LOCKHEED MARTIN ENERGY RESEARCH LIBRARIES
3 4456 0514058 5

CENTRAL RESEARCH LIBRARY
DOCUMENT COLLECTION

OAK RIDGE NATIONAL LABORATORY
operated by
UNION CARBIDE CORPORATION
for the
U. S. ATOMIC ENERGY COMMISSION

ORNL - TM - 2807

42

RADIOISOTOPE PROGRAM (8000) PROGRESS REPORT

FOR OCTOBER 1 THROUGH NOVEMBER 30, 1969

A. F. Rupp

OAK RIDGE NATIONAL LABORATORY
CENTRAL RESEARCH LIBRARY
DOCUMENT COLLECTION
LIBRARY LOAN COPY
DO NOT TRANSFER TO ANOTHER PERSON
If you wish someone else to see this document, send in name with document and the library will arrange a loan.
UCN-7969
(3 3-67)

NOTICE This document contains information of a preliminary nature and was prepared primarily for internal use at the Oak Ridge National Laboratory. It is subject to revision or correction and therefore does not represent a final report.

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

ORNL-TM-2807

Contract No. W-7405-eng-26

ISOTOPES DEVELOPMENT CENTER

RADIOISOTOPE PROGRAM (8000) PROGRESS REPORT FOR OCTOBER 1 THROUGH NOVEMBER 30, 1969

A. F. Rupp

DECEMBER 1969

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee
operated by
UNION CARBIDE CORPORATION
for the
U.S. ATOMIC ENERGY COMMISSION

LOCKHEED MARTIN ENERGY RESEARCH LIBRARIES

3 4456 0514058 5

CONTENTS

Page

RADIOISOTOPE PRODUCTION AND MATERIALS DEVELOPMENT 1

NEUTRON PRODUCTS - 08-01-01 1

A. Biomedical Radioisotopes 1

1. Thulium-171 1
2. Phosphorus-33 1
3. Potassium-43 2
4. Rubidium-84 4

B. Research Radioisotopes 5

1. Lanthanum-137 5

C. Isotopic Power 5

1. Thulium-170 5

D. Miscellaneous 6

1. Terbium-157 6

E. Supporting Technology 8

1. Radioisotope Characterization 8
2. Radioisotope Special Analysis and Quality Control . . 8

CYCLOTRON PRODUCTS - 08-01-02 10

A. Biomedical Radioisotopes 10

1. Rubidium-84 10
2. Gallium-67 10
3. Indium-111 11

B. Miscellaneous 12

1. Bismuth-206 12
2. Cobalt-56 13

FISSION PRODUCTS - 08-01-03 14

A. Krypton-85 Enrichment 14

B. Cesium-137 Process and Product Technology 14

1. Fission-Product Process Study 14
2. Cesium Chloride Slab Sources 16

Page

RADIATION SOURCE DEVELOPMENT - 08-01-04 16

A. Strontium-90 Silicate Sources 16

1. Matrix Studies 16
2. Encapsulation 16
3. Characterization 19

B. Fabrication of Carbon-14 and Technetium-99 Radiographic Sources 19

C. Alpha Source Fabrication Development 19

D. Sources for Medical Applications 20

RADIOISOTOPE SOURCE SAFETY TESTING - 08-01-04 21

A. Special Form Prototype Testing 23

B. Testing Sources with Manufactured Leaks 23

C. Surveillance of Regulations and Cask Evaluations 23

D. Source Leak Test Guide 24

E. Sealed Source Testing 24

F. Unsealed Source Testing 25

G. Examination of Commercial Sources 25

TECHNOLOGY UTILIZATION - 08-01-05 25

A. Information Center 25

B. Isotopes and Radiation Technology 27

C. Publications 27

RADIOISOTOPE APPLICATIONS DEVELOPMENT 28

BASIC TECHNOLOGY DEVELOPMENT - 08-03-01 28

A. Liquid Scintillation Development 28

1. Alpha Radiation Damage in Phosphors 28
2. Cerenkov Counting 29

Page

SYSTEMS ENGINEERING APPLICATIONS - 08-03-02 30

A. Oceanographic Systems Study 30

1. Point Mugu Experiment 32

B. National Security 33

RADIATION PROCESSING APPLICATIONS DEVELOPMENT 33

ORGANIC SYNTHESIS WITH ISOTOPIC SOURCES - 08-04-01 33

A. Woolen Mill Effluent 33

RADIOISOTOPE SALES 34

ISOTOPES SALES AND SHIPMENTS 36

RADIOISOTOPE PRODUCTION 36

CESIUM-137 PROCESSING 36

STRONTIUM-90 PROCESSING 37

MAINTENANCE 38

SOURCE FABRICATION 38

A. Cesium-137 38

B. Strontium-90 39

SHORT-LIVED FISSION PRODUCT OPERATIONS 40

SPECIAL FORM MATERIALS 40

OPERATIONAL SUMMARY 40

PROCESS STATUS 41

MISCELLANEOUS RADIOISOTOPE PROCESSING 41

ADMINISTRATIVE SECTION 41

RADIOISOTOPE PROGRAM (8000) PROGRESS REPORT FOR OCTOBER 1 THROUGH NOVEMBER 30, 1969

A. F. Rupp

RADIOISOTOPE PRODUCTION AND MATERIALS DEVELOPMENT

NEUTRON PRODUCTS - 08-01-01

A. Biomedical Radioisotopes

1. Thulium-171

Thulium-171 is a 1.9-year beta emitter with a maximum beta energy of 0.097 MeV. These characteristics make it useful as a tracer for biological studies. It can best be made, free of the more energetic thulium-170, by irradiating enriched erbium-170.

Initial work will concentrate on separating thulium-171 from previously irradiated erbium-170. Samples of thulium-171 will be supplied to the Lovelace Foundation for use in lung inhalation studies. The ORNL Biology Division also has an inhalation program to study the relation of air pollutants to lung carcinomas, and thulium-171 samples will be provided for use as a tracer radionuclide to study long-term effects. Future work will entail tagging the thulium-171 on a suitable aerosol carrier of known particle size distribution.

2. Phosphorus-33

The purpose of this project is to develop methods of preparing hundred-millicurie quantities of carrier-free phosphorus-33 containing $\leq 5\%$ phosphorus-32. This radioisotope of phosphorus has a lower-energy beta and a longer half-life than phosphorus-32, making it useful for autoradiography work in biochemistry, agriculture, and metallurgy.

