author returned from the Manhattan District to the Solar Energy Project at Massachusetts Institute of Technology, the rapidity with which new research data began to accumulate made it difficult to digest and fit into the frame work of the monograph."

The rapid growth in work on photosynthesis was, of course, principally due to the tremendous impetus given to it by radioisotopes.

Metabolism Studies

The study of the course and fate of various elements in the metabolism of plants and animals is another important use of isotopes in biology and agriculture (Figure IV-4). The growth of papers in this field has been at least as explosive as that in photosynthesis. Because complete coverage of this large amount of work would require all of our available space, we will only note some of the books on metabolism which contain much work using isotopes.

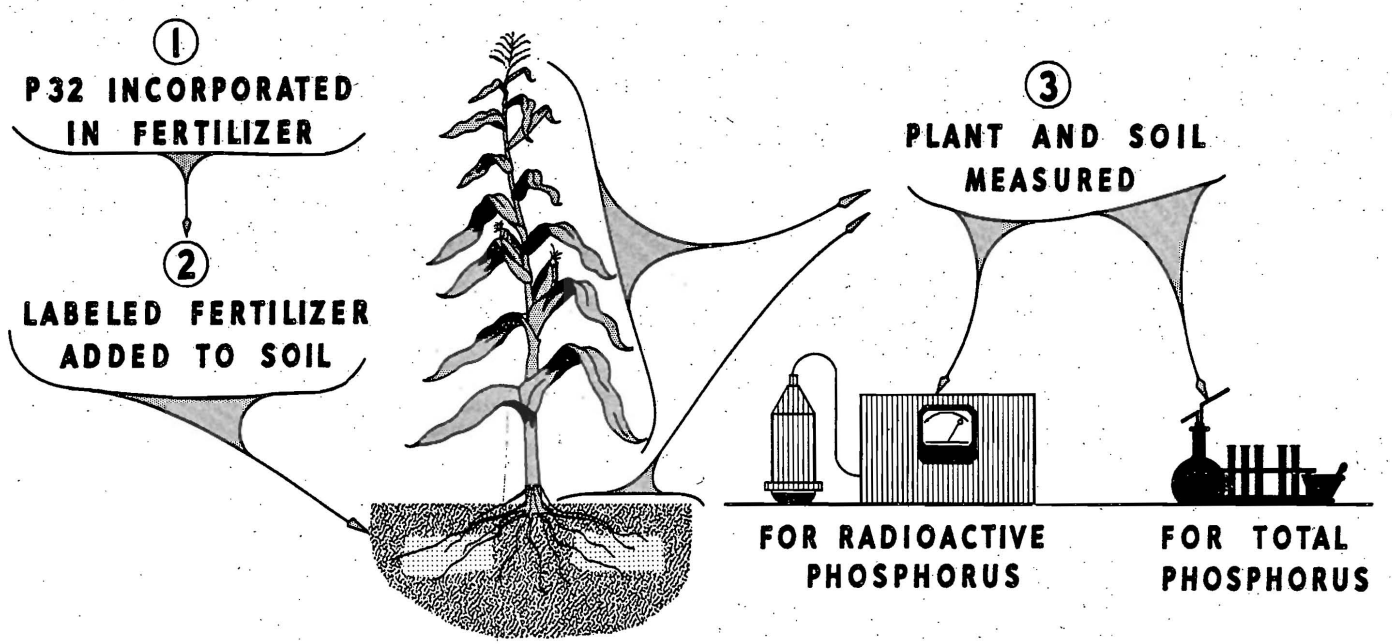
A very dramatic example of how the literature in this field has grown is in the study of mineral metabolism in humans. In 1939, Shohl's book on this subject, Mineral Metabolism,[63] was almost 400 pages long. Publication of a new 2-volume, 4-part, book on Mineral Metabolism,[64] which was intended to have the same scope and coverage as Shohl's book, began in 1960 and the last 2 parts have just been released. These 4 parts are now over 1600 pages.

In biology and agriculture, two of the most important elements in plant and animal metabolism are nitrogen and phosphorus. The metabolism of each of these elements has been covered in at least two books. In a symposium at The Johns Hopkins University in 1955, the function of metalloflavoproteins in inorganic nitrogen metabolism was discussed, and these papers have been published.[65] Another book published recently that covers nitrogen metabolism in plants[66] has a bibliography of 215 pages. This gives some idea of the amount of work being done in this field.

Figure IV-4

RADIOACTIVE PHOSPHORUS – P32

FOR STUDY OF PHOSPHATE FERTILIZER UPTAKE



SHOWS:

1- FIXATION OF PHOSPHORUS BY SOIL
2- PHOSPHORUS UPTAKE BY PLANT
3- PROPER TYPE AND PLACEMENT OF FERTILIZER
4- EFFICIENCY OF FERTILIZER

USAEC-ID-179

Symposia were held at The Johns Hopkins University in both 1951 and 1952 on the role of phosphorus in the metabolism of plants and animals. The proceedings of these symposia were published in two volumes.[67]

The metabolism of at least three other elements has also been covered in monographs. The relationship of animals, plants, and the soil in copper metabolism was discussed at a symposium[68] on copper metabolism held at Johns Hopkins in 1950. Because of the short half-life of Cu^{64} ($t_{1/2}$ = 12.8 hr), isotopes were not used extensively in the studies reported; however, a good paper describing isotopic methods for elements having relatively short half-lives was given. In The Metabolism of Sulfur Compounds,[69] published in 1958, the authors note that of the 800 references cited, over half of them were published after 1950. A book on calcium metabolism[70] was published in 1957. The International Atomic Energy Agency sponsored a conference on medical uses of Ca^{47} in December 1961. Although the title is "medical uses," several good reports on calcium metabolism in humans and on techniques for use of this new isotope were given.[71]

New Work

It was noted earlier that there was a large increase in isotopic work in biology when long-lived isotopes of carbon and hydrogen were discovered. Unfortunately, there is still no good long-lived radioactive isotope of oxygen. As a rule, when an oxygen tracer is needed, the stable isotope O^{18} is used. Workers at Washington University,[72,73] St. Louis, Mo., have recently been producing O^{15} ($t_{1/2}$ = 2-1/4 min.) by the deuteron bombardment of nitrogen in the air by a cyclotron. The O^{15}

thus produced by the $N^{14}(d,n)O^{15}$ reaction is immediately purified and the experimental animal breathes it. With the experimental set-up shown in Figure IV-5, it is possible to use O^{15} to study the physiology of oxygen in the body. Studies with humans have shown that it is also feasible to use this method as a diagnostic technique in clinical medicine.

Recently radioisotopes have been used to study the movement of an element in the different parts of an ecological system. These studies have now become so extensive that the word "radioecology" has been coined to describe them. An example of this type of study is an experiment done at Oak Ridge National Laboratory.[74] Two-millicurie portions of Cs^{134} were put in the trunks of several white oak trees. Figure IV-6 shows where the activity was found 165 days later. Potassium-42 was also injected in some of the trees, and the differences in behavior between Cs^{134} and K^{42} were found.

One of the most exciting new uses of radioisotopes in biology is the experiment -- hopefully named "Gulliver" -- designed to try to show whether or not there is life on Mars.[75,76] This experiment is based on the assumption that if there is life as we know it on Mars, there will certainly be microorganisms present in the soil. Following landing on Mars, the apparatus is designed to shoot two 25-ft strings coated with a sticky silicone grease out into the soil and then pull them back in. A culture medium containing sodium formate-C^{14} and glucose-C^{14} (uniform label) will then be put over the string. If there is life present, the radioactive carbon dioxide which will be given off can be determined by a counter (placed so that the C^{14} in the culture medium does not affect it) and the data telemetered back to earth. The current version of this

Figure IV-5

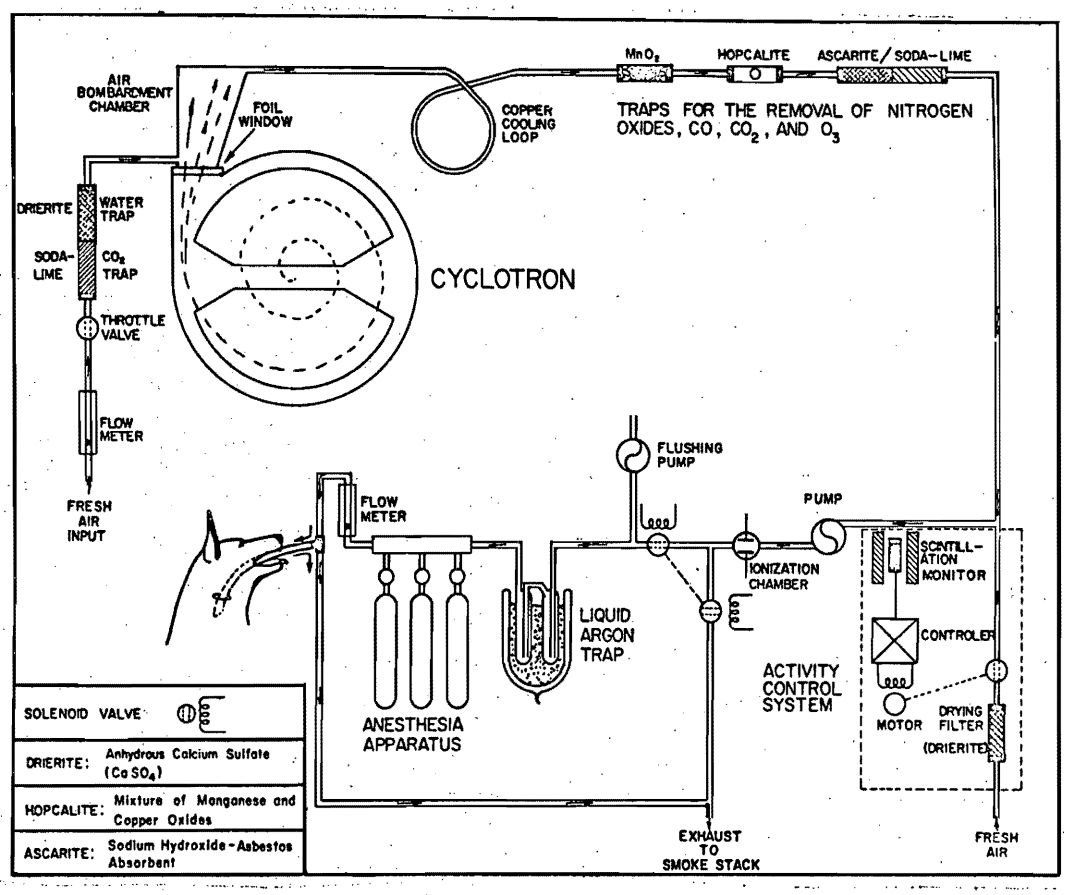
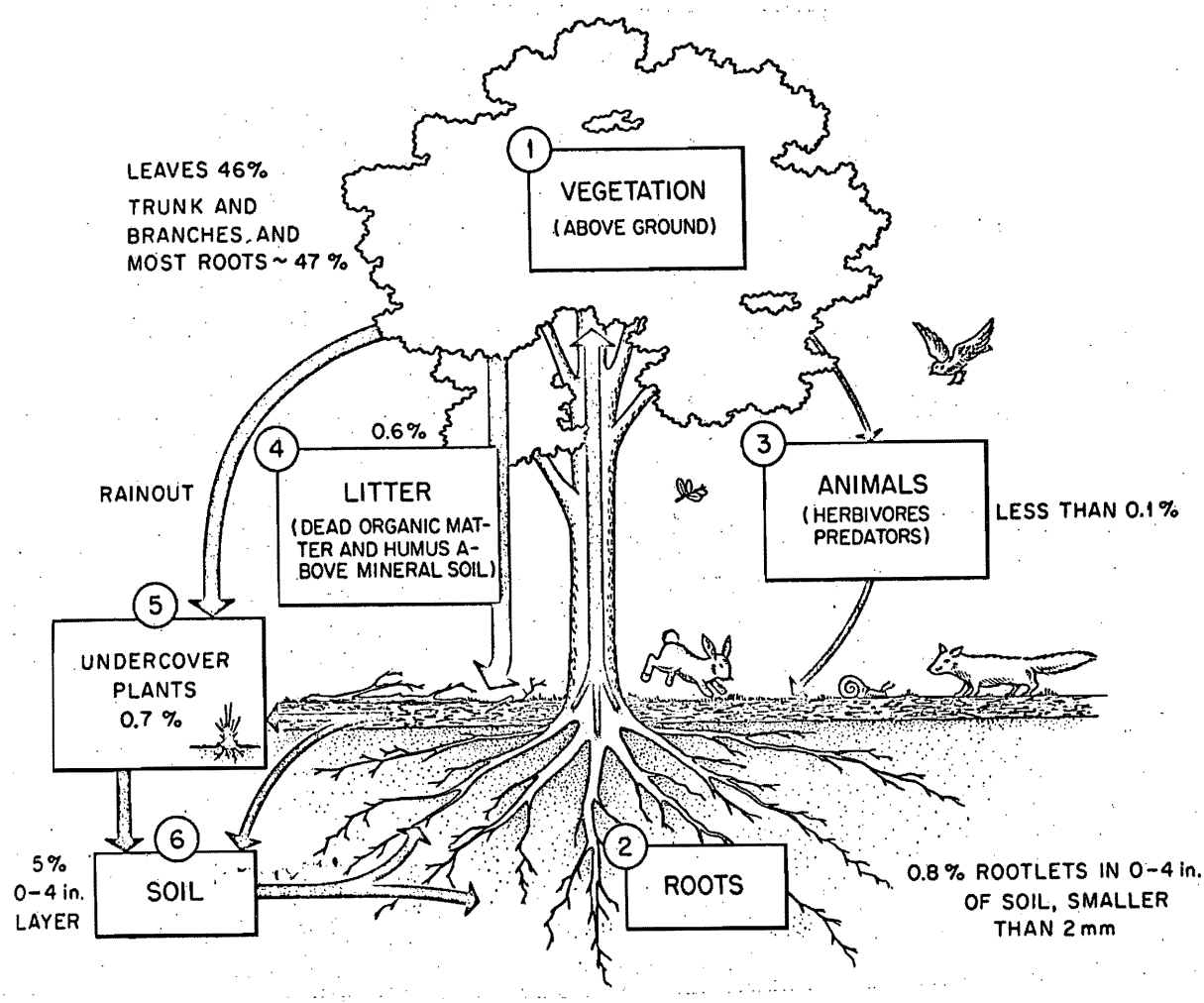


Diagram shows the method[72] devised for the utilization of O^{15} in studies on the physiology of oxygen in the body. The radioisotope has a half-life of only 2-1/4 minutes and must be used immediately after production. The radioactive oxygen is produced by bombarding air with a deuteron beam from the cyclotron, then passed through filters that remove oxygen compounds other than molecular oxygen before being administered to test animals by means of a trachial tube.

From: Atomic Energy Research 1961, A special Report of the United States Atomic Energy Commission, December 1961 (p. 10).

Figure IV-6



The rapid ecological movement of cesium 134 is illustrated in the above drawing of a white oak tree whose trunk was tagged with two millicuries of the radioisotope. Within 165 days, the tracer had become redistributed in different parts of the ecological system and was again entering the tree, this time through the root system.

From: Atomic Energy Research 1961, A Special Report of the United States Atomic Energy Commission, December 1961 (p. 115). This drawing is also available as ORNL-LR-DWG-68158.

1-1/3 lb. instrument has been tested successfully in Rock Creek Park in Washington.

RADIOCHEMISTRY

Radioisotopes have been used for a wide variety of applications in inorganic and analytical chemistry research. Since there is a large area of overlap in these fields, it is difficult to divide the literature into two arbitrary parts. Rather than make this split, we will combine the discussion of literature common to the two fields and then present short sections on each subject separately.

Radiochemistry has been defined by Paneth[77] as "the chemistry of bodies detected through their nuclear radiation," and Broda[78] has divided the field into fundamental and applied radiochemistry. In fundamental radiochemistry, the radioelement is the object of the research; in applied radiochemistry the radioisotope is used as a tool. Inorganic research falls in the category of fundamental radiochemistry because the goal is to learn about the preparation and properties of an isotope; on the other hand, analytical chemistry is applied radiochemistry since the isotope is a tool. The general techniques are the same, but the goals are different.

The literature of radiochemistry was reasonably uncomplicated and easy to follow until shortly after the discovery of artificial radioactivity in 1933 by M. and Mme. Curie-Joliot. For many years, two books by Paneth[77] and Hahn[79] published as the result of their Baker Non-Resident Lectureships at Cornell University were the standard references. In these books, published in 1928 and 1936 respectively, only naturally radioactive isotopes were discussed although Hahn, in a postscript to his introduction, noted that the discovery of artificial radioactivity "will without doubt lead in the future to an extraordinary broadening of the field of Applied Radiochemistry." Hevesy and Paneth in the second edition of <u>A Manual of</u>

Radioactivity[80] covered both artificial and natural isotopes; however, the largest part of this book was devoted to the physics of isotopes and radioactivity rather than to radiochemistry. Broda[78] in Advances in Radiochemistry covered the literature of radiochemistry up to December 1949. This book, written in the early post-war period, was among the first to consider the impact of the nuclear reactor and other wartime developments on radiochemistry.

Bibliography

The Subcommittee on Radiochemistry of the NAS-NRC has published an excellent bibliography, Source Material for Radiochemistry.[81] W. W. Meinke, the subcommittee chairman, describes the reason for this bibliography and the method used to compile it in the foreword:

> "The field of radiochemistry has no journal or other publication outlet to call its own. Some abbreviated material is published in analytical or nuclear journals, but seldom is a detailed radiochemical separation procedure or an elaborate counting technique given full play in the regular literature. Often different groups record their procedures in documents which are circulated within the framework of the Atomic Energy Commission. Most of these documents are also available for sale but many times the "outsider" is not aware of the report or of its availability.
>
> This compilation then is an attempt to list current source material of interest to the radiochemist. It has been compiled from lists submitted by each member of the Subcommittee on Radiochemistry. Emphasis has been placed on documents and reports of a review nature which have proven useful to members of this Subcommittee. No attempt has been made to include standard analytical reference works which, of course, are indispensible in planning new procedures."

Each of the entries in this bibliography is evaluated as to its potential usefulness in radiochemical work. It is divided into the following major sections:

Books on radiochemistry, nuclear chemistry, and nuclear physics

- Textbooks and reference works
- Textbooks and instruction manuals with emphasis on techniques
- General compilations of references pertinent to radiochemistry

Collections of radiochemical procedures

- General
- Monograph series on the radiochemistry of the elements

Ion exchange and solvent extraction

Activation analysis

Nuclear geochemistry, oceanography, geochronology

Counting techniques

Nuclear data compilations

Where and how to obtain radioactive isotopes

Laboratory design and safety

Since in our opinion this a reasonably complete and up-to-date compilation, only a few references from it are used in this review.

Many good references on radiochemistry can also be found in Special Sources of Information on Isotopes,[82] which is prepared and distributed by the Division of Isotopes Development, U. S. Atomic Energy Commission.

Radiochemistry of the Elements

As Meinke noted above, radiochemical separation procedures are widely scattered and sometimes written in abbreviated form. Some AEC

installations have compiled their procedures and issued them in the form of reports.[83]

The best compilation of radiochemical procedures now available is the monograph series on radiochemistry of the elements, being prepared under the sponsorship of the Subcommittee on Radiochemistry of the National Research Council and printed by the U. S. Atomic Energy Commission. Each of these one-volume monographs collects the information needed for radiochemical work with an individual element or a group of closely related elements. All the elements except hydrogen, helium, lithium, and boron will be covered in 60 monographs when this series is complete. There are four volumes still to be issued. Table IV-2 is a cross-referenced table of contents for the series, which may be obtained from the Office of Technical Services for the prices shown in the table.

Radiochemical Techniques

In general, radiochemical separation techniques are those which have been used for analytical separations for many years. It is noteworthy, therefore, that the analytical chemist by using radioisotopes has been able to strengthen and re-establish many old separation techniques. For example, the traditional solvent extraction and ion exchange techniques did not become powerful analytical tools until tracer techniques had been used on the Manhattan Project to develop them for the separation of uranium, plutonium, and fission products.

The close similarity between inorganic radiochemical and analytical chemical techniques was noted by Finston and Miskel in an article in Annual Reviews of Nuclear Science[84] for 1955:

Table IV-2 NATIONAL ACADEMY OF SCIENCE - NUCLEAR SCIENCE SERIES ON RADIOCHEMISTRY OF THE ELEMENTS

Element	Symbol	NAS-NS, No.	Other Elements	Authors	Price,$
Actinium	Ac	3020	RE, Sc, Y	P. C. Stevenson W. E. Nervik	3.00
Aluminum	Al	3032	Ga	J. E. Lewis	0.50
Americium	Am	3006	Cm	R. A. Penneman T. K. Keenan	0.75
Antimony	Sb	3033	--	W. Maeck	0.50
Argon	Ar	3025	Ne, Kr, Xe, Rn	F. F. Momyer	0.75
Arsenic	As	3002	--	H. C. Beard	0.50
Astatine	At	3012	--	E. H. Appleman	0.50
Barium	Ba	3010	Ca, Sr	D. N. Sunderman C. W. Townley	1.25
Berkelium	Bk	3031	Trans-curium	G. H. Higgins	0.50
Beryllium	Be	3013	--	A. W. Fairhall	0.75
Bismuth	Bi			W. Gibson	
Boron	No radiochemistry				
Bromine	Br	3005	F, Cl, I	J. Kleinberg G. A. Cowan	0.50
Cadmium	Cd	3001	--	J. R. DeVoe	0.75
Calcium	Ca	3010	Ba, Sr	D. N. Sunderman C. W. Townley	1.25
Californium	Cf	3031	Trans-curium	G. H. Higgins	0.50
Carbon	C	3019	N, O	J. Hudis	0.50
Cerium	Ce	3020	RE, Sc, Y, Ac	P. C. Stevenson W. E. Nervik	3.00
Cesium	Cs	3035	--	H. L. Finston M. T. Kinsley	0.75
Chlorine	Cl	3005	F, Br, I	J. Kleinberg G. A. Cowan	0.50
Chromium	Cr	3007	--	J. Pijck	0.50

Table IV-2 Continued

Element	Symbol	NAS-NS, No.	Other Elements	Authors	Price,$
Cobalt	Co	3041	--	L. C. Bate G. W. Leddicotte	1.00
Copper	Cu	3027	--	F. F. Dyer G. W. Leddicotte	0.75
Curium	Cm	3006	Am	R. A. Penneman T. K. Keenan	0.75
Dysprosium	Dy	3020	RE, Sc, Y, Ac	P. C. Stevenson W. E. Nervik	3.00
Einsteinium	Es	3031	Trans-curium	G. H. Higgins	0.50
Erbium	Er	3020	RE, Sc, Y, Ac	P. C. Stevenson W. E. Nervik	3.00
Fermium	Fm	3031	Trans-curium	G. H. Higgins	0.50
Fluorine	F	3005	Cl, Br, I	J. Kleinberg G. A. Cowan	0.50
Francium	Fr	3003	--	E. K. Hyde	0.50
Gadolinium	Gd	3020	RE, Sc, Y, Ac	P. C. Stevenson W. E. Nervik	3.00
Gallium	Ga	3032	Al	J. E. Lewis	0.50
Germanium	Ge	3043	--	J. A. Marinsky	0.50
Gold	Au	3036	--	J. F. Emery G. W. Leddicotte	0.50
Hafnium	Hf	3011	Zr	E. P. Steinberg	0.50
Helium	No radiochemistry				
Holmium	Ho	3020	RE, Sc, Y, Ac	P. C. Stevenson W. E. Nervik	3.00
Hydrogen	No radiochemistry				
Indium	In	3014	--	D. N. Sunderman C. W. Townley	0.50
Iodine	I	3005	F, Cl, Br	J. Kleinberg G. A. Cowan	0.50
Iridium	Ir	3045	--	G. W. Leddicotte	0.50
Iron	Fe	3017	--	J. M. Nielsen	0.50
Krypton	Kr	3025	Ne, Ar, Xe, Rn	F. F. Momyer	0.75

Table IV-2 Continued

Element	Symbol	NAS-NS, No.	Other Elements	Authors	Price,$
Lanthanum	La	3020	RE, Sc, Y, Ac	P. C. Stevenson W. E. Nervik	3.00
Lead	Pb	3040	--	W. M. Gibson	1.75
Lithium	No radiochemistry				
Lutetium	Lu	3020	RE, Sc, Y, Ac	P. C. Stevenson W. E. Nervik	3.00
Magnesium	Mg	3024	--	A. W. Fairhall	0.50
Manganese	Mn	3018	--	G. W. Leddicotte	0.50
Mendelevium	Md	3031	Trans-curium	G. H. Higgins	0.50
Mercury	Hg	3026	--	J. Roesmer P. Kruger	0.50
Molybdenum	Mo	3009	--	E. M. Scadden N. E. Ballou	0.50
Neodymium	Nd	3020	RE, Sc, Y, Ac	P. C. Stevenson W. E. Nervik	3.00
Neon	Ne	3025	Kr, Ar, Xe, Rn	F. F. Momyer	0.75
Neptunium	Np		Pu	N. Jackson	
Nickel	Ni	3051	--	L. J. Kirby	0.50
Niobium	Nb	3039	Ta	E. P. Steinberg	0.75
Nitrogen	N	3019	C, O	J. Hudis	0.50
Nobelium	No	3031	Trans-curium	G. H. Higgins	0.50
Osmium	Os	3046	--	G. W. Leddicotte	0.50
Oxygen	O	3019	C, N	J. Hudis	0.50
Palladium	Pd	3052	--	O. T. Høgdahl	0.75
Phosphorus	P	3056	--	W. T. Mullins G. W. Leddicotte	0.50
Platinum	Pt	3044	--	G. W. Leddicotte	0.50
Plutonium	Pu		Np	N. Jackson	
Polonium	Po	3037	--	P. E. Figgins	0.75
Potassium	K	3048	--	W. T. Mullins G. W. Leddicotte	0.50

Table IV-2 Continued

Element	Symbol	NAS-NS, No.	Other Element	Authors	Price,$
Praseodymium	Pr	3020	RE, Sc, Y, Ac	P. C. Stevenson W. E. Nervik	3.00
Promethium	Pm	3020	RE, Sc, Y, Ac	P. C. Stevenson W. E. Nervik	3.00
Protactinium	Pa	3016	--	H. W. Kirby	1.00
Radium	Ra		--	M. L. Salutsky	
Radon	Rn	3025	Kr, Ar, Xe, Ne	F. F. Momyer	0.75
Rhenium	Re	3028	--	G. W. Leddicotte	0.50
Rhodium	Rh	3008	--	G. R. Choppin	0.50
Rubidium	Rb	3053	--	G. W. Leddicotte	0.50
Ruthenium	Ru	3029	--	E. I. Wyatt R. R. Rickard	1.00
Samarium	Sm	3020	RE, Sc, Y, Ac	P. C. Stevenson W. E. Nervik	3.00
Scandium	Sc	3020	RE, Y, Ac	P. C. Stevenson W. E. Nervik	3.00
Selenium	Se	3030	--	G. W. Leddicotte	0.50
Silicon	Si	3049	--	W. T. Mullins G. W. Leddicotte	0.50
Silver	Ag	3047	--	D. N. Sunderman C. W. Townley	0.75
Sodium	Na	3055	--	W. T. Mullins G. W. Leddicotte	0.50
Strontium	Sr	3010	Ba, Ca	D. N. Sunderman C. W. Townley	1.25
Sulfur	S	3054	--	G. W. Leddicotte	0.50
Tantalum	Ta	3039	Nb	E. P. Steinberg	0.75
Technetium	Tc	3021	--	E. Anders	0.50
Tellurium	Te	3038	--	G. W. Leddicotte	0.50
Terbium	Tb	3020	RE, Sc, Y, Ac	P. C. Stevenson W. E. Nervik	3.00
Thallium	Tl			E. L. Brownlee	
Thorium	Th	3004	--	E. K. Hyde	0.75

Table IV-2 Continued

Element	Symbol	NAS-NS, No.	Other Elements	Authors	Price,$
Thulium	Tm	3020	RE, Sc, Y, Ac	P. C. Stevenson W. E. Nervik	3.00
Tin	Sn	3023	--	W. E. Nervik	0.75
Titanium	Ti	3034	--	C. K. Kim	0.50
Tungsten	W	3042	--	W. T. Mullins G. W. Leddicotte	0.50
Uranium	U	3050	--	J. Grindler	3.50
Vanadium	V	3022	--	J. L. Brownlee	0.75
Xenon	Xe	3025	Ne, Ar Kr, Rn	F. F. Momyer	0.75
Ytterbium	Yb	3020	RE, Sc, Y, Ac	P. C. Stevenson W. E. Nervik	3.00
Yttrium	Y	3020	RE, Sc, Ac	P. C. Stevenson W. E. Nervik	3.00
Zinc	Zn	3015	--	H. G. Hicks	0.75
Zirconium	Zr	3011	Hf	E. P. Steinberg	0.50

"The problem of separation of radionuclides, except for the special case of the Szilard-Chalmers and recoil separations, are essentially the problems of analytical chemistry. The analytical chemistry literature has been carefully scrutinized, and suggestions are made as to the application of procedures which may not have been used for radiochemical separations."