Two methods of preparing phosphorus-33 are being evaluated. One method is the irradiation of sulfur-33 enriched to greater than 92% sulfur-33 in a fast neutron flux to prepare the radioisotope by the $^{33}S(n,p)^{33}P$ reaction. The other method of preparation is the fast-neutron irradiation of chlorine-36 to prepare phosphorus-33 by the $^{36}Cl(n,\alpha)^{33}P$ reaction. Highly enriched targets are necessary to reduce the phosphorus-32 content of the phosphorus-33 produced from either of the targets. The yield and purity of phosphorus-33 product prepared by the two methods are discussed below.

a. Phosphorus-33 Preparation from Enriched Sulfur-33

The 96-mg sulfur-33 target ($K_2{}^{33}SO_4$) was enriched to 92.15% sulfur-33, with 7.49% sulfur-32 content. The average yield of phosphorus-33 with $\leq 5\%$ phosphorus-32 content after a 2-1/2-month irradiation at 2.5×10^{14} n/cm^2.sec fast flux was 255 mCi. The target material is expensive and is recovered each time for re-irradiation. Chemical process losses of $K_2{}^{33}SO_4$ are 8% per irradiation.

The phosphorus-32 content of the phosphorus-33 product has been increasing each time the target is irradiated and has not been explained. The K_2SO_4 target material is being assayed at present.

The sulfate target material is more difficult to purify for re-irradiation than KCl targets. Traces of elements that behave chemically like sulfate [such as CrO_4(II-)] tend to get into the target and result in gamma impurities in the phosphorus-33 product after irradiation. These impurities are difficult to remove.

b. Phosphorus-33 Preparation from Enriched Chlorine-36

The purpose of this work is to prepare target material for future phosphorus-33 production by the $^{36}Cl(n,\alpha)^{33}P$ method and to supplement current phosphorus-33 needs now normally supplied by the $^{33}S(n,p)^{33}P$ method.

The chlorine-36 target material was prepared by a 10-year irradiation of natural chlorine in a Hanford reactor. At the time of reactor discharge from Hanford, the isotopic abundance of chlorine was 28% chlorine-35, 47% chlorine-36, and 25% chlorine-37. This material was prepared as KCl and irradiated in a fast neutron flux position of the HFIR (thermal = 8×10^{14} n/cm^2.sec; fast = 2.5×10^{14} n/cm^2.sec).

The irradiation accomplished two purposes: (1) to further enrich the chlorine-36 by the $^{35}Cl(n,\gamma)^{36}Cl$ reaction by the thermal component of the neutron flux and (2) to prepare phosphorus-33 by the $^{36}Cl(n,\alpha)^{33}P$ reaction by the fast neutron flux.

The first HFIR irradiation of the target material (47% chlorine-36 - 28% chlorine-35) at a fast neutron flux of 2.5×10^{14} n/cm^2.sec gave 700 mCi of phosphorus-33 and 265 mCi of phosphorus-32 per gram of KCl target. After a 345-day irradiation at a thermal neutron flux of 8×10^{14} n/cm^2.sec, the chlorine-36 content had increased to approximately 60% chlorine-36 and 15% chlorine-35. Irradiation of this material in a fast neutron flux of 2.5×10^{14} n/cm^2.sec gave 820 mCi of phosphorus-33 and 93 mCi of phosphorus-32 per gram of KCl target. The increase in the yield of phosphorus-33 and decrease in the yield of phosphorus-32 is a result of the enrichment of chlorine-36 and depletion of the chlorine-35 by thermal neutron capture.

An additional 300-day irradiation at a thermal flux of 8×10^{14} n/cm^2.sec is expected to give an isotopic mass composition of 8% chlorine-35, 67% chlorine-36, and 25% chlorine-37. This target will yield 850 mCi of phosphorus-33 with ≤ 40 mCi of phosphorus-32 per gram of KCl target.

3. Potassium-43

The objectives of this project are to prepare potassium-43 by the $^{43}Ca(n,p)^{43}K$ reaction using isotopically enriched ^{43}CaO targets in quantities sufficient for medical and biological experiments, to define a method for separating potassium-43 from the target in a purity suitable for medical use, and to establish cooperative programs with medical institutions interested in evaluating its usefulness.

Potassium-42 has been used, primarily in animals, to locate tumors, to tag red blood cells, and to study blood flow, but has very limited use in humans because of its high-energy (1.52 MeV) gamma-ray emission and its relatively short half-life (12.4 hr). Potassium-43, with a half-life of 22 hr and gamma-ray emissions of 0.373 and 0.617 MeV, is more suitable for metabolic and clinical studies. The lower dose rate would permit multiple doses. It has been suggested as a possible tool for studying blood flow through the heart and for diagnosing myocardial infarctions or immune rejection of transplanted organs.

Several targets of 63.6% isotopically enriched calcium-43 as CaO have been irradiated in a fast-neutron flux of approximately 10^{15} n/cm^2.sec. The average product yield is approximately 10 mCi for a target of 45 mg of calcium-43. Several ion-exchange techniques were evaluated for separation of the carrier-free potassium-43. A major problem has been the presence of sodium-24, leached from the quartz ampule and from sodium contamination in the target, and excessive amounts of inactive salts in the initial preparations. The most successful chemical procedure has been the acid elution of a strong cation exchange resin in the hydrogen form. The calcium-43 target and the scandium radionuclides produced from the decay products of calcium-45 and calcium-47 in the target are strongly bound to the column. Using graduated acid strengths, contamination activities are eluted first, followed by sodium-24 and finally potassium-43 activities. A successful separation of potassium-43 and sodium-24 was achieved. The calcium is finally stripped from the column using 5 M HCl. The calcium target has been successfully recovered for reuse using quantitative oxalate precipitations.

Samples of potassium-43 were supplied for evaluation to Johns Hopkins and also to the University of Mississippi Medical Center. They reported satisfactory results with no observed adverse effects.

A recurrent problem in the recent process runs has been the presence of iridium-192 in the target. This radiocontaminant is produced in large quantities, but fortunately almost all of it washes out of the column in the initial 4 column volumes of eluate. A colloidal form is suspected and consistent with this is the observation of traces of iridium-192 activity in a product sample which has decayed for ten half-lives. The potassium-43 is eluted after 36 column volumes of HCl has been passed through the column. The amount of iridium-192 in the product is estimated to be approximately 0.01 μCi/ml, but the source of the contaminant has not been established. It may have originated from the irradiation in the platinum crucible which was used to calcine the recovered calcium at 1100°C. This possibility is under investigation.

There is considerable uncertainty in the decay scheme of potassium-43. The most recent published work is by Taylor *et al.*[1] and our own spectral studies using a Ge(Li) detector confirms the absence of the 388-keV X ray

---

[1]H. W. Taylor, J. D. King, H. Ing, and R. J. Cox, Note on the Decay of ^{43}K, *Can. J. Phy.* 47: 1539 (1960).

reported in the Table of Isotopes.[2] Our gamma rays are in general agreement with those reported by Taylor et al. and in addition we have observed new weak transitions of 404, 801, 990, 1021, and 1394 keV which fit the level scheme given in the Table of Isotopes.

The relative intensities of the gamma rays and the error limits of the energies will be determined. Our tentative value for the half-life supports the 22.4-hr value quoted in the Table of Isotopes. A tentative calcium-43 level scheme is shown in Fig. 1. The electron spectrum will be studied in a cooperative study with the Chemistry Division.

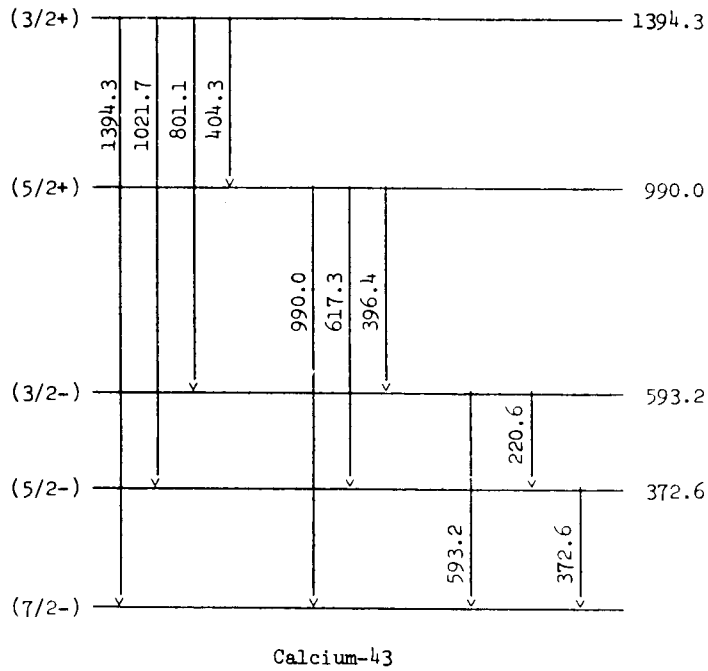


Fig. 1. Calcium-43 Decay Level Scheme.

4. Rubidium-84

Rubidium-84 is used for diagnostic studies of heart muscle. The 0.51-Mev annihilation radiations of rubidium-84 are used in these studies.

The objective of this work was to prepare rubidium-84 by fast neutron irradiation of enriched strontium-84 [$^{84}Sr(n,p)^{84}Rb$ reaction] and determine yield of rubidium-84 and the rubidium-83 and rubidium-86 content. Rubidium-83, which has gamma energies near 0.51 MeV, is an undesirable radionuclide impurity in rubidium-84 products.

A 190-mg SrO target (75% isotopic enrichment of strontium-84) was irradiated for 46 days at a fast neutron flux of 2.6×10^{14} n/cm^2.sec. The anticipated yield and purity is approximately 14 mCi of rubidium-84 with $\leq 3\%$ rubidium-83 and rubidium-86.

[2] C. M. Lederer, J. M. Hollander, and I. Perlman, Table of Isotopes, (6th ed.), John Wiley & Sons, Inc., New York, 1967.

A process has been developed for purifying the carrier-free rubidium-84 and recovery of the enriched strontium-84 target. The process is based on precipitation of strontium by fuming nitric acid, leaving rubidium in solution. The rubidium and traces of strontium are then separated on a Dowex-50 ion-exchange column using HCl as the eluant. This process is expected to give a minimum of 95% recovery of rubidium-84 and 95% recovery of strontium-84.

The target has been discharged and will be processed in November. The product yield and purity will be examined for rubidium-84 and rubidium-86 content and for impurities. If the product is satisfactory it will be shipped to the National Institute of Health for heart studies. This completes this work.

B. Research Radioisotopes

1. Lanthanum-137

Lanthanum-137 has a half-life of 6×10^4 years. It possesses an excited state at 0.010 MeV with an 89-nsec lifetime, which makes this nuclide suitable for the resonant absorption applications of the Mossbauer technique. The objective of this study is to determine the production parameters and separation procedures for making carrier-free lanthanum-137 available in significant quantities.

A cooperative program was initiated with Hebrew University of Jerusalem to produce approximately 150 μg of lanthanum-137. A target containing 61 mg of enriched $^{136}CeO_2$ (22%) was irradiated for one 23-day cycle in a flux of 2×10^{15} n/cm^2.sec. After a period of decay to allow short-lived activities to subside, the target was dissolved in nitric acid and the cerium was oxidized with $NaBrO_3$ and extracted with di(2-ethylhexyl)-phosphoric acid using the procedure described by M. Deschuyter et al.[3] The cerium will be recovered by oxalate precipitation after the cerium-141 has decayed substantially. Analysis by a spark source mass spectrometer indicated a yield of approximately 250 μg of lanthanum-137, but the product was contaminated with europium-156 from impurities in the enriched target material. The product will be purified by ion exchange. A portion will be analyzed by mass spectrometry to deduce cross-section data. In the future, the enriched target will be purified before irradiation, because the Ce(IV) is easily separated from contaminant rare earths by solvent extraction. Future plans also include recovery of the enriched target after the residual activity has decayed sufficiently.

C. Isotopic Power

1. Thulium-170

The objectives of this research are to provide sufficient data on thulium-170, which has been proposed as an isotopic-power source for short-duration missions, to permit reasonable assessment of its potential application and to permit preliminary engineering design of power sources. The economics for reactor production of thulium-170 (128.5 day; $E_{\beta-av}$ = 0.32 MeV) from natural thulium-169 are favorable and the sesquioxide offers a promising

[3] M. Deschuyter, D. L. Massart, A. Speecke, and J. Hoste, Radiochemica Acta 10: 11-14 (1968).

fuel compound, having a practical specific power of approximately 2 W/g and a power density of approximately 16 W/cm^3. The study will include measurements of thermal conductivity and thermal diffusivity, leach rates for safety analysis, high-temperature compatibility with suitable containment metals, vapor pressure, and fuel form densification.

The thermal conductivity measurements, which include specimens of Tm_2O_3, Yb_2O_3 (ytterbium is the decay product of thulium-170), and equimolar mixtures of the two, are for the purpose of demonstrating that the thermal conductivity of thulia-ytterbia mixtures is a linear function of the concentrations. A Yb_2O_3 pellet has been hot-pressed and the thermal conductivity measurement will be made during the next month.