The Subcommittee on Radiochemistry has now started a series of monographs on radiochemical techniques,[85] which is a companion series to the Radiochemistry of the Elements monographs. These are intended to supplement rather than replace or duplicate existing reviews or monographs. For instance, Morrison and Freiser[86] have written a general textbook on solvent extraction and also a review specifically on solvent extractions in radiochemical separations;[87] but the NAS monograph series covers in detail two very specific and useful types of solvents widely used in radiochemistry -- high-molecular-weight-amines[85a] and tri-n-octylphosphine oxide.[85b] Other monographs in this series cover low-level radiochemical separations, rapid radiochemical separations, detection and measurement of nuclear radiation, and paper chromatography and electromigration techniques.

Because of the importance of solvent extraction and ion exchange techniques to radiochemistry, Source Material for Radiochemistry,[81] has a special section devoted to them. To give detailed reviews of these or other separation methods is beyond our scope; however, the April issue of Analytical Chemistry in even-numbered years gives excellent and complete reviews of these and many other separation methods.[88]

One specialized radiochemical technique is activation analysis, in which a sample is irradiated with neutrons and the resulting radioactive

elements are identified and the amount present determined (Fig. IV-7). This technique was originated in 1936 by Hevesy and Levi,[89] who exposed samples of rare earths to a radium-beryllium neutron source to detect and determine impurities. However, it did not gain wide acceptance until nuclear reactors became available to provide a high-flux source of neutrons. The literature in this field up to 1960 is well covered in Source Material in Radiochemistry.[81] Several new and interesting uses of this technique that have recently been developed are discussed later in this section.

New Developments in Inorganic Chemistry

In general, new developments in inorganic radiochemistry are covered in the Annual Reviews of Nuclear Science. The 1962 volume[90] contains the following articles that may interest readers of this review:

R. L. Mössbauer, Recoilless Nuclear Resonance Absorption, p. 123-52

L. Yaffe, Preparation of Thin Films, Sources, and Targets, p. 153-88

R. H. Herber, Isotopic Exchange Reactions in Nonaqueous Systems, p. 329-58

G. Burbidge, Nuclear Astrophysics, p. 507-76

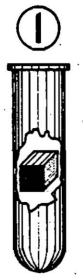
Since 1959, there has been a review series, Advances in Inorganic Chemistry and Radiochemistry. The three volumes that have appeared to date have had more inorganic than radiochemistry, but the 1961 volume[91] had an article by A. G. Maddock and E. H. Willis on "Atmospheric Activities and Dating Procedures."

The first book of Inorganic Isotope Syntheses[92] was published in 1962 and contains procedures for the synthesis of a wide variety of inorganic compounds containing either radioactive or selected separated

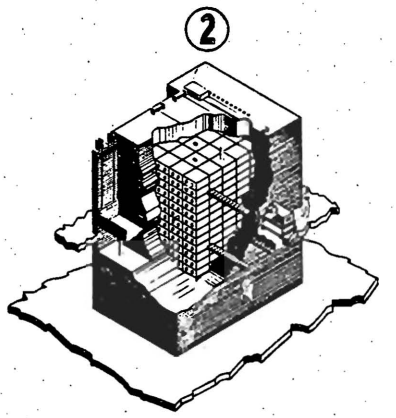
Figure IV-7

INDUCTION OF RADIOACTIVITY

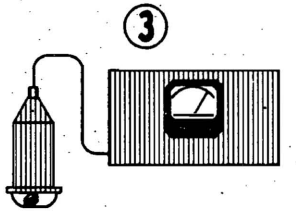
FOR ACTIVATION ANALYSIS



SAMPLE CONTAINING UNKNOWN ELEMENTS



SAMPLE IRRADIATED WITH NEUTRONS IN NUCLEAR REACTOR



INDUCED RADIOISOTOPES IDENTIFIED BY RADIATION CHARACTERISTICS

ADVANTAGES:

1 - ELIMINATES TEDIOUS CHEMICAL PROCEDURES

2 - MORE SENSITIVE THAN CHEMICAL AND SPECTROSCOPIC METHODS

3 - METHOD IS BOTH QUALITATIVE AND QUANTITATIVE

DISADVANTAGES:

1 - LARGE SOURCE OF NEUTRONS SUCH AS A REACTOR IS NEEDED

2 - SIMILAR RADIATION PROPERTIES MAY PREVENT DIFFERENTIATION OF INDUCED RADIOISOTOPES

USAEC-ID-229

stable isotopes. A chapter on general procedures is followed by chapters on methods for synthesis of compounds containing deuterium, tritium, N^{15}, O^{18}, P^{32}, S^{35}, Cl^{36}, and I^{131}.

New Developments in Analytical Chemistry

New developments in the applications of radiochemistry to analysis are reviewed every other year in Analytical Chemistry. The most recent review, by Leddicotte, was in 1962.[93] The three earlier reviews were by Meinke.[94] These four review articles contain over 4000 references; but in the last two Meinke and Leddicotte stated that they were limiting their coverage to selected articles rather than trying to cover all the papers in the field.

An interesting new analytical technique for determination of dissolved oxygen in water was recently described by H. G. Richter and A. S. Gillespie, Jr., of the Research Triangle Institute. The fact that, even in extremely low concentrations, oxygen dissolved in water rapidly and quantitatively oxidizes thallium metal is the basis for this method. This fact, discovered by Wright and Lindsay,[95] had been used in an industrial instrument which measured the electrical conductivity of the effluent stream. By substituting Tl^{204} and "counting" the effluent stream, Richter and Gillespie could determine 0.2 ppm of oxygen in pure water and were able to use the method in water containing many more impurities than in the original method. The authors believe that this method has a potential sensivity greater than any other known method for this determination.

New Developments in Activation Analysis

Although activation analysis has been known for a relatively long time, very few analyses are run routinely by this technique. Meinke and

Shideler[97] have suggested that activation analysis will come into general use in the laboratory only when a neutron source suitable for use in an individual laboratory is 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 developments in this field are directed toward the solution of both of these problems. Several laboratories are working on small neutron sources that are relatively portable and not too expensive, whereas others are evaluating ways to speed up analyses and put them on a routine basis.

Neutron Sources. Olive, Cameron, and Clayton of the UKAEA Isotope Research Group have recently reviewed high-intensity neutron sources and their applications in industry.[98] Nuclear reactors are not considered, but radioisotope sources and accelerating machines are discussed in some detail. These authors conclude that radioisotope sources such as those shown in Table IV-3 will usually be limited to a neutron flux of about 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 will probably be more economical.

Hennelly[99] has 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 easily regenerated in a reactor. He estimates that it could be operated for $5000 per year. The British review[98] 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.

Table IV-3 RADIOISOTOPE NEUTRON SOURCES

Source	Half-Life	Yield $n\ sec^{-1}\ curie^{-1}$
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}

Strain and Leddicotte[100] have recently published work on 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. This resulted in a flux of 2.36×10^4 n cm^{-2} sec^{-1}. Although this flux is low, they believe it would be suitable for macro concentrations of a large number of materials. As Am^{241} becomes available, this type of source should be more important.

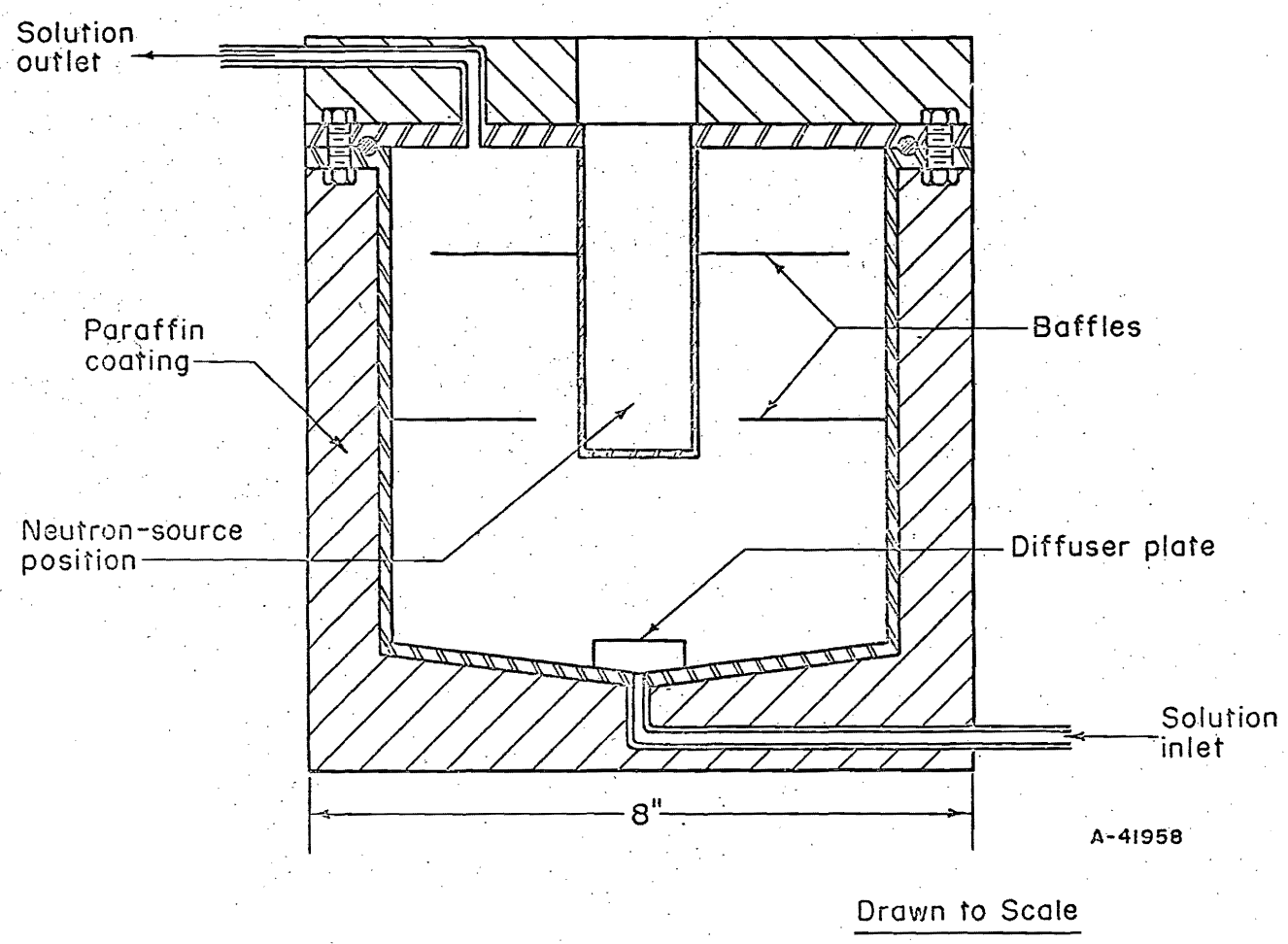
Nucleonics[101] recently reviewed the use of accelerators for the production of neutrons. Currently the major interest in neutron sources for activation analysis seems to be in the small particle accelerators which generate neutrons by the $H^3(d,n)He^4$ reaction. In the British review mentioned above,[98] data from the Nucleonics article on such machines, which cost about $20,000 (Table IV-4), are summarized.

Two recent reports[97,101] describe the installation and use of Texas Nuclear Corporation Cockcroft-Walton neutron generators. The installation at the University of Michigan[97] is routinely producing a thermal neutron flux of 2×10^8 n cm^{-2} sec^{-1}, which the authors believe is about the limit with present target designs. The Oak Ridge National Laboratory[101] generator flux is about 3.5×10^8 n cm^{-2} sec^{-1}.

An interesting type of neutron source has recently been described by Gluck, McFarling, Kircher, Townley, and Sunderman at Battelle Memorial Institute in a study done for the USAEC Division of Isotopes Development.[102] Designed for activation analysis on a flowing stream, the irradiator uses a Po-Be source that can be lowered into the activation cell shown in Fig. IV-8. The activation achieved with this cell in a flowing stream was tested with several concentrations of manganese, indium, vanadium, and silver solutions.

Table IV-4 LOW-COST ACCELERATOR NEUTRON SOURCES

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



DETAIL OF ACTIVATION CELL

Fig. IV-8

Rapid Methods of Analysis. In addition to small portable neutron generators, Meinke also stated that analyses should be done rapidly. Three different approaches are being investigated: (1) Meinke has been working on quick radiochemical separation methods[85d] following activation so that the isotopes with short half-lives can be determined;[103,104] (2) Guinn and co-workers[105,106] are working on instrumental methods in which neutron activation is followed by gamma-ray spectrometry with no chemistry; frequently the gamma-ray spectrum is determined several times to determine half-lives of various activities; (3) Wainerdi and co-workers at Texas A & M,[107,108] are studying very elaborate techniques in which the activation and counting sequences are controlled by a computer. The samples are introduced into the system in batches of 100, activated in the reactor, counted in a gamma-ray spectrometer, and the results are calculated and printed out. Since the entire sequence is controlled by the computer, this method should be ideal for routine analyses of large numbers of samples.

Uses of Activation Analysis. Several laboratories have recently looked at the possibilities of using activation analysis to aid in the examination of evidence for law enforcement purposes.

V. P. Guinn[109] 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} and Ba^{139}, and determining these two activities by gamma-ray spectrometry. Guinn is now working with the Los Angeles Police Department on the application of this method.

Bate, Pro, and Leddicotte of Oak Ridge National Laboratory and the Alcohol and Tobacco Tax Laboratory, Internal Revenue Service, have examined a number of samples of black automobile paint and some soil and soot samples,[110] and they have concluded that activation analysis would be a useful technique for the forensic scientist to use in identification of these materials.

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 suspected arsenic poisoning.[111] 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. They report a case in which activation analysis of a hair was introduced in court.[112] In this case 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 and the results shown in Table IV-5 were obtained. The difference between the P^{32}/Sb^{35} ratio in the two hairs was presented in the Canadian courts as evidence of identity.

Table IV-5 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

ORGANIC CHEMISTRY

For years the organic chemist had needed a method to distinguish between carbon atoms in various parts of an organic molecule. With the availability of radioisotopes, his need was answered; and now, for the first time, he was able to say with assurance how the various parts of a molecule behaved in a reaction. Obviously, the most important isotope in this work is C^{14}, but the stable isotope C^{13} is also used when two carbon tracers are needed. Tritium serves extensively for tracing hydrogen, and if a second tracer is needed, deuterium is available. Sulfur-35, P^{32}, and I^{131} are also used to a limited extent in organic research. Several good books that summarize the information in this field are available. These books, described briefly in the following paragraphs, provide not only a good background in this subject but also a history of its development over the past 15 years.

The first book devoted entirely to the use of carbon isotopes in organic and biochemistry, Isotopic Carbon,[113] was published in 1949, and describes techniques using C^{11}, C^{13}, and C^{14}. In the introduction, Calvin indicates the tremendous interest which had started to develop in this subject:

> "It is our conviction that isotopic carbon as a tracer will very soon find its place as a routine tool in many kinds of scientific laboratories. Its use on a routine basis for the investigation of physical, chemical, and biochemical transformations is comparable with that of the microscope, spectroscope, or x-ray analyses in their respective spheres. Because of their great diversity, the applications of this tool are beyond the scope of this book, and they have been included only as a bibliography, Appendix X, which contains all published tracer applications of isotopic carbon up to July, 1948." [Appendix X contains about 195 references.]

After 14 years, Calvin's book is still a valuable reference. He describes most of the basic techniques for working with both C^{14} and C^{13}. In one chapter of almost 100 pages, he summarizes all of the syntheses of organic compounds with isotopes published up to April 1948. Since most of the compounds described were the fundamental ones needed in many organic syntheses, this chapter is still useful if one wants to synthesize, rather than buy, a specific compound.

Nevenzel and co-workers at the Medical School of the University of California at Los Angeles have prepared two bibliographies of the literature on synthesis of organic compounds containing isotopes. Their original bibliography[114] covered compounds published through 1952. This report contained 748 references in 175 pages. The organic compounds are listed by name in an empirical formula index, and references are given to a list of original literature citations. There is also an author index. In 1957, the same workers published a supplement to the original bibliography[115] which contained corrections to the original work and covered literature for the years 1953 and 1954. This bibliography was indexed in the same way as the original and contained about 550 additional references. At this point, the authors gave up their original plan to follow all literature on the synthesis of organic compounds containing tracers:

> "The originally planned yearly supplement to this bibliography has proved impractical with the limited manpower presently available. Future supplements will be compiled on a reduced scale with emphasis being placed on abstraction of more obviously preparative papers."

Murray and Williams in 1958 published a two-volume work, Organic Syntheses with Isotopes.[116] The first volume describes syntheses with C^{14} and the second discusses compounds labeled with isotopes of the halogens, hydrogen, nitrogen, oxygen, phosphorus, and sulfur. The authors cover the literature through 1955. They include chemical but not biological syntheses. Catch,[117] in discussing Murray and Williams' book says:

> "This last book, after a brief and useful introduction -- too brief unfortunately to cover the field adequately -- embodies about 800 practical methods for isotopic syntheses, of which over 400 in volume I deal with carbon-14. References and brief information on many more preparations are also given. The methods are presented in the style of Organic Syntheses, and this may lead the incautious reader to assume that (as in that unrivalled collection) all the methods have been independently checked; but this in general appears not to be so, and a corresponding degree of dependability is not to be assumed. With this reservation (which is not a criticism) the book will be found well worth its rather high price."

Three other books on the subject of isotopes in organic chemistry should also be mentioned. The first, by Burr, Tracer Applications for the Study of Organic Reactions,[118] was published in 1957. The second, by Catch, Carbon-14 Compounds,[117] was published in 1961. The third and most recently published book in the field is Isotopes in Organic Chemistry,[119] by Miklukhin which was published in Russia in 1961.

The book by Burr is written primarily for the organic chemist, and the isotopic applications are arranged according to the reaction classifications most familiar to the organic chemist. This arrangement results in the subordination of techniques used in tracer work and emphasis on the results. Burr's text covers the literature through 1952; however, he has

added an appendix in which literature citations, arranged by chapters, are given for material that appeared in Chemical Abstracts up to early 1955.

Catch's book is arranged with emphasis on techniques for working with C^{14}, and is thus a valuable complement to Burr's. He states in his introduction that he has not tried to cover the literature comprehensively; therefore, his bibliographies are selective and should be more valuable than if they had been exhaustive. He covers the literature through 1958 with a few references to 1959 and 1960. An appendix provides some coverage through 1960.

No English translation of Miklukhin's book is available at this time. Most of the material is in tables and references, and both the formulas of the compounds and the references to all but the Russian work can be followed with only a very slight knowledge of Russian. This book is divided into three parts: (1) "Methods for Obtaining and Physical Properties of Labeled Organic Compounds" with a bibliography of about 1300 references; (2) "Isotope Exchange Reactions" with 1400 references; and (3) "Research on Mechanism of Chemical Reactions" with about 1300 references. The references cover the literature to the beginning of 1957.

Today the organic chemist who wishes to work with tracers frequently will not have to synthesize a compound before he starts work. The 1962 edition of The Isotope Index[120] lists about 1275 compounds with carbon-14 tracers that are commercially available. This number counts separately each organic compound having a label in a different location, but does not count compounds having the same label but different specific

activities. In addition, about 225 compounds are reported containing tritium labels, 95 labeled with S^{35}, 50 with P^{32}, and 40 with I^{131}. The International Directory of Radioisotopes,[121] which was published in 1962 and covered the entire world, listed the following numbers of available tracer organic compounds: C^{14} about 1280; tritium about 250; I^{131} about 55; P^{32} about 50; S^{35} about 120.

GEOPHYSICS

Isotopes have assumed an important place in many fields of geophysics research in recent years. The International Atomic Energy Agency held a symposium on one of these fields, radioactive dating, in Athens, November 19-23, 1962. The January 1963 International Atomic Energy Agency Bulletin had a good report of this meeting, "Dating the Past with Radioactivity." We reprint this article here with a few editorial changes and the addition of some references which may add to its value.

"Very soon after the discovery of radioactivity, measurements of the heavy radioactive elements existing in nature were undertaken to determine the age of rocks by Rutherford in 1904,[122] but only when Professor Willard Libby in an evening lecture during the first United Nations Conference on the Peaceful Uses of Atomic Energy in Geneva in 1955 described how he and his research team had evolved the C^{14} dating technique did research on radioactive dating receive a real impetus.[123] Since that time laboratories in many countries have made rapid progress and geologists, geophysicists, geochemists, and archaeologists have joined forces with nuclear scientists to perfect the techniques and widen the range of its applications."

"Professor Libby had first worked out the radiocarbon dating technique in theory; his team had found and extracted radiocarbon from sewage slushes in Baltimore, conducted a worldwide assay of woods and developed the necessary counting procedures and equipment. They then conducted actual datings, both of samples of known age, which proved that the method "worked", and of objects of hitherto unknown age. He first published his findings in 1952.[124] Interest in the 1950's centred mainly round the Libby method using C^{14} as the "chronometer" and the dating of archaeological

and other relatively recent objects.[125] Lately the interest and techniques have again shifted to the age determination of the very distant past down to the very origin of our solar system and to the application of the radioactive dating method to problems in the earth sciences, such as the movements of air and water masses, the origin and nature of rock formations, and the history of meteorites.

"This shift of interest was clearly discernible at a five-day international symposium organized by IAEA in Athens, 19-23 November 1962, which, although also dealing with C^{14} and archaeology, mainly discussed geological and meteoritic problems. The symposium was co-sponsored by the Joint Commission on Applied Radioactivity of the International Council of Scientific Unions (ICSU); the Greek Atomic Energy Commission acted as host.

Methods and Possibilities

"The general principle of radioactive dating is based on measuring the decay of certain given radioisotopes and/or the build-up rate of other radioisotopes in the decay chain. To obtain results with only small margins of error it is necessary to know as exactly as possible the half-life of the radioisotope concerned and to be sure that in the sample to be measured the activity of the radioisotopes has not escaped by other means than the natural decay. One must also be able to measure small amounts of radiation, which is difficult because of the background radiation that is always present from other sources. In radioactive dating one uses both natural radioisotopes with very long half-lives which have survived since the formation of the earth and the solar system (nucleosynthesis) and -- more recently -- radioactive isotopes

formed by the cosmic ray bombardment both directly on the surface of the earth and in the atmosphere and beyond.

"The half-life of C^{14}, formed in the atmosphere by the neutrons from cosmic rays being absorbed by nitrogen and transformed into this radioactive isotope of carbon, had been assumed in the earlier days of radiodating to be about 5600 years.[124] In a paper by two Swedish research workers from Uppsala University (Olsson and Karlén)[126] it was pointed out that recent investigations at Uppsala, the US National Bureau of Standards[127] and Aldermaston (UK)[128] seemed to agree that the half-life value more correctly should be set at 5700 years.[129] The determination of the half-life of C^{14} is particularly important as this radioisotope is the main tool for measurements up to 50,000 years back and consequently the most used method in archaeology. For dating samples of much greater age, radioisotopes occurring in nature are used. A paper by a German and a Swiss scientist (Herr and Hirt) reported the use of Re^{187} with a half-life of 48 million years to measure the age of iron meteorites. The results corresponded very well with the general assumption, based on the results from other radioactive dating methods, that meteorites and the crust of the earth itself were formed approximately at the same time, some 4500 million years ago. Before the use of the radioactive dating method there were no means of determining the age of the formation of the earth with any degree of reliability and even this method has only recently become reasonably precise.

"Some of the elements in the uranium decay chain emit alpha particles which make up nuclei of helium. The helium can be measured by mass spectrometry and a report on this method was presented in a paper by two

scientists from the University of Arizona (Damon and Green). One main difficulty in this method is to determine the loss of helium in the sample through reasons other than the natural decay, as for instance through "escape" caused by the high emission velocity of the alpha particles, radiation damage, and heat diffusion.

"The thermoluminescence method was discussed in a couple of papers. This is based on measuring the glow of a sample when exposed to heat. Certain elements undergo electronic rearrangement when exposed to radiation, with the electrons accumulating in "traps". This rearrangement is stable at the normal temperature of the environment of the element, but when subjected to heat the electrons recover their pre-radiation position and in this process they emit light which can be measured. The intensity of the light is in direct proportion to the original exposure to radiation.

"The first commonly used method of dating uranium-bearing rocks was to measure the build-up of Pb^{206}, which is a stable isotope at the end of the uranium disintegration chain, and to compare this with the amount of U^{238} contained in the sample. The same principle can be used in comparing the amount of Pb^{206} with other preceding radionuclides in the chain. Experiments reported by Yugoslav scientists (Gojkovic, Deleon, and Cervenjak) showed that by determining the Pb^{206}/Pb^{210} ratio results were obtained which corresponded well with results from other methods.

"One of the main tasks in present-day radioactive dating is to develop counting techniques and equipment which can be used to measure the very low levels of activity usually involved. H. Oeschger, of the Physical Institute of the Berne University, surveyed present low-level radiation detectors (gas and scintillation counters) and described

methods for the suppression of the background radiation. He was of the opinion that although currently used techniques were adequate for samples not older than 50,000 years, improvements in techniques and devices -- which seemed quite feasible -- would open up new promising possibilities. In that case, one might, for instance, use C^{14} for dating up to 60,000 to 70,000 years; one might not have to collect as large samples as at present; and the study of oceanic circulation processes and of radioactivity in terrestrial materials would be eased. It was also most probable, he said, that with higher sensitivity in counting techniques unknown radioisotopes with very long half-lives would be detected.

"By using a sensitive mass spectrometer one can obtain reliable results with the K-Ar dating method even when the samples have low concentration or are of relatively short age. This was reported by two German scientists (Gentner and Lippolt) who had experimented with fossil bones from the Black Forest and volcanic minerals from the Eifel Mountain.

Geology and Applications in Meteorology and Hydrology

"It became clear in the course of the Athens symposium that in the last few years the radioactive dating method has not only sharpened its sensitivity, but also increased its time dimensions in both directions. The progress in methods to determine the ratio between U^{234} and U^{238}, for instance, has made it possible to make more precise age determinations of samples aged 100,000 to more than one million years (Thurber). At the other end of the scale, it was shown that the use of Pb^{210}, formed in the atmosphere through the decay of the short-lived gas Rn^{222}, is useful for dating geochemical processes which have taken place within the last

hundred years or so. This was shown in a paper from California University (Goldberg), which reported on studies of Greenland glaciers and the Colorado and Sacramento rivers. These studies also indicated that the composition of river waters is determined not only by the geology of the drainage basin, but also by inorganic or chemical reactions occurring in the waters.

"The use of C^{14} and other radionuclides formed by cosmic radiation in finding out about the large-scale movements of air in the atmosphere were discussed. In an Indian paper (Lal) it was stated that it was now possible to determine the rates of accumulation of extra-terrestrial dust on the earth in the last few million years by measuring the Al^{26} and Mn^{53} concentrations in these sediments. At present it is estimated that one thousand tons of such dust reaches the earth every day.

"According to the same paper, radioactivity formed on the surface of the earth by cosmic radiation can also be used for erosion studies, although normal erosion rates are so high that surfaces exposed at present would have been tens of meters underground some hundred thousand years ago. This method is therefore suitable only for measuring relatively recent erosion rates. Studies of cosmic ray-produced radioisotopes have also been found useful in the study of oceanic circulation processes and the chronology of oceanic sediments.

"It was, for instance, reported (Bien, Rakestraw, and Suess) that the estimated south-north speed of the water in the Pacific Ocean amounts to 0.04 dm/second; i.e. it takes approximately 400 years for the water at 40° south latitude to reach the 40° north latitude.

K. O. Münnich from Heidelberg University discussed the measuring of artificially produced C^{14} in the high atmosphere; he was of the view that the old "box-model" to explain the slow intermixing of air masses both north/south and between altitudes should be replaced by a "continuous model" based on the diffusion theory.

"The conventional method of determining rock ages has been to measure separated radioisotopes from a sample. This method has the defect that some decay products from the isotope to be measured have been lost during the cooling time of the rock. Strontium-87, a daughter isotope of Rb, seems, however, to remain in the rock; a method of measuring a whole-rock sample, rather than the separated isotopes, for its concentration of Rb in relation to Sr therefore seems promising. A paper from the Massachusetts Institute of Technology (Hurley, Fairbairn, Faure and Pinson) reported on the development of this method. Its success depends partly on determining the initial Sr^{87}/Sr^{86} ratio in the rock at the time of its formation. The authors reported that for most rocks this value is close to 0.708 and this value can be used without having to make individual measurements for each sample.

"Results of age determination of uranium minerals in granite rock formations in Northern Bavaria showed the the uranium had been formed much later than the general deposits of the area, probably by uranium dissolved in acid water flowing through alkaline limestone and thus falling out as uranium salt (Lenz and Wendt).