Specimens of hot-pressed thulia have been sent to Savannah River for encapsulation and irradiation in the high flux experiment (greater than 10^{15} n/cm^2.sec). The irradiation, which should begin in December, will demonstrate the capability of producing a fuel having a specific power of approximately 3 W/g and the specimens will then be used for compatibility exposures with refractory alloys at temperatures up to 2000°C.

Sol-gel thulia microspheres were fabricated in sizes ranging from 70-600 μm and were examined in detail. The microspheres had been sintered at 1475°C. The porous structure of the microspheres is shown in the micrographs in Fig. 2. Note that the epoxy resin penetrates the microsphere completely in the mounted specimen. This program is about 60% complete.

D. Miscellaneous

1. Terbium-157

The purpose of this work is to prepare sufficient terbium-157 (150 years) for evaluation of the Mossbauer effect of this radioisotope. This material is being prepared for the Physics Division at ORNL.

Terbium-157 was prepared by the following nuclear path:

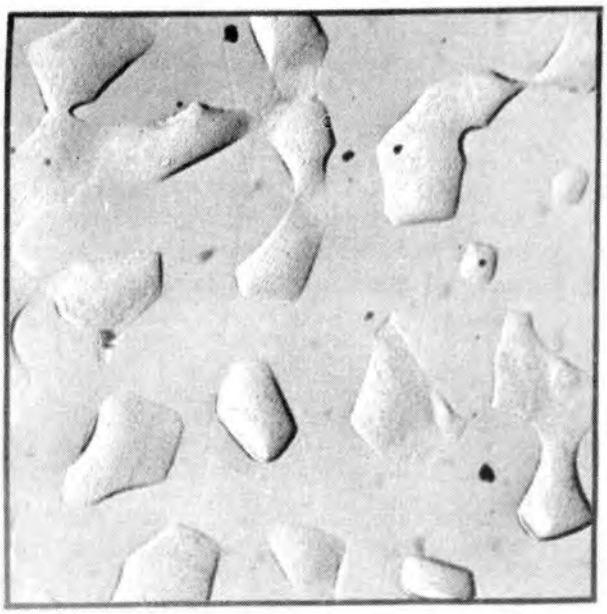
$$^{156}Dy(n,\gamma)^{157}Dy \xrightarrow{8.5\ h} {}^{157}Tb$$

The 100-mg target of enriched dysprosium-156 (12% isotopic enrichment) was irradiated 20 weeks in a thermalized neutron flux of 2.5 x 10^{15} n/cm^2.sec. The terbium activities were separated from the Dy_2O_3 target material and other rare-earth activities by high-pressure ion exchange.

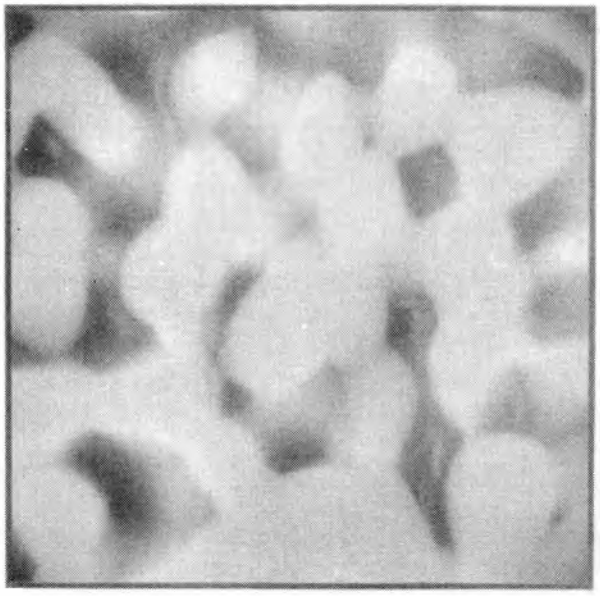
Isotopic mass analysis of the terbium and yield of terbium-160 (73 days) were used to calculate the yield (by weight) and composition:

Isotope	At. %	Terbium Isotope Yield, mg
Terbium-157	2.81	0.064
Terbium-158	1.04	0.024
Terbium-159	93.17	2.153
Terbium-160	2.98	0.069
		2.310

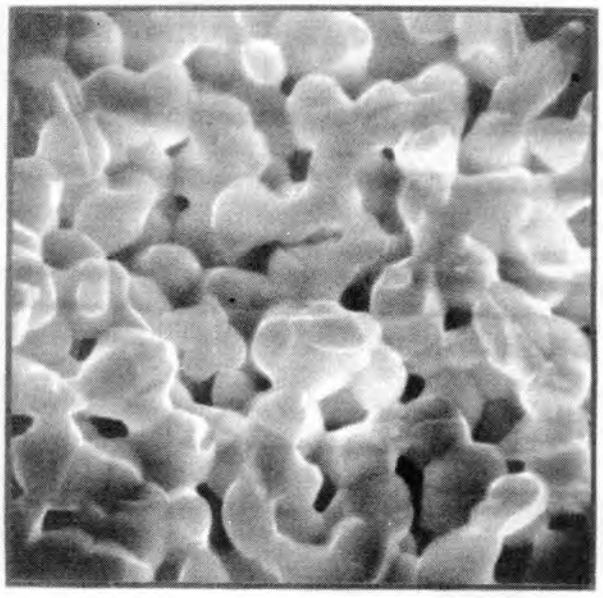
OPTICAL MICROSCOPY 500X
(A Microsphere That was Mounted in Epoxy Resin and Metallographically Prepared In a Conventional Manner)



ELECTRON MICROSCOPY 16,000X
(Indirect Carbon Replica)



SCANNING ELECTRON MICROSCOPY 10,000X
(Direct Image of Polished Section)



SCANNING ELECTRON MICROSCOPY 10,000X
(Direct Image of Fractured Surface of a Microsphere)

Fig. 2. Porous Structure of Tm_2O_3 Microspheres.

Yields of terbium-157 and terbium-158 were a factor of 4 lower than calculated, indicating possible extensive neutron capture by these isotopes at the high neutron flux. The yield of terbium (2.31 mg) is near that anticipated.

The Physics Division will evaluate this material, and if the terbium-157 content is high enough a Mossbauer source will be prepared. Arrangements have been made to have the terbium-157 separated and implanted in a molybdenum matrix using a calutron adapted for separations of radioactive materials.