"The K-Ar dating method has been used to determine the age of tektites (Zähringer), which are found in three main areas: North America, Czechoslovakia, and Australia. The australites seem to be very young,

about 600,000 years old, the Czech tektites about 15 million years, and those from North America more than 30 million years. Tektites are spheric, dark-greenish, glassy objects, commonly assumed to be of cosmic origin. Professor Vinogradov (USSR), however, pointed out in the discussion that available physical and chemical data, particularly the high potassium and uranium content similar to that in acid earth-formed rocks, would lead to the conclusion that tektites were of terrestrial genesis.

"The discrepancies found in using the radioisotopic method for geological dating depend very largely on temperature changes and base exchange by ground water, stated J. L. Kulp of Columbia University. He reported on recent experimental work on the base exchange properties of biotite, where age determination results had been particularly discrepant. His conclusions were that thermal effects rarely caused discrepancies of more than 10 per cent, whereas ground water exchanges would explain the differences that have resulted from using the Rb-Sr or K-Ar methods. These discrepancies have their value, however, in providing valuable information on a region's geochemical history.

"A. P. Vinogradov, of the Biogeochemical Institute of the Soviet Academy of Sciences, reported on C^{14} dating of peat deposits along the lower Indighirka river in East Siberia and on the dating of mammoths (12,000 years) and the change from fresh to brackish water in the Black Sea (8,000 years).

Meteorites

"The determination of the age of the formation of meteorites, the time of their fall to earth, and the production of radioactivity in man-made satellites was the subject of a number of papers. Special attention

was paid to determination of the intensity of cosmic rays in time and space and the formation of penetrating particles in space.

" A United States paper (Fisher) stated that data obtained from using lead as a dating tool showed that stone meteorites had an age of 4500 million years, that is, the same as the crust of the earth. The evidence in respect of iron meteorites, using the K-Ar ratio method, sets their age at up to almost twice as much. There are no other data that support the commonly held assumption that iron and stone meteorites and the earth itself were formed at the same time. With the results at present available one must conclude that iron meteorites are much older than stone ones.

" The same techniques that are used for measuring the age of meteorites has also been applied lately to the fragments of man-made satellites. A paper from a research group at the Smithsonian Astrophysical Observatory (Fireman, de Felice and Tilles) gave a survey of the work carried out there. The paper stressed that the production of radioactivity in satellites is quite different from its production in meteorites. In meteorites radioactivity is uniform; in satellites considerable variations are observed because of their excentric trajectories and their interaction with the Van Allen radiation belt. Samples of some of the Discoverer satellites, it was reported, had been analysed for tritium in lead, steel, and aluminium. These investigations have a direct bearing on the plans for manned space travel.

" Other papers described techniques for the use of C^{14} or other radionuclides produced by cosmic rays to date the time of the fall to earth of meteorites. The results so far available tend to prove that many meteorites

have had longer "terrestrial lives" than has hitherto been assumed. It was generally agreed that C^{14} provided a good tool for dating meteorites and that further developments in this technique would increase its accuracy and range, which might prove particularly interesting in dating prehistoric meteorite craters.

"To obtain the same kind of information that has resulted from the study of recently fallen meteorites by "direct" observation would require a two-year long space travel in an appropriately constructed space vehicle.

"The spatial and time distribution of cosmic radiation intensity were dealt with in several papers. In a paper from Brookhaven National Laboratory (Davis, Steenner, and Schaeffer), a method of comparing the activity of a short-lived radioisotope with a long-lived one to establish the spatial density of cosmic radiation was described. Argon-37, with a half-life of 35 days, and Ar^{39}, with a half-life of 325 years, were used. The results have led to the conclusion that the present cosmic ray intensity at one Astronomical Unit (the earth orbit) is within 15 per cent of the intensity at a distance of several Astronomical Units. The results also showed that the production of radionuclides by cosmic rays is independent of the meteorite's orbit. Special study has also been made of the effects of solar flares, as for instance the one that occurred in November 1960, comparing the Ar^{37} content of stainless steel samples from Discoverer 17, which was launched during the intense solar flare in November 1960, with samples from Discoverer 18 and 26.

"Another paper from the United States (Goel and Kohman) dealt with variations in cosmic ray intensity in time. Cosmic rays are generally

assumed to originate in stars or stellar systems and as these vary with time and position within the galaxy, long-term variations would be expected. The paper concludes, on the basis of Cl^{36} and Ar^{36} investigations, that cosmic ray intensity has been decreasing over the past few hundred million years. The research also indicates that erosion of the meteorite during its space travel has been "general and significant".

"These conclusions were different from those reached in two other papers. In a paper by two scientists from the Max-Planck Chemical Institute, Mainz, Federal Republic of Germany (Voshage and Hintenberger), it was concluded that the cosmic ray intensity in the last million years had been greater than the average during the entire life-span of the meteorites. Another paper from the same Institute (Vilosek and Wänke) concluded that space erosion had been rather small. According to this paper, the relatively short life of stone meteorites is not due to erosion, but to collisions in space which would be catastrophic for stone but not for iron. A Belgian-US paper (Crèvecour and Schaeffer) also dealt with this question and stated that definite conclusions as to the changes in cosmic ray intensity in time could be made only after such problems as the erosion of meteorites in space and the production rate of radio-isotopes in meteorites had been solved, although measurements of the Al^{26}/Be^{10} ratio indicated that cosmic radiation intensity had been constant in the last ten million years.

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V. APPLICATIONS IN AGRICULTURE

In addition to their application as tracers in studying metabolic processes, as mentioned in the section on research in biology and agriculture, radioisotopes are being investigated for use in eradicating insects, developing new varieties of plants, preventing loss of agricultural products during storage, and tracing movements of insects and small animals.[1] Since this field will be covered in the third issue of this quarterly progress review, it is discussed here only very briefly. Much irradiation work has been done with x rays, but Co^{60} and Cs^{137} are being used increasingly, and some experiments have been made with solutions of such isotopes as S^{35} and P^{32}, in which seeds, for example, can be soaked.

The most spectacular result of the use of radiation in agriculture is the eradication of the screwworm fly -- a pest that attacks farm animals in the southern U.S., Mexico, and Caribbean areas -- with an estimated annual saving of some 20 million dollars.[1c] Huge numbers of male flies were sterilized by irradiation and released, and females (which mate only once) mated with these, did not lay hatchable eggs. Other insects being investigated for such treatment include the boll weevil, European corn borer, mosquito, and tsetse fly.

Amateur gardeners are familiar with Co^{60}-irradiated seeds and bulbs, with 27 varieties of flower and vegetable seeds being advertised by a single company.[2] Even more important, of course, are the commercial crops that have been improved by irradiation from various sources: mustard, tobacco, beans, oil rape, fodder peas, navy beans, barley, peanuts, and oats. Various other crops are being studied. The

success of this procedure led to the development of a portable, 1-ton, Cs^{137} source, which can be used in the field.[3]

Irradiation of potatoes to prevent sprouting in storage has been under investigation for some time. No chemical change has been found in irradiated potatoes that would make them unsafe for eating, and Canada has recently approved this method of potato preservation. The effect of radiation on onions and other crops is being studied. Irradiation "pasteurization" (i.e., irradiation sufficient to kill most, but not all, of the organisms in products that are to be refrigerated) is also being actively investigated. Since the amount of radiation to pasteurize foods is only 5% of that required to kill all organisms, this procedure is cheaper and less damaging to flavor and texture than complete sterilization. Among the foods being investigated for such processing are corn, broccoli, strawberries, bananas, seafood, and meat. Irradiation is also being considered for killing insects and animal pests in stored crops.

Various other beneficial uses of radiation have been reported. Output of penicillin, which, of course, is not a strictly agricultural product, has been increased a thousandfold by repeated mutations resulting from irradiation of the microorganisms producing it. More and better silk is obtained when the silkworm in the cocoon is killed by irradiation instead of heat.[4] Russian scientists are reported to be irradiating silkworm ova to produce a higher percentage of males, which give more silk than females.[5] Caviar production has been reported to be increased by irradiation, and irradiation of pullets before hatching is reported to result in a higher egg production.[6]

The density of silage may be determined by external readings of the radiation passing through the silo contents from a Cs^{137} source placed in the center during filling.[7]

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VI. APPLICATIONS IN EDUCATION

Radioisotopes have been described as the most important tool for scientific research since the microscope. Although for many years research workers have accepted isotopes as an important tool, educators have been rather slow to add instruction in the use of isotopes to the chemistry, biology, and physics curricula. Of course, in a historical sense, the term "slow" is relative and the addition of instruction with radioisotopes to college courses probably has not been slow since it is doubtful that any colleges or universities gave instruction in the use of the microscope as soon as thirty years after its invention. The only reason we might expect to do better now is that communications have improved and the pace of scientific advances has become much faster.

The Division of Isotopes Development of the Atomic Energy Commission has sponsored projects to develop experiments for use in high school, college, and industrial courses, and descriptions of many such experiments are now available. Choppin, in A Cross Referenced Index of Radiochemical Teaching Experiments Applicable to Chemistry,[1] has summarized and indexed radiochemical experiments in 15 books that are readily available in the United States. His index divides the experiments into categories analytical chemistry, biochemistry, general and inorganic chemistry, organic chemistry, and physical chemistry; and breaks these broad subjects down into specific types of experiments. For instance, Physical Chemistry is divided into: crystal studies (surface area, exchange,etc.), determination of dissociation constants, kinetic and exchange studies, Szilard-Chalmers experiments, and radiation chemistry. This index is valuable for any teacher wishing to enrich his teaching with experiments using radioactive isotopes. Rather than simply duplicate this index,

this review will indicate the scope of educational material now available and will supplement the Choppin index in cases of items that it did not cover. Publications indexed by Choppin are designated by (C) when referred to in the text and in the bibliography.

Almost any modern chemistry, physics, or biology textbook will have some information that is basic to the use of radioisotopes. This kind of information is not covered here; instead, only educational material developed specifically for teaching the use of isotopes is reviewed. For the sake of simplicity, the material has been divided into three main sections describing the use of isotopes in (1) high schools, (2) colleges and universities, and (3) industrial training programs. The division into these three categories is arbitrary and there is considerable overlapping among them; therefore an experiment should not be ignored at the college level simply because it is listed in the high school section or visa versa. The section ends with a brief discussion of other educational aids such as training movies and charts.

License-exempt Isotopes

Under current AEC regulations, small but significant amounts of most radioisotopes may be bought under what is known as a "general license." This means that a teacher or student does not have to get a license from the AEC if he limits his use of isotopes to the amounts shown in Table VI-1. At any one time, no person shall possess more than a total of ten such scheduled quantities. Since these quantities are ample for most experiments, the job of educators in obtaining isotopes is greatly simplified.

Table VI-1 QUANTITIES OF ISOTOPES AVAILABLE WITHOUT AEC AUTHORIZATION

Radioisotope	Unsealed Source (μc)	Sealed Source (μc)
Antimony 124	1	10
Arsenic 76	10	10
Arsenic 77	10	10
Barium 140-Lanthanum 140	1	10
Beryllium 7	50	50
Cadmium 109-Silver 109	10	10
Calcium 45	10	10
Carbon 14	50	50
Cerium 144-Praseodymium 144	1	10
Cesium 137-Barium 137	1	10
Chlorine 36	1	10
Chromium 51	50	50
Cobalt 60	1	10
Copper 64	50	50
Europium 154	1	10
Fluorine 18	50	50
Gallium 72	10	10
Germanium 71	50	50
Gold 198	10	10
Gold 199	10	10
Hydrogen 3 (Tritium)	250	250
Indium 114	1	10
Iodine 131	10	10
Iridium 192	10	10
Iron 55	50	50
Iron 59	1	10
Lanthanum 140	10	10
Manganese 52	1	10
Manganese 56	50	50
Molybdenum 99	10	10
Nickel 59	1	10

Table IV-1 Continued

Radioisotope	Unsealed Source (μc)	Sealed Source (μc)
Nickel 63	1	10
Niobium 95	10	10
Palladium 109	10	10
Palladium 103-Rhodium 103	50	50
Phosphorus 32	10	10
Polonium 210	0.1	1
Potassium 42	10	10
Praseodymium 143	10	10
Promethium 147	10	10
Rhenium 186	10	10
Rhodium 105	10	10
Rubidium 86	10	10
Ruthenium 106-Rhodium 106	1	10
Samarium 153	10	10
Scandium 46	1	10
Silver 105	1	10
Silver 111	10	10
Sodium 22	10	10
Sodium 24	10	10
Strontium 89	1	10
Strontium 90-Yttrium 90	0.1	1
Sulfur 35	50	50
Tantalum 182	10	10
Technetium 96	1	10
Technetium 99	1	10
Tellurium 127	10	10
Tellurium 129	1	10
Thallium 204	50	50
Tin 113	10	10
Tungsten 185	10	10
Vanadium 48	1	10

Table VI-1 Continued

Radioisotope	Unsealed Source (μc)	Sealed Source (μc)
Yttrium 90	1	10
Yttrium 91	1	10
Zinc 65	10	10
All others not listed	1	10

Sealed sources in license-exempt quantities are available from most of the companies who sell isotopes. A number of companies also sell individual isotopes or educational kits in license-exempt quantities. These isotopes usually come in a solution of a simple compound such as chloride or carbonate, and the average cost is about $5 to $10 per isotope. The following are some of the companies who sell license-exempt isotopes specifically for educational purposes (other companies will be listed as we get information):

Abbott Laboratories, Oak Ridge, Tennessee

2-pack kit - P^{32}, I^{131}

9-pack kit - P^{32}, I^{131}, Cr^{51}, S^{35}, Zn^{65}, Ca^{45}, As^{74}, Hg^{203}, Co^{60}

Atomic Corporation of America, 14725 Arminta St., Panorama City, Calif.

Kits with all equipment including isotopes to do definite experiments. For example: "Bone Metabolism in Goldfish," or "Transport of Phosphates in Tomato Plants." They also sell the following individual isotopes: I^{131}, P^{32}, Ca^{45}, C^{14}, Cs^{137}-Ba^{137}, Cl^{36}, Cr^{51}, Co^{60}, Fe^{59}, Ni^{63}, Ru^{106}-Rh^{106}, Na^{22}, Sr^{89}, Sr^{90}-Y^{90}, S^{35}, Tl^{204}, Zn^{65}

Bio-Rad Laboratories, 32nd and Griffin Ave., Richmond, Calif. (This company is supplying free isotopes to science teachers for fiscal year 1963 under the program mentioned below.)