E. Supporting Technology

1. Radioisotope Characterization

Properties of radionuclides such as half-lives, gamma energies and percentages, and beta and alpha heat outputs must be known to permit handling, analysis, and application. Selection of values frequently has to be done by personnel directly involved in production or analysis, because compilations and adoption of values by information groups necessarily lag several years behind availability of that information. In other cases, information of sufficient quality is simply not available and must be determined. The Isotope User's Guide (report ORNL-IIC-19) contains a large table of properties of about 240 common radionuclides, that can be readily interpreted by nonspecialists. It appeared in time for showing at the Gatlinburg Analytical Conference, and response was enthusiastic, particularly for the tabulated data. Many requests for copies have been received, both domestic and foreign. Recent work to keep data up-to-date included a critical review of half-life determinations for thallium-204, with selection of a value of 3.78 ± 0.02 years, scanning of two large tables of gamma energies in recent issues of a journal, and recommendation of gamma-intensity values for cesium-134 (see section below).

An important aspect of the whole program of quality control and characterization is furnishing of critically evaluated information to ORNL workers and other personnel. This is done by preparation of articles and compendia and responding to telephone or letter inquiries. Much of this work is done in collaboration with the Isotopes Information Center.

2. Radioisotope Special Analysis and Quality Control

The analysis of routine and special radioisotope products is broken down into assay and quality control. The former refers to content of the desired radionuclide and involves standards and calibration techniques, while the latter includes purity and chemical characteristics. It is apparent that ORNL products must be assayed as accurately as is feasible, because some users accept the stated concentration of a shipment without verifying it, and others attempt to confirm the measured concentration and protest if they cannot. Further, the products must obviously be reasonably free of impurities, equaling or exceeding catalog specifications of purity. This work must in general be of an *ad hoc* nature, responding to locally detected deviations of assay or purity, or to

inquiries or complaints of users. Methods and instrumental techniques are reviewed on occasion, new ones are recommended or developed, and special analyses are sometimes done.

An essential part of the quality control program with respect to assay is the supplying of standards. Some are prepared locally, and others are purchased from reputable suppliers. However, some items marketed as standards are of poor quality, with disintegration rates quoted erroneously or containing impurities. Growing awareness of this fact prompted the initiation of a survey by the National Academy of Sciences, through its Subcommittee on Use of Radioactivity Standards. The first step of the survey involved the sending of a questionnaire to users of radioisotopes who presumably needed standards and analysis of the questionnaire responses. A panel of specialists in various fields is now being selected to review the first step and to make suggestions about the next phase, which is the elucidation of details by personal interviews with people who are engaged in large standardization programs or have instructive problems. Finally a report will be prepared, and it will be issued after review and appropriate revision by the panel. The analysis of questionnaire responses, the interviews, and the preparation of the report are to be done by S. A. Reynolds of the Isotopes Development Center. A newly formed committee of the American National Standards Institute (formerly USASI) has indicated an interest in the program and information has been forwarded.

A half-day session on radioactivity standards was held at the recent Gatlinburg Conference on Analytical Chemistry in Nuclear Technology, followed by a half-day meeting of the NAS-NRC Subcommittee on Use of Radioactivity Standards. Both were well attended, and discussion was very active. The standards-evaluation program was described in a paper in the conference session, and the Subcommittee reviewed it and made suggestions for subsequent phases. Returns of questionnaires now number about 400, with 60% expressing satisfaction with available standards. Fruitful discussions were held with individual members of the Subcommittee, H. Houtermans of IAEA, and others engaged in preparations or use of standards.

The Subcommitte sponsored a round-robin involving preparations of often-used radioisotopes. The very satisfactory results by our routine laboratory were 1.7% higher than the average on cobalt-57 and 1.3% lower than the average on iodine-125. A standard of cesium-134 from a commercial organization was recently assayed by means of a gamma ionization chamber, and the measured values averaged 2.4% above the certified value, which was said to have an "effective standard deviation" of 1.9%.

CYCLOTRON PRODUCTS - 08-01-02

A. Biomedical Radioisotopes

1. Rubidium-83

The chief objective of this work is to supply rubidium-83 (83 days) for medical and research applications under cooperative programs and to develop the technology for preparing a high-purity product acceptable for human use. Rubidium-83 is prepared by cyclotron bombardment of strontium-84 by the following reactions:

$$^{84}Sr(p,2n)^{83}Y \xrightarrow[EC]{\beta^+} {}^{83}Sr \xrightarrow[EC]{\beta^+} {}^{83}Rb$$

$$^{84}Sr(p,pn)^{83}Sr \xrightarrow[EC]{\beta^+} {}^{83}Rb$$

$$^{84}Sr(p,2p)^{83}Rb$$

Two cooperative programs have been established with the University of Utah College of Medicine and National Center for Radiological Health and excess development material is distributed to these participants for evaluation. No progress to report this month.

2. Gallium-67

The objectives of this program are twofold in nature: to determine the optimum target configuration for producing gallium-67 (78 hr) in acceptable purity and to provide gallium-67 for medical applications research and development. Interest in this isotope has been spurred by evidence obtained by the Medical Division of Oak Ridge Associated Universities (ORAU), in both animals and humans, of a high uptake of carrier-free gallium-67 by lymphoid tumors. This interesting discovery was made during the evaluation of gallium-67, produced in the ORNL 86-Inch Cyclotron by the $^{68}Zn(p,2n)^{67}Ga$ reaction, as a bone-scanning agent for a patient with Hodgkin's disease.

Four gallium-67 preparations were made and shipped to ORAU; three smaller orders for gallium-67 were filled for clinicians interested in evaluating the potential of this radionuclide for their applications.

Work has continued on cyclotron target development. The target previously used was 1/2-in.-dia zinc tubing of 20-mil wall thickness, but this tubing would often creep and fail at a beam current of 300 μA. Sustained irradiation was limited to 200- to 250-μA currents. A new 3/8-in.-dia, 20-mil wall thickness zinc tubing was tested for tolerance to higher beam currents. This new tubing was irradiated at a beam current of 325 μA for 1.3 hr without failure and is routinely irradiated at 315 μA for up to 3 hr without difficulty.

The feasibility of employing zinc-65 as an internal quality control for zinc content in the gallium-zinc separation process was evaluated. From a comparison of gamma-ray spectra obtained with a Ge(Li) detector and a

4096 channel analyzer of unprocessed and of purified samples, it was concluded that the zinc-65 specific activity was too low for detection of multimicrogram amounts of zinc within reasonable counting times. A chemical spot test using a 1% aqueous solution of $K_4[Fe(CN)_6]$ was also investigated and found to be sensitive to 20 μg of zinc per milliter.