Individual Isotopes: C^{14}, Co^{60}, S^{35}, Fe^{59}, Zn^{65}, Radium-D, P^{32}, I^{131}

Nuclear-Chicago Corporation, 333 E. Howard Ave., Des Plaines, Ill.

Kit of 5 isotopes: Pb^{210}-Bi^{210}, Co^{60}, Ce^{144}-Pr^{144}, Zn^{65}, C^{14}*, *(As Na_2CO_3)

Individual Isotopes: The five above and C^{14} (as glycine, acetic acid, or acetic anhydride), S^{35} (in toluene or as sulfuric acid) P^{32}, Ca^{45}

Nuclear Consultants Corporation, 9842 Manchester Rd., St. Louis 19, Mo.

Kit of 10 isotopes: C^{14}, Co^{60}, Cs^{137}, Na^{22}, Zn^{65}, U^{238}, I^{131}, P^{32}, Cr^{51}, Fe^{59}

Each of the above isotopes can also be bought separately.

Radiation Equipment and Accessories Corporation, 665 Merrick Rd., Lynbrook, N. Y.

14-pack kit - Ba^{133}, Cs^{137}, Mn^{54}, C^{14}, Tl^{204}, S^{35}, Co^{60}, Fe^{55}, Co^{57}, Na^{22}, Pm^{147}, Zn^{65}, Uranyl nitrate, Ni^{63}

6-pack kit - Any of 6 above

Replacements - Any of 3 above

U. S. Nuclear Corporation, P. O. Box 208, Burbank, Calif.

Set of 3 - C^{14}, I^{131}, P^{32}

Set of 4 - Ce^{144}-Pr^{144}, Cs^{137}-Ba^{137}, Co^{60}, Uranyl nitrate

Set of 5 - Pb^{210}-Bi^{210}, Co^{60}, Ce^{144}-Pr^{144}, Zn^{65}, C^{14}

Set of 3 - Pm^{147}, Na^{22}, Sr^{90}-Y^{90}

Isotopes may also be ordered separately.

Isotopes in High School

With the increased emphasis on science in high schools has come pressure to upgrade the courses with "new and modern techniques." To meet this goal, the National Science Foundation and the Atomic Energy Commission are sponsoring summer institutes to teach high school science teachers how to use the modern tools of science. Radioisotopes have been emphasized in these institutes. In these courses the teachers are given

a kit of equipment and supplies that can be used in their own classrooms. Each year the U. S. Atomic Energy Commission contracts with one isotope supplier to furnish license-exempt isotopes free of charge to teachers who have taken the courses.

Experiments developed in these courses are, of course, excellent supplementary teaching material for high school chemistry, biology, or physics courses. A booklet, Laboratory Experiments with Radioisotopes for High School Science Demonstrations (C)[2]

includes 20 experiments ranging from a determination of the half-life of I^{131} to a study of the removal of P^{32} from water by goldfish. It was supplemented and extended by Teaching with Radioisotopes (C).[3] Since this book is tailored for use with kits furnished to teachers taking a summer institute course in radiobiology, most of the experiments are biological. There are many more quantitative experiments in this book than in the earlier book--typical ones being "Determination of Range and Energy of Alpha Particles," "Distribution of P^{32} in an Animal," and "Soil Adsorption and Leaching."

Nuclear-Chicago Corporation has compiled reprints of 36 papers which appeared in the Journal of Chemical Education (C)[4] between February 1949 and June 1959, and which describe experiments and methods for teaching with radioisotopes. Many of these experiments are suitable for use with high school classes.

"The Amateur Scientist" column in Scientific American for May 1960[5] described a number of experiments that could be performed in high school or even in a home laboratory. The advantage of these experiments is that in many of them the radioactivity is detected by autoradiography, thus eliminating counting equipment. For those who wish to make a Geiger counter, the article gives a simple circuit that can be assembled from commercial parts for about $25.

Scientists at Argonne National Laboratory have been giving a short course in basic nuclear science for high school teachers since 1957. This course, including both lectures and laboratory work, covers more than radioisotopes; however, it is a good introduction to the subject. Both the lectures and the laboratory experiments were recently published in

Introduction to Nuclear Science.[6] In addition to chemistry, biology, and physics experiments, this book also contains some metallurgy demonstrations, but these are more concerned with reactor fuel elements than with isotopes.

EXPERIMENTS IN "INTRODUCTION TO NUCLEAR SCIENCE"[6]

Physics

Ratio of charge to mass

Magnetic field assembly for e/m apparatus

Notes for a high-voltage power supply

Diffusion cloud chamber

Electroscope

Strontium bead sources

Demonstration of radioactive decay

Uranyl oxalate actinometer

Biology

Plant irradiation effects

Uptake and distribution of P^{32} in tomato plant

The response of yeast cells to irradiation

Effects of radiation of mammals

Chemistry

Liquid-liquid extraction separation

Coprecipitation experiment

Ion exchange separation

Determination of solubility product of silver iodide using iodine tracer

Experiment on isotopic exchange

Metallurgy

Safety precautions

Method of conducting experiments

Experiment 1: make uranium-aluminum alloy by melting the elements in a high-frequency induction furnace

Experiment 2: hot-rolling the uranium-aluminum alloy

Experiment 3: cast cladding

Experiment 4: rolling the fuel plate

Experiment 5: inspection of the fuel element

Samples for teaching aids

Isotopes in Colleges and Universities

There are two approaches to the introduction of isotopes into courses of instruction at both the undergraduate and graduate levels. In one, experiments using isotopes and radiation are incorporated into the traditional curriculum in places where isotopes are the best tool for a job. In the other, the use of isotopes is taught in a separate course or courses which cut across disciplinary lines. If isotopes are a tool like the microscope, then more and more isotope experiments should be added to the traditional courses. The number of college and university courses on the use of the microscope is extremely small, while the number of courses using microscopes is very large.

Some of the high school experiments mentioned above are also suitable for use in the enrichment of traditional college courses. In particular, a number of experiments in the Nuclear-Chicago collection[4] from the Journal of Chemical Education may be more suitable at the college level.

The Atomic Energy Commission and the Nuclear-Chicago Corporation have co-sponsored the preparation of a laboratory manual, which was designed expressly to introduce radioisotope experiments to the chemistry curriculum. The purposes and philosophies behind the preparation of this manual, Radioisotope Experiments for the Chemistry Curriculum (C)[7] are described by its editor, Radin, in the Journal of Chemical Education.[8] An instructor's manual is also available.[9]

TYPICAL RADIOISOTOPE EXPERIMENTS FOR THE CHEMISTRY CURRICULUM[8]

Introductory Chemistry:

Determination of atomic weights, with S^{35}-sulfate

Chemistry of sulfur; thiosulfate, sulfur, and sulfite

Secular equilibrium; Pb^{210}-Bi^{210}

Qualitative Analysis:

Determination of a solubility product for calcium sulfate

Separation of ions by liquid/liquid extraction

Coprecipitation phenomena; calcium and permanganate with barium sulfate

Quantitative Analysis:

Determination of phosphate in a phosphate rock by an isotopic yield method

Determination of the formation constant of a complex ion; calcium citrate vs an ion exchange resin

Determination of calcium; removal of interference by ion exchange and chelatometric titration

Organic Chemistry:

Synthesis of S^{35}-sulfanilamide

Synthesis of C^{14}-acetylsalicyclic acid

Identification of amines by acetylation with C^{14}-acetic anhydride

Physical Chemistry:

Dynamic nature of equilibria between ions in solution and a salt

Exchange between a metal and ions in solution

Analysis of the phase diagram of a three component system; dimethylaniline, water, acetic acid-C^{14}

Demonstration of the adsorption isotherm with charcoal and acetic acid

Radioactive decay and growth; Ce^{144}-Pr^{144}

Instrumental Analysis:

Preparation of samples for Geiger counting

Properties of Geiger counters

Statistical variation in radioactivity measurements

Biochemistry:

Ion exchange and paper chromatography; separation of C^{14}-glycine and glutamic acid; isotopic dilution determination

Properties of enzymes, studied with an isotopic substrate made by the student

Incorporation of C^{14}-glycine into a protein fraction of mouse spleen; properties of the in vitro system

The William H. Johnston Laboratories, under an AEC contract, have developed a series of experiments on radiation chemistry which are intended to be used as supplementary material to other courses.[10,11] These experiments require the use of a 25-curie Co^{60} source, for which they have supplied detailed engineering drawings.[12]

TRAINING EXPERIMENTS IN RADIATION CHEMISTRY[10]

Interaction of alpha particles with matter

Interaction of electrons with matter

Interaction of gamma rays with matter

Fricke dosimeter

Gas-liquid chromatography and chemical effects of radiation

Radical trapping I: radiolysis of water and decolorization of methylene blue

Radiolysis of methane

Polymerization of styrene

Radical trapping II: sol-gel cultures and radiation damage to living cells

Viscometric determination of the radiation degradation of polymethylmethacrylate in solution

A second volume[11] presents the theories on which these experiments are based and includes a reprint from the Journal of Chemical Education of the Lind Jubilee Symposium on the development of radiation chemistry.

Johnston Laboratories have also developed a number of pieces of equipment (Table VI-2) for use in college and university laboratories for teaching with radioisotopes.[13] The aim in developing these systems was to provide training laboratories with equipment of higher training value, lower cost of construction, and greater general usefulness than present in existing commercial equipment.

A number of textbooks and laboratory manuals have been prepared to teach the use of isotopes in college or university courses. Choppin has indexed the following textbooks intended for use in courses on isotope methodology: Experimental Nucleonics,[14] Principles of Radioisotope Methodology,[15a] Experimental Nuclear Chemistry,[16] Modern Radiochemical Practice,[17] Radioisotope Techniques,[18] and A Laboratory Manual for

Table VI-2 SYSTEMS AND EQUIPMENT FOR RADIOISOTOPE TRAINING[13]

Title	Drawing Number
Radiochemical instrumentation	
Liquid scintillation counting of C^{14} and H^{3}	CAPE-812
Low level beta counting	CAPE-814
Monitoring of natural radon	CAPE-815
Tracer leak detection	CAPE-816
Gamma ray spectrometer	CAPE-817
Solid state alpha detector	CAPE-818
Industrial gauging	
Beta thickness gauging	CAPE-811
Density and level gauging	CAPE-813
Tracer flow rate training system	CAPE-819
Tracer wear studies	CAPE-820
Industrial research and process analysis	
Gas chromatography and radiotracer system	CAPE-821
Carbon-hydrogen analyzer	CAPE-822

These drawings are described and listed in Unclassified Engineering Materials List, TID-4100, Supplements 13-16, and are available in packets from Oak Ridge Reproduction Service, P. O. Box 363, Oak Ridge, Tennessee.

Radiochemistry.[19] Many of the experiments in these courses could also be used as supplementary material in traditional courses, and this index is a good guide to use in choosing experiments.

Principles of Radioisotope Methodology[15b] was revised in 1962 and Chase has added several experiments not listed in Choppin's index. In his index Choppin included the contents of his own book (see p. of this issue), Experimental Nuclear Chemistry,[16] which was probably in press at the time.

Most of the books that have been written for practical training in radioisotope techniques have emphasized chemistry and physics. Choppin's index is confined to chemistry; however, he does list some parts of Techniques of Radiobiochemistry.[20] This book is in two parts; the first deals with general principles of techniques and methods and the second with experiments in radiobiology, with emphasis on C^{14} work. Isotopic Tracers[21] and Radioisotopes in Biology and Agriculture[22] are also useful as textbooks to teach the use of radioisotopes in biology.

The first part of Isotopic Tracers discusses the theoretical considerations in the use of radioisotopes in biological systems, and the second part is a practical laboratory course. Following a series of basic physics experiments on counting methods, statistics, and half-lives, a number of biological experiments are given.

BIOLOGY EXPERIMENTS IN ISOTOPIC TRACERS[21]

Determination of atom per cent excess N^{15} in a sample of an ammonium salt enriched with N^{15}

Incorporation of P^{32} into hens' eggs during their development

Persistence of isotopically labeled red cells in the bloodstream

after injection, and calculation of the circulating red cell volume and blood volume

Labeling of red cells simultaneously with P^{32} and Cr^{51}

Use of I^{131}-trace-labeled proteins (a) to study the persistence in the bloodstream of injected proteins, and (b) to determine the plasma volume of an animal

Determination of the intercellular plasma of centrifuged red blood cells

Use of a trace-labeled protein antigen in the determination of the composition of serological precipitates

Distribution of P^{32} in the tissues following the injection of P^{32}-containing inorganic phosphate into mice

Incorporation of C^{14}-labeled acetate into the lipids of rat liver

Determination of exchangeable sodium in rabbits, using Na^{24}

Experiments on thyroid

Measurement of thyroid activity by determination of protein-bound radioiodide in the blood after injection of I^{131}

Localization of injected I^{131} in the thyroid

Paper chromatography with I^{131}-containing urines

Experiments with radioactive iron

Binding of Fe^{59} by plasma proteins

Persistence of plasma-bound iron in the circulation

Comar's book,[22] is not a laboratory manual since it contains no specific laboratory experiments. Instead, it discusses basic techniques and principles for experimentation with animals and plants. The subjects covered are indicated quite well by the chapter headings.

RADIOISOTOPES IN BIOLOGY AND AGRICULTURE

Principles of Tracer Methodology

Basic Difficulties in Tracer Methodology

Health Physics and Radiation Protection

Facilities and Handling of Radioisotopes with Animals and Plants

General Procedures for Radioassay

Properties and Procedures for Individual Radioisotopes

Autoradiography

Paper Chromatography

Ion Exchange

Radioactivation Analysis

The chapter on Properties and Procedures for Individual Radioisotopes contains much information about individual isotopes that were available from Oak Ridge National Laboratory when the book was written. Although the prices and some of the specific activities are no longer valid, the book still contains a large number of useful data. For each element, intake levels (toxicity), radioassay method, chemistry, and typical methods for use in biology are given.

Industrial Training

The line between industrial training manuals and books intended for academic use frequently is either rather fine or nonexistent. For example, Radioisotope Techniques[18] is used in an academic course at Rensselaer Polytechnic Institute and also for industrial courses at Oak Ridge Institute of Nuclear Studies. We have arbitrarily categorized these manuals according to what appears to have been the primary goal of the author.

Choppin lists two industrial training manuals in his index in addition to Overman and Clark's book: Radioisotopes in Industry Training Program,[23] and Short Course in Basic and Applied Isotope Technology.[24] Radioisotopes in Industry Training Program is based on a course, given at the General Motors Research Laboratories, which comprises 45 lectures requiring 170 hr and 20 experiments requiring 135 hr. Short Course in Basic and Applied Isotope Technology was prepared by the William H. Johnston Laboratories. It includes 26 experiments that are intended to be the background of a six-week course in isotope technology for scientific personnel with the equivalent of at least a bachelor's degree.

Radioisotope Laboratory Techniques[25] is used in the Isotope School at Harwell, England. This book covers many aspects of laboratory work with isotopes, but it is not a "cookbook" type of laboratory manual. It would be most valuable if used by the student for background prior to work in the laboratory or by the teacher in preparing a laboratory manual. The subjects covered are shown by the chapter headings:

RADIOISOTOPE LABORATORY TECHNIQUES

Elements of Nuclear Physics

Properties of Radiations

Production of Radioisotopes

Introduction to Health Physics

The Laboratory

Hazard Control

Decontamination and the Disposal of Waste

Laboratory Apparatus

Introduction to Electronic Techniques

Detection Devices: Integrating Types

Particle Detectors and Their Use

Associated Equipment

Errors and Corrections

The Statistics of Counting

The Choice of Counting Equipment

Special Counting Techniques

Feasibility (Will a radioisotope do the job at hand?)

Some Chemical Applications

Autoradiography and Gamma Radiography

Applications of Isotopes

Radiochemical Procedures: Work with Radioisotopes[26] is a translation of a Yugoslav textbook, written in 1959, intended for use by students in the School for Training of Personnel in Handling Radioactive Materials, conducted by the Federal Nuclear Energy Commission of Yugoslavia. This book is used in three different courses: (1) Fundamentals of Radioactivity and the Possibilities of Their Practical Application, (2) Introduction to Basic Operating Techniques, Specifically to the Techniques for the Practical Application of Radioisotopes, and (3) Introduction to the Most Important Safety Measures in Handling Radioactive Materials. The text is divided into ten main subject headings:

RADIOCHEMICAL PROCEDURES: WORK WITH RADIOISOTOPES

General Concepts

The Measurement of Nuclear Radiation

Physical Methods in Radiochemistry

Chemical Methods in Radiochemistry

The Radiochemical Laboratory and the Principles of Work with Radioactive Isotopes

Dosimetry

The Effect of Radiation on the Organism

Some Examples of the Application of Radioisotopes

Chemical Changes Caused by Radiation

Appendix (numerical data)

This book is chiefly of interest as a source of information on the type of training courses given in another country.

Stanford Research Institute, under a contract with the Atomic Energy Commission, has developed three industrial training courses concerned with radioisotopes, radiation, and atomic power entitled: Survey of Nuclear Fundamentals and Radioisotope Applications in Industry,[27] Survey of Nuclear Energy and Power Plant Applications,[28] and Radiation Safety.[29] These courses, in the form of lectures, could be used as background material for a laboratory course on the use of radioisotopes, or they could be used without laboratory experiments as background material for workers in the field. These courses, each of which requires about 32 hr of lecture time to present, are designed for laymen without a scientific background. The instructors' manuals for these courses provide excellent source material for teachers in almost any aspect of isotope and radiation technology at a popular level. A number of good charts are provided as well as good lists of reading material and movies at a popular level. The philosophy, aims, and background considered in the development of these courses are given in other reports,[30,31,32] and the courses have been evaluated for use with union workers,[33] adult education programs,[34] and workers in a naval shipyard.[35]

SURVEY OF NUCLEAR FUNDAMENTALS AND RADIOISOTOPE APPLICATIONS IN INDUSTRY[27]

Introduction (1 hour)

Some Principles of Atomic Structure (3 hours)

- The Structure of Matter
- The Structure of the Atom and Atomic Classification
- Isotopes

Some Principles of Radioactivity (4 hours)

- Nature and Types of Radioactivity
- Radioactivity Measurement Units
- Effects of Radioactivity
- Uses of Radioactivity

Radiation Safety (4 hours)

- Hazards of Radiation
- Control of Radiation
- Detection and Monitoring
- Safety Practices in the Use of Radioisotopes

Radiography (4 hours)

- Fundamentals of Radiography
- Radiographic Techniques
- Special Factors Related to Radiography
- Industrial Uses

Radiation Gages (4 hours)

- Fundamentals of Radiation Gaging
- Types of Radiation Gages and Suitability for Production Control

Procedures in Using Radiation Gages

Industrial Uses

Radioisotope Tracers (2 hours)

Principles

Factors in Using Tracers

Other Applications of Radiation Sources (2 hours)

Well Logging

Industrial Radiation Processing

Handling Radioactive Materials (4 hours)

Sealed and Unsealed (Open) Sources

Transportation

Waste Disposal

Accident Procedures

Radioisotopes Utilization and People (2 hours)

Job Opportunities

Labor Relations and Public Relations

Review and Examination (2 hours)

Other Training Materials

The Atomic Energy Commission has an active program of assistance to provide teachers and students with information on the various phases of atomic energy. Students and teachers may write to the Commission's Division of Technical Information Extension, Educational Materials Section, Reference Branch, P. O. Box 62, Oak Ridge, Tennessee, and receive available information. Currently, the Educational Materials Section has over 200 different items in 20 subject categories. They have a good collection of information on isotopes.

The Division of Isotopes Development has prepared a series of about one hundred-fifty 8 by 10-in. line drawings, which are available without charge from them or from the Division of Technical Information Extension, Oak Ridge, Tennessee. One of these charts is shown in Figure VI-1 to illustrate general style and quality. Essentially all aspects of isotope work are covered graphically by these charts, and they are very useful in teaching about isotopes at almost any level. Several of these charts are also used in other sections of this review.

Many 16-mm moving pictures have been made on isotope technology in the past 15 years. Most of these films are available from the Atomic Energy Commission's Motion Picture Libraries and can be obtained on loan free of charge for educational use. These films are listed in two pamphlets,[36,37] which are available from the Commission. The following moving pictures of particular interest as visual aids in teaching about isotopes were released in 1961 and 1962:

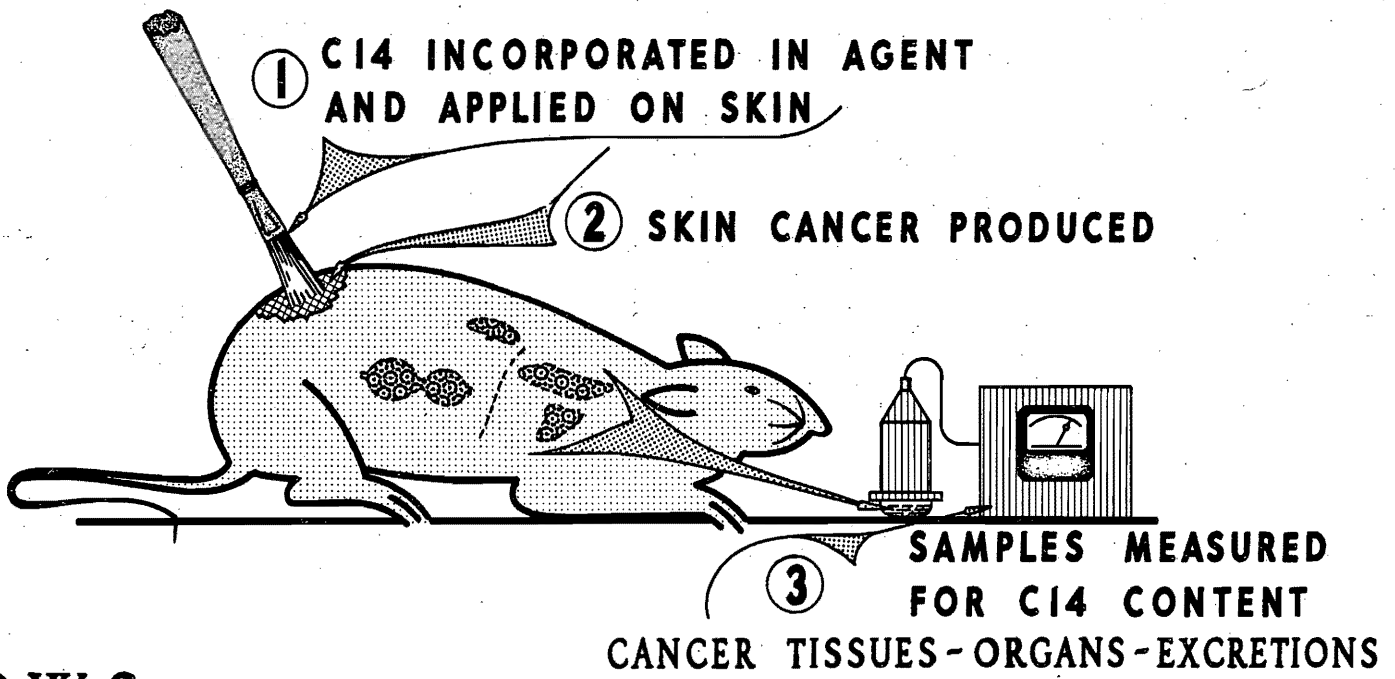
OUR NEAREST STAR (Isotopic Power System for the Transit Satellite). 12 minutes, 16mm, color (1961). A SNAP isotopic power system has been placed in orbit aboard the Transit Four-A navigation satellite. This simple, powerful device is the first application of nuclear power in space. Against a background of the Transit Program, this semitechnical film follows the development and testing of the radioisotope fuel capsule and the thermoelectric generator that make up this SNAP system.

ATOMIC WEATHERMAN: STRONTIUM-90 ISOTOPIC APPLICATIONS. 18-1/2 minutes, 16mm, color (1961). This semitechnical film describes the world's first radioisotope-powered weather station, which is operating

Figure VI-1

RADIOACTIVE CARBON – C14

FOR STUDYING CANCER PRODUCING AGENTS - CARCINOGENS



SHOWS:

1 - AMOUNT OF AGENT IN CANCER AND OTHER TISSUE
2 - LOCATION OF BREAKDOWN PRODUCTS OF THE AGENT
3 - MODE OF ACTION OF CANCER PRODUCING AGENT

USAEC-ID-189

unattended at a remote site in the Canadian Arctic. The "atomic" weather station is powered by a thermoelectric unit in which the heat from the decay of strontium-90 (Sr^{90}) is directly converted into electricity.

OPPORTUNITY UNLIMITED: FRIENDLY ATOMS IN INDUSTRY. 28-1/2 minutes, 16mm, color (1962). This popular-level film, narrated by news commentator John Daly, surveys the widespread use of radioisotopes by American industry to make better products--from ships to nylon hose--more efficiently and with an impressive record of safety.

UNDERSTANDING THE ATOM: ALPHA, BETA, AND GAMMA. 44 minutes, 16mm, black and white (1962). The film gives some insight into the origin and nature of alpha, beta, and gamma radiation.

UNDERSTANDING THE ATOM: RADIATION AND MATTER. 44 minutes, 16mm, black and white (1962). The film, which considers the interaction of radiation with matter, develops the various processes by which alpha, beta, and gamma radiation give up energy to their surroundings.

UNDERSTANDING THE ATOM: RADIATION DETECTION BY IONIZATION. 30 minutes, 16mm, black and white (1962). The basic principles of ionization detectors are described, particularly in relation to the pulse height as a function of voltage curves. Brief descriptions of ionization chambers, proportional counters, and Geiger counters are included, and examples of instruments operating in these regions are shown.

UNDERSTANDING THE ATOM: RADIATION DETECTION BY SCINTILLATION. 30 minutes, 16mm, black and white (1962). A short review of gamma interactions with matter is shown, with particular reference to useful scintillation crystals. The scintillation process is described, and the efficiency of the conversion of gamma radiation to visible light in the scintillator is discussed.

UNDERSTANDING THE ATOM: PROPERTIES OF RADIATION. 30 minutes, 16mm, black and white (1962). This film includes a discussion of general problems of radiation decay, such as the laws of radioactive decay, including the concept of half-life. Statistical considerations are introduced, and the basic notion of the standard deviation in counts expected in various experiments is described.

CHALLENGE: INVISIBLE BULLETS. 29 minutes, 16mm, black and white (1962). The meaning of radiation, its natural sources, the various forms it takes, the difference between alpha and beta particles and between gamma and x rays, how radiation is used in research.

CHALLENGE: WORKING WITH RADIATION. 29 minutes, black and white (1962). Precautions used in working with radiation, and research to learn more about handling radiation. Illustrated are "hot caves" with mechanical slave manipulators or electric manipulators, and giant "caves" using heavy duty manipulators. Demonstration outlines method of disposing of radioactive wastes.

CHALLENGE: TRACING LIVING CELLS. 29 minutes, black and white (1962). The use of radioactive isotopes in the study of cell division and in

medical therapy; use of radioactive tracers in blood and cancer research.

CHALLENGE: THE ART OF SEPARATION. 29 minutes, black and white (1962). With radiation, the chemist is able to work with much greater speed and ease in the field of chromatography--the separation of chemical compounds into basic substances. Basic principles are explained, with a demonstration of an actual separation of a chemical compound.

CHALLENGE: FOUNDATIONS FOR THE FUTURE. 29 minutes, black and white, (1962). Discussion of what the future will bring in the various areas of basic nuclear research--the effects of radiation, the peaceful uses of radiation, and the dangers of radiation.

Another source of educational material which should not be overlooked is the technical bulletins issued by companies selling isotopes. It is not possible to give a complete listing of this type of information, but some are mentioned to illustrate the wide variety of available material.

Nuclear-Chicago Corporation has prepared a number of bulletins that emphasize techniques for working with radioisotopes. The titles of the bulletins indicate some of the subjects covered. A bulletin of particular interest, Diagnostic Applications of Radioactive Isotopes, deals with the medical applications of isotopes.

NUCLEAR-CHICAGO BULLETINS

1. How to Use Radioisotopic Yield Determination in Quantitative Analysis
2. How to Use Radioisotopic Analysis for Mixing Evaluation

3. How to Use the Radioisotopic Derivative Method in Quantitative Analysis

4. How to Use Radioactivity in Paper Chromatography

5. How to Evaluate an Analytical Method with the Aid of Radioisotopes

6. How to Make Quantitative Determinations by Radiometric Analysis

7. How to Prepare Radioactive Samples for Counting on Planchets - Part 1

7B. How to Prepare Radioactive Samples for Counting on Planchets - Part 2

8. How to Compute Absorption and Backscattering of Beta Rays

10. Quantitative and Qualitative Analysis through Neutron Activation

11. How to Prepare Samples for Liquid Scintillation Counting

12. How to Compute Neutron Transmission and Diffusion

13. How to Determine Efficiency Automatically in Liquid Scintillation Counting

14. How to Apply Statistics to Nuclear Measurements

15. Continuous Scintillation Counting of Weak Beta Emitters in Flowing Aqueous Streams

Bulletins 1-7B have also been issued combined in one document, Analytical Procedures with Radioactive Isotopes.