Gallium-67 will be supplied on a biweekly basis to ORAU per the cooperative agreement and will be sold to other researchers.

3. Indium-111

The purposes of this project are twofold in nature: to determine the optimal indium-111 production parameters and to establish cooperative programs with qualified researchers for its evaluation as a diagnostic radionuclide in medicine (e.g., such applications would include spinal-cerebral cisternography, aerosol lung studies, delayed brain scanning, visualization of the lymphatic system, metabolic studies of indium-labeled macroaggregates and colloids, and possible applications of tumor localizations.)

Indium-111 has both a favorable decay scheme [gamma emissions of 173 keV (89%) and 247 keV (94%)] ideally suited for external detection and an optimal half-life (2.81 days) for compound labeling and distribution studies which must be carried out over 24-hr periods or longer. The use of indium in such lengthy studies is currently impossible with the indium isotope now in general use, indium-113m (1.73 hr).

The objectives of the indium-111 product development work being carried out are to elucidate, define, and optimize pertinent production parameters (e.g., cyclotron target material, cyclotron beam energy and current conditions, adverse side reactions leading to unwanted isotopic impurities, target processing chemistry and product purification procedures, and target recovery, if necessary) and to make sufficient material available to cooperative participants for applications research. As an example of the varied production possibilities, indium-111 may be produced in the ORNL 86-Inch Cyclotron by three entirely different nuclear reactions [$^{111}Cd(p,n)^{111}In$, $^{112}Cd(p,2n)^{111}In$, and $^{112}Sn(p,2n)^{111}Sb \xrightarrow{\beta^+,EC} {}^{111}Sn \xrightarrow{\beta^+,EC} {}^{111}In$] all of which require different target and irradiation conditions, and two of them require completely different processing for chemical separations.

Work was continued to determine thick-target production rates and actual production yields for the $^{111}Cd(p,n)^{111}In$ reaction. Preliminary production rate yields of 420 mCi/mA-hr and 80 mCi/hr, respectively, were determined using a 460-mg target of 96.5% enriched ^{111}CdO. These values are in good agreement with earlier values obtained for a similar irradiation configuration.

Due to unacceptably low chemical yields (approximately 30%) obtained recently for the separation of indium-111 from cadmium target material by extraction into acetylacetone, development work on a HBr-isopropyl ether extraction procedure has been initiated. The safety hazards of using isopropyl ether in a hot cell were evaluated and were judged acceptably

low for the small amounts required (approximately 40 ml total). A ^{113}Sn-^{113m}In isotope generator was prepared to supply tracer amounts of carrier-free indium-113m for preliminary hood-level tracer experiments. These experiments were performed to determine feasibility of this separation system for carrier-free amounts of indium (all literature references pertaining to this system describe separations involving macro amounts of indium), and to gain safe handling experience for the ether system before attempting the in-cell operation.

The results of the preliminary experiments were encouraging so one full-scale production run was scheduled, the target irradiated and processed, and two shipments of approximately 50 mCi each were shipped to two of our three cooperative program participants. A chemical yield of approximately 98% was obtained from the processing. Preliminary chemical spot tests indicated less than 20 μg of cadmium per ml in the final product.

The use of this product in humans (a responsibility of the medical researchers) makes quality control a most important item. The future work on this project will be devoted to both chemical and radionuclidic quality control. Extensive chemical analysis will be performed to determine the quality of the product as obtained from the current separation procedure. Preliminary consideration will also be given different types of cyclotron targets so that the (p,2n) reactions mentioned previously may be investigated under the most favorable conditions.

B. Miscellaneous

As part of a continuing effort to assist cooperative participants to fill specialized requirements for specific radionuclides not usually available, miscellaneous, short-term tasks are occasionally undertaken. The demand or potential usefulness of these radionuclides is not considered adequate to justify extensive effort or a continuing development program, but the services performed are valuable to meet program objectives.

1. Bismuth-206

A radiogenic lead-206 cyclotron target was bombarded with 22-MeV protons in the ORNL 86-Inch Cyclotron and then chemically processed for use by Vanderbilt University under our cooperative program. The desired nuclear reaction was $^{206}Pb(p,n)^{206}Bi$. An attempt was made to maximize this reaction and minimize the unwanted side reaction $^{206}Pb(p,2n)^{205}Bi$ by using a lead-206 absorber to degrade the proton beam energy to approximately 15 MeV. When the sample was opened for processing, it was discovered that the lead-206 absorber and target had fused together. As a result, both the absorber and sample were dissolved and processed together.

Preliminary thick-target production rates and actual cyclotron yields for both the (p,n) and (p,2n) reactions are shown in Table 1. It should be noted that the yields listed in Table 1 do not represent the maximum possible yields. The target was intentionally bombarded at a low beam current in an attempt to prevent fusing of the target and absorber.

Table 1. Preliminary ^{206}Pb Thick-Target Production Data[a]

Nuclear Reaction	Production Rate, mCi/mA-hr	Actual Cyclotron Yields, mCi/hr
$^{206}Pb(p,2n)^{205}Bi$	210	11
$^{206}Pb(p,n)^{206}Bi$	230	12

[a]All data normalized to end of bombardment.

2. Cobalt-56

In radiographic work where high-energy photons are needed for penetration, there is a limited choice of radioisotopes which decay with gamma energy greater than 2 MeV and have a sufficient half-life. Cobalt-56, with a half-life of 77.3 days and gamma photons of up to 3.26 MeV, has proved to be useful.

Production of cobalt-56 by the $^{56}Fe(p,n)^{56}Co$ reaction has a reported average yield of 70 mCi/mA-hr for an internal target in the ORNL 86-Inch Cyclotron.[4]

The iron target is a thin foil soldered to a copper flat plate. The copper plate is heated until the solder melts and the iron foil can be removed. Excess molten solder is scraped from the iron target to prevent undue difficulties during the chemical purification process.

The front surface of the iron target which has received the proton bombardment is placed downward in a crystallizing dish and sufficient 6 N HCl solution is added to dissolve only from the front surface. After several dissolvings the residual radiation is measured as an indicator of the remaining activity in the target. Greater than 95% of the radioactivity is removed by this method.

The inactive iron is separated from cobalt-56 by extracting with 4-methyl-2-pentanone (MIBK). The cobalt-56 remains in the 6 N HCl solution. The separation factor of iron from cobalt is 10^6 with this system.[5] Because of the large quantity of iron in the 6 N HCl solution, it is necessary to extract approximately 10 times with MIBK. Radiochemical analyses show that the production yield is 86.0 mCi/mA-hr and that the loss during the chemical processing is less than 8%. Four gamma sources containing 20 mCi each of cobalt-56 were fabricated for a customer.

[4]G. I. Gleason, I. J. Gruverman, and J. L. Need, Int. J. Appl. Rad. Isotopes 13: 223-8 (1962).
[5]J. J. Pinajian, The Methyl Isobutyl Ketone (4-Methyl-2-Pentanone) Extraction of Trivalent Iron from Hydrochloric Acid Solutions and the Effect of an Inert Diluent on the Distribution Coefficient, ORNL-TM-1481 (April 1966).

FISSION PRODUCTS - 08-01-03

A. Krypton-85 Enrichment

Operation of all six columns has continued routinely during the month. The following product fractions were removed:

510 Ci of 29.5% krypton-85
385 Ci of 24.0% krypton-85
194 Ci of 10.8% krypton-85

The 29.5% and 24.0% material was taken from one column which has been operating on second-pass feed material. The 10.8% material came from a single-pass operation starting with 3 to 5% krypton-85.

One column which has been operating on stable krypton for 65 days to provide enriched target material for cyclotron operations was emptied and the isotopic analysis of the product fractions is given in Table 2. The column has been recharged with radioactive krypton.

Table 2. Isotopic Analysis of Krypton-85[a] Enrichment Product Fractions

Section	Flask Vol, ml	Pressure, in. of Hg	Gas Vol, ml	Mass Concentration, vol %					
				^{78}Kr	^{80}Kr	^{82}Kr	^{83}Kr	^{84}Kr	^{86}Kr
Light End									
1st cut	500	14	266	3.3	19.0	37.9	8.6	30.7	0.5
2nd cut	500	14	266	1.8	16.4	59.8	18.8	3.2	-
3rd cut	500	10.5	325	0.9	11.8	59.5	23.1	4.7	-
4th cut	500	12	300	0.9	11.9	59.5	22.9	4.8	-
5th cut	500	10	334	0.1	3.4	51.0	33.5	11.8	0.2
6th cut	500	5	416	-	1.0	34.8	39.9	24.2	0.1
Heavy End	250	10	167	-	-	-	-	8.8	91.2
Middle									
1st cut	980	3 psig	1180	-	-	0.2	4.1	95.3	0.4
2nd cut	1030	7	790	-	-	0.1	1.5	96.5	1.9
3rd cut	981	1 psig	1050	-	-	0.1	1.5	95.7	2.7
4th cut	600	10.5 psig	1030	-	-	0.3	5.0	92.9	1.8
Total gas volume			6124						

[a]Starting feed - 0.35 vol % ^{78}Kr, 2.27 vol % ^{80}Kr, 11.6 vol % ^{82}Kr, 11.5 vol % ^{83}Kr, 57.0 vol % ^{84}Kr, and 17.3 vol % ^{86}Kr.

Of current interest is the testing of a self-illuminating traffic sign by the Arizona Highway Department in Phoenix, Arizona. The self-luminous modules used in the sign were developed by American Atomics Corporation of Tucson, Arizona, and contain enriched krypton-85 furnished by ORNL.

B. Cesium-137 Process and Product Technology

1. Fission-Product Process Study

With the prediction of rapidly increasing amounts of spent nuclear fuel to be reprocessed in the near future, the problem of storing radioactive

wastes should become more and more acute accordingly. Of the various fission products contained in the wastes, the long-lived cesium-137 and strontium-90 have to be removed to prevent generation of an excessive amount of heat in the stored wastes.

Cesium-137, in particular, is an attractive source of radioisotope radiation in view of its relatively high power output (4.8×10^{-3} W/Ci) and its long half-life (30 years). However, the selling price (and therefore the production cost) of cesium-137 must come down considerably in order for cesium-137 to compete with other radiation sources. The purpose of this project is the study and development of economical processes for recovery, purification, and conversion of cesium-137 obtained from reactor fuel reprocessing operations.

The project objectives are a study and economic evaluation of various potential processes for recovery, purification, and conversion of cesium-137; development of economical recovery processes, for possible application to commercial-scale production; and preparation of project reports.

The current objective is the continuation of the work to develop a generalized semiempirical method for evaluating various conceptual process designs.

Preparation of the conceptual flow diagrams with processing conditions has been continued for the following processes for purifying and converting cesium-137 loaded on the inorganic ion exchanger:

1. Use of HCl as the eluant, followed by concentration of the eluate in an evaporating-crystallizer to purify and recover CsCl.

2. Use of HNO_3 as the eluant; evaporation and crystallization of the eluate to recover $CsNO_3$, followed by reaction with either HCl gas or aqueous solution to convert to CsCl.

3. Use of $(NH_4)_2CO_3$-NH_4OH as the eluant; partial evaporation of the eluate to concentrate Cs_2CO_3 and to drive off ammonia. Reaction of the concentrate with HCl gas converts Cs_2CO_3 to CsCl, which is purified and recovered by evaporation and calcination to dryness.

These conceptual flow diagrams are to be used in determining the number of functional units (N) in the equation presented in <u>IDC Newsletter for July 1969</u> (ORNL CF-69-8-13). The processing conditions and materials of construction would permit estimation of the process complexity factor (f_c).

Preparation of the conceptual flow diagrams will be continued, and the estimation of the number of functional units and the process complexity factor will be made. The compilation of cost data will continue.

2. Cesium Chloride Slab Sources

The study into the causes of the swelling of some CsCl (BNL type) slab sources was continued. To examine gases that form within the sources and determine if a difference could be found between pelletized and powder CsCl, five cans of CsCl powder each containing 12,000 Ci were punctured and the contained gases sampled. The measured gas pressures varied from 13.3 to 19.1 psia. The gas space within the powder cans was measured to be approximately 150 cm^3. The gases from the powder cans were passed into evacuated sample flasks and subjected to mass spectrographic and gas chromatographic analysis.

Mass spectrographic analysis showed the main gas to be nitrogen (assuming no CO) that varied from 63.6 to 87.4%, which would be from air sealed within the powder cans. The main contaminating gas was CO_2, which varied from 6.2 to 29.5%. This amount of CO_2 would be equivalent to a content of 39-176 ppm of CO_2 in the CsCl powder, which is below the amounts normally found as residual in routine powder analysis. The CO_2 content could be from carbonates left over from the oxalate precipitation step in the powder process. Other gases measured were O_2, which varied from 0.1 to 5.6%, and H_2, which varied from <0.01 to 3.9%.