New England Nuclear Corporation publishes a monthly bulletin, Atomlight, which generally features articles on the biological or medical aspects of isotope use.

Abbott Laboratories have published several bulletins on medical uses of isotopes. These bulletins include:

Radioisotopes in Medicine: A Guide for Physicians and Hospital Personnel, April 1962

Nuclear Medicine: for the Modern Physician and his Hospital, 1956

Radio-iron Preparations, July 1962

The Isotope Research Division of the United Kingdom Atomic Energy Authority, is now publishing a series of single page bulletins on various phases of isotope use. These may be obtained from the Wantage Research Laboratory (AERE), Wantage, Berks., England.

BULLETINS ISSUED BY UNITED KINGDOM ATOMIC ENERGY AUTHORITY TO MARCH 1, 1963

B.6 Neutron Detectors

B.7 Semi-Conductor Radiation Detectors

B.10 Statistics of Measurement

C.1 Radioactive Tracers; Physical Principles

C.2 Principles of Tracer Technique (Chemical)

C.4 Radioactivation Analysis

C.6 Measurement of Liquid Flow

C.10 Measurement of Bulk Flow and Mixing Efficiency

C.12 Filter Testing

C.13 Siltation Studies

C.14 Ground Water Tracing

D.1 Thickness and Density Gauges (Transmission)

D.2 Thickness and Density Gauges (Backscattering)

D.7 X-ray Fluorescence Spectrometry

E.1 Advice and Services Available in the UK

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4. Nuclear-Chicago Corporation, A Collection of Papers from the Journal of Chemical Education Related to Training and Experiments in Radioactivity, June 1959, 114 p. (C)

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10. William H. Johnston Laboratories, Inc., Training Experiments in Radiation Chemistry, Experiments, USAEC Report JLI-650-2-1 (Vol. I) June 1960, 171 p., OTS $2.75.

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15b G. D. Chase, J. L. Rabinowitz, Principles of Radioisotope Methodology, 2nd ed., Burgess Publishing Co., Minneapolis, 1962, 372 p., $6.00.

16. G. R. Choppin, Experimental Nuclear Chemistry, Prentice-Hall, Inc., Englewood Cliffs, N. J., 1961, 226 p., $10.00; text ed. $7.50. (C)

17. B. G. Cook, J. F. Duncan, Modern Radiochemical Practice, Clarendon Press, Fair Lawn, N. J., 1952, 407 p., $8.40. (C)

18. R. T. Overman, H. M. Clark, Radioisotope Techniques, McGraw-Hill Book Co., New York, 1960, 476 p., $10.50. (C)

19. A. F. Voigt, A Manual for Radiochemistry, College Book Store, Iowa State University, Ames, Iowa, 1960, $2.40. (C)

20. S. Aronoff, Techniques of Radiobiochemistry, Iowa State College Press, Ames, 1956, 228 p., $5.95. (C)

21. G. E. Francis, W. Mulligan, A. Wormall, Isotopic Tracers, 2nd ed., The Athlone Press, London, 1959, 524 p., $8.40.

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23. J. P. Danforth, R. P. Stapp, Radioisotopes in Industry Training Program, General Motors Institute, Flint, Mich., 1959, 2 vol., 400 p. each, $5.00 each. (C)

24. R. W. Kiser, A. F. Findeis, H. M. Rosenstock, N. W. Desrosier, W. H. Johnston, Short Course in Basic and Applied Isotope Technology, USAEC Report JLI-650-1-3, William H. Johnston Laboratories, Inc., 1960, 216 p., OTS $3.00. (C)

25. R. A. Faires, B. H. Parks, Radioisotope Laboratory Techniques, 2nd ed., George Newnes Ltd., London, 1960, 244 p., $3.59.

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27. L. H. Towle, Instructors Guide: Survey of Nuclear Fundamentals and Radioisotope Applications in Industry, USAEC Report SRIA-19, Stanford Research Institute, July 1960, 91 p., OTS $0.75.

38. H. M. Vollmer, B. J. Maynard, Instructors Guide: Survey of Nuclear Energy and Power Plant Applications, Revised Edition, USAEC Report SRIA-11a, Stanford Research Institute, Sept. 1959, 75 p., OTS $0.50.

29. L. H. Towle, Instructors Guide, Radiation Safety, USAEC Report SRIA-21, Stanford Research Institute, July 1960, 92 p., OTS $0.75.

30. H. M. Vollmer, L. H. Towle, B. J. Maynard, Development of a Training Program for Radioisotope Workers in Industry, USAEC Report SRIA-20, Stanford Research Institute, July 1960, 125 p., OTS $0.75.

31. H. M. Vollmer, Development of Training Programs for Operating and Maintenance Personnel in Nuclear Power Generation Systems, USAEC Report SRIA-4, Stanford Research Institute, Feb. 1959, 71 p., OTS $0.45.

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VII. STABLE ISOTOPES

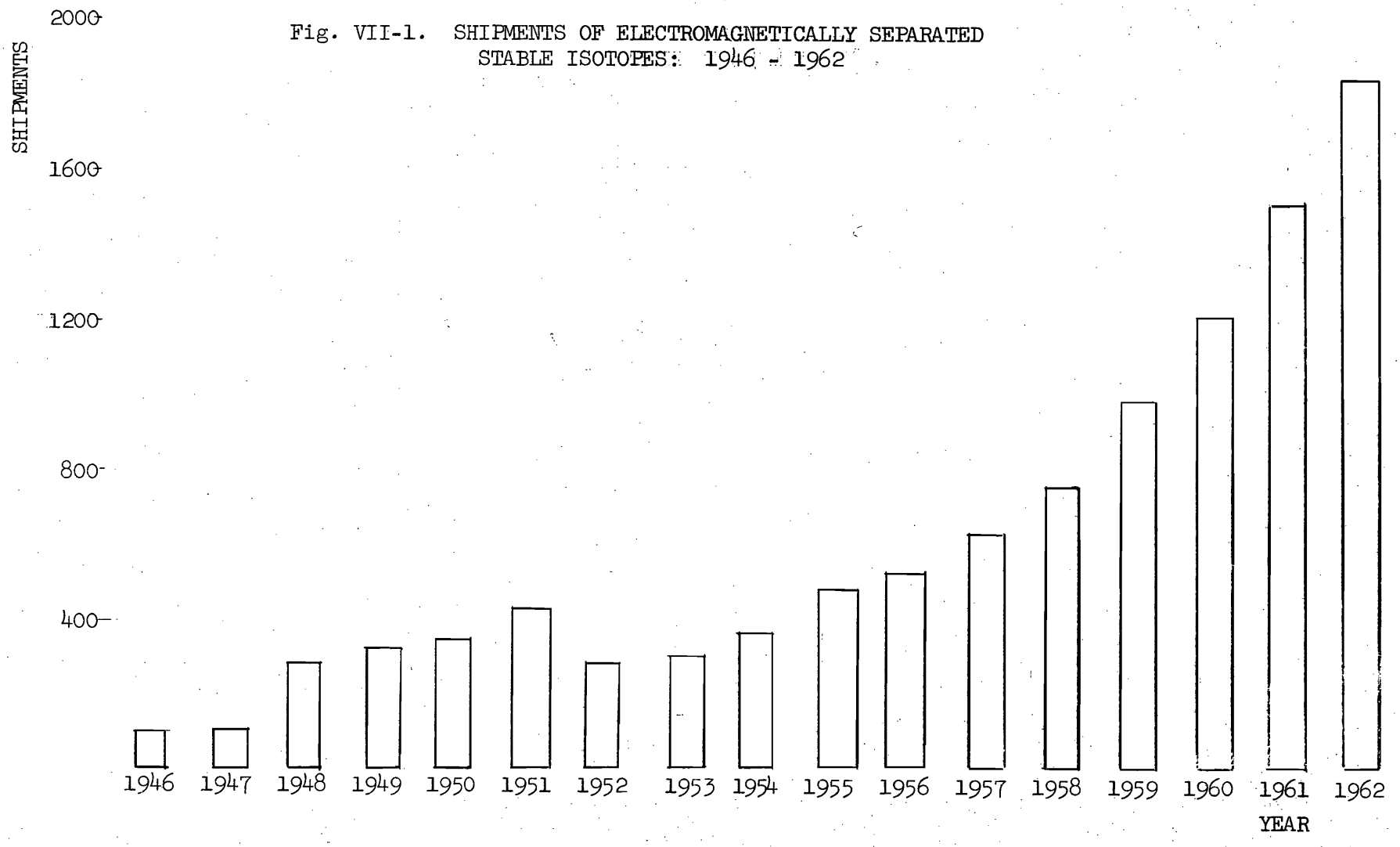
Although popular conception of the atomic energy program associates the isotopes business with the glamorous "twinkling atoms," the increasing importance of separated stable isotopes should not be overlooked; these silent partners play a rather important role in the overall applications program. Figure VII-1 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. These methods will be discussed more thoroughly in the second issue of this review.

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 reduced extraneous activity usually results from both increased concentration of desired target nuclide and decreased concentration of undesired target nuclide. Table VII-1 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}. The 4.5-day isotope when prepared from the 0.003% Ca^{46} in normal calcium is

Fig. VII-1. SHIPMENTS OF ELECTROMAGNETICALLY SEPARATED STABLE ISOTOPES: 1946 - 1962



completely overshadowed (by a ratio of about 25:1) by the 164-day Ca^{45} produced by the 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 VII-1. 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 VII-2 shows some typical uses for stable nuclides.[1] A recent bibliography covering these and other applications is included in ORNL 3266.[2]

Table VII-2.* 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.[2a]

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.[3] Today much of the effort is involved in the various cross-section studies.[4,5] However, very interesting work continues on energy levels,[6] decay schemes,[7] and half-lives,[8] and on such other topics as hyperfine structure,[9] the measurement of bulk properties,[10] and Mössbauer studies.[11,12]

The use of enriched stable isotopes in isotope dilution as an analytical tool has good potential.[13] Their use in tracer work involving eventual activation analysis is also a powerful tool.[14] 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. This is also mentioned under "Medical Uses of Radioisotopes."

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VIII. 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.)

Dans la preface, le docteur M. Tubiana, Professeur Agrégé à la Faculté de Médecine de l'Université de Paris, dit,((Les auteurs] ont réussi ... à écrire un livre où le spécialiste trouvera un grand nombre de données utiles et où le non-spécialiste pourra sans être rebuté s'initier à ces méthodes.)) Certainement, beaucoup d'information est contenu dans le livre. Pour le novice il-y-a un discussion de la structure du noyau atomique, de la décroissance radioactive, et de l'activité spécifique. Pour le travailleur scientifique il-y-a une liste convenable des principaux radioéléments utilisés en biologie et médecine avec leurs périodes de demi-vie et leurs énergies de β et γ.

Bienque ce petit livre n'est pas documenté, il forme un excellent précis de l'utilisation des radio-traceurs dans l'étude des maladies du sang, de la circulation sanguine, de la digestion et de l'absorption intestinale, de l'eau et des electrolytes, l'exploration de la fonction thyroidienne, le diagnostic des tumeurs et l'application des isotopes radioactifs en thérapeutique.

Le livre ne présente pas trop de difficultes pour le technicien de langue anglaise qui ne parle pas beaucoup de français. (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.)

BOOKS RECEIVED BUT NOT REVIEWED IN THIS ISSUE

Radioactive Isotopes in Medicine and Biology: Medicine, S. Silver, Lea & Febiger, Philadelphia, 1962, $8.00.

Radioactive Isotopes in Medicine and Biology: Basic Physics and Instrumentation, E. H. Quimby and S. Feitelberg, Lea & Febiger, Philadelphia, 1963, $8.00

Le cesium-137 in téléthérapie, R. Amalric and J. P. Vigne, Gauthier-Villars, Paris, 1962, NF 20.

La Télécobalthérapie en cancérologie, H. Pourquier, Masson et Cie, Paris, 1962, NF 60.

Telecobaltoterapia, G. Pisani, C. Cortissone, G. Savino, A. Malaspina, Edizioni Minerva Medica, Turin, 1962, L 5000.

Progress in Radiation Therapy, Vol. II, F. Buschke, Grune & Stratton, New York, London, 1962, 266 pp., $12.50.

Immediate and Low-level Effects of Ionizing Radiations, A. A. Buzzati-Traverso, Taylor & Francis, London, 1959.

Radiation Biophysics, Howard L. Andrews, Prentice-Hall, Inc., Englewood Cliffs, N. J., 1961, 328 pp., $11.65.

Radiation Therapy, Walter T. Murphy, W. B. Saunders Co., Philadelphia, Pa., 1959, 1041 pp., $25.00.

Basic Principles of the Tracer Method, C. W. Sheppard, John Wiley & Sons, Inc., New York, 1962, 282 pp., $8.00

International Directory of Radioisotopes, International Atomic Energy Agency, Johannes Kmoch, Vienna, Austria, 1962, 700 pp., $9.00.

Principles of Radioisotope Methodology, G. D. Chase and J. L. Rabinowitz, Burgess Publishing Co., Minneapolis, Minn., 1962, 2d ed., 372 pp., $6.00.

Advances in Tracer Methodology, Vol. 1, S. Rothchild, Ed., Plenum Press, New York, 1962, 332 pp., $12.00.

Advances in Nuclear Science and Technology, Vol I, E. J. Henley and Herbert Kouts, Academic Press, New York, 1962, 355 pp., $12.00.

Les Applications Industrielles des Radioéléments, A. Blondel, R. Colas, R. Cornuet, R. Hamelin, A. Kohn, A. Lamm, P. Leclerc, P. Lévêque, M. Quesson, R. Rabot, P. Rochas, J. Thiery, Éditions Eyrolles 61, boulevard Saint-Germain, Paris, 1962, 352 pp., $9.30.

Radioisotopes in the Physical Sciences and Industry III, International Atomic Energy Agency, Globus, Druck- und Verlagsanstalt, Vienna, Austria, 1962, 640 pp., $8.00.

IX. RECENT NEWS ITEMS

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 March 1963, concentrates of the mid-position isotopes, 10% Ne^{21} and 4% Ar^{38}, became available in very limited amounts.

Dr. Martin D. Kamen, co-discoverer of C^{14}, gets the nod for the American Chemical Society award for Nuclear Applications in Chemistry sponsored by Nuclear-Chicago Corp. Dr. Kamen is Professor of Chemistry at the University of California, San Diego.

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, and Texas for transferring to the states some of its regulatory responsibility for licensing, rule-making, and enforcement in the uses of radioisotopes. A similar agreement proposed by Arkansas is being considered.

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