To further study the gas buildup in the CsCl sources, four BNL-type slab-source liners and capsules have been designed and fabricated with 15-psig gages connected by 1/8-in.-dia stainless steel tubing. The liners were loaded with CsCl pellets and then inserted into the capsules which were then welded closed. These are to be watched and the pressures recorded over a period of time. Great care was taken to see that the gages, liners, and capsules were absolutely clean and that they remained clean during assembly and welding. This work was carried out in a clean room and glove box to lessen the chances of organic contamination which could cause gas generation and buildup.

RADIATION SOURCE DEVELOPMENT - 08-01-04

A. Strontium-90 Silicate Sources

1. Matrix Studies

Nothing to report.

2. Encapsulation

The recently developed process in which strontium silicate minibeads are imbedded in an aluminum matrix was used to fabricate 155 6-in. sources and nineteen 3-in. sources using thin-walled (0.006 in.) rectangular (0.375 in. by 0.085 in. inside dimensions) capsules. Approximately 5000 Ci will be loaded in twenty 6-in. tubes and two 3-in. tubes. These sources and those previously delivered will be used in beta source work at the Brookhaven National Laboratory.

Eighty sources, using the same procedure, were fabricated using (0.010-in. wall) capsules (0.375 in. by 0.085 in.) approximately 4 in. long. Sixteen of these sources (approximately 2100 Ci) were loaded into a circular-type array and installed in a beta irradiator (Fig. 3) capable of withstanding 2000 psi for development work at ORNL.

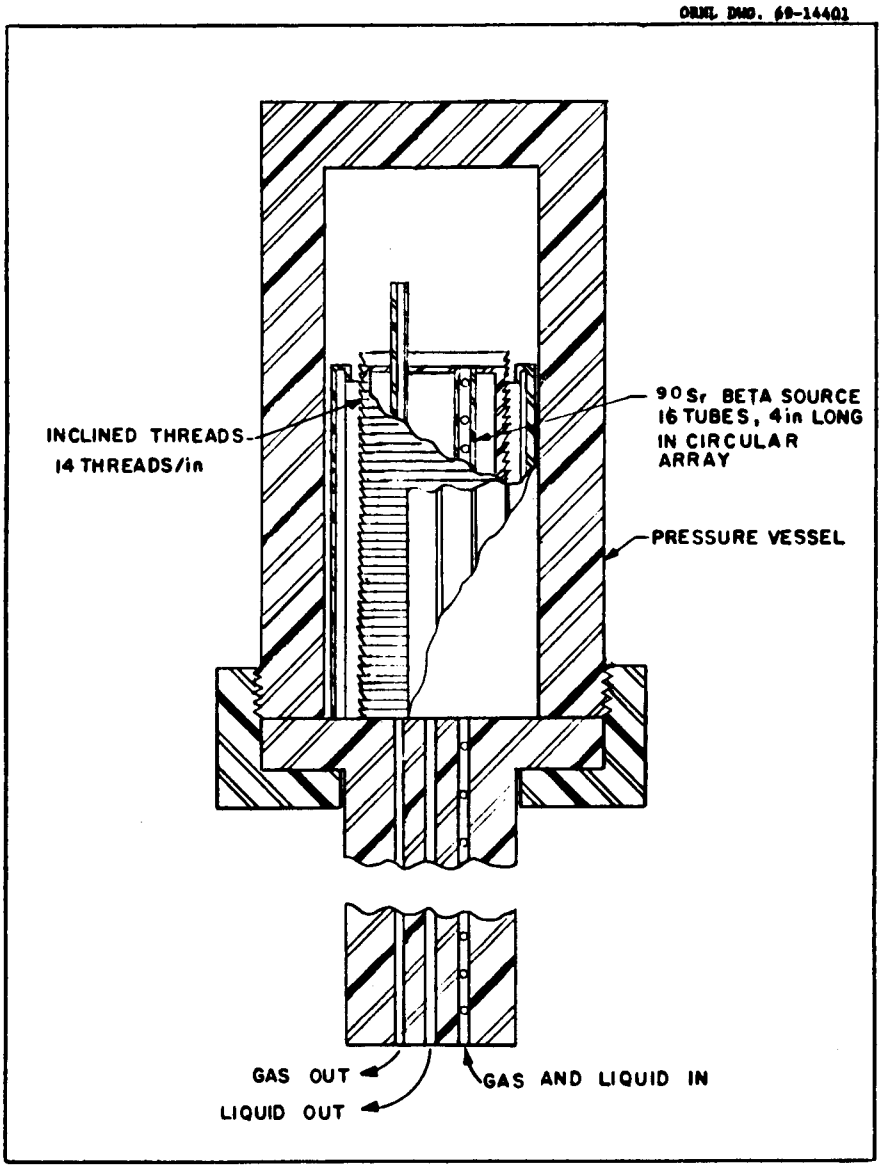


Fig. 3. High-Pressure Beta Process Irradiator.

This irradiator consists of a commercially available high-pressure reactor vessel (Paar bomb) containing the beta source, a lead shield which was fabricated from a reconditioned carrier, and a high-pressure piping system which serves as a means for pumping liquid into and out of the reaction chamber.

The reactor vessel is constructed of stainless steel with a cavity 2-1/2-in. dia by 6-7/32 in. long. It is equipped with a stainless steel top with extended tubing connections capable of penetrating the bottom of the shield to prevent radiation escape at this point and also to provide means for making tubing connections outside the shield.

The source container consists of a thin-walled cylinder with an outside diameter of 2.475 in., an inside diameter of 2.380 in., and an overall length of 4.150 in. The bottom was machined with a 0.050-in.-thick lateral ring and an inside diameter of 2.125 in. A longitudinal ring with a 2.050-in. inside diameter was then machined 0.200 in. long. The source tubes were placed with ends resting on the lateral ring and held in place by the longitudinal ring. A top, machined in the same manner, was placed in position and tack welded to form a compact integral unit holding sixteen 4-in. sources (a total of 2100 Ci) in a 2.125-in.-dia circle.

The liquid sample to be irradiated was recirculated by a gas lifting device consisting of 3/8-in. autoclave tubing formed in the shape of a U. The admission of oxygen on one side of the U-tube carried the liquid sample to the reactor vessel above where the oxygen was disengaged and the liquid flowed down a threaded cylinder 2 in. in diameter and 3-1/2 in. long inside the source array. A sight glass and manual regulating valves completed the piping system. Dose rate measurements with ceric sulfate solution showed that 4.55×10^7 rad/hr were absorbed in a 25-ml sample.

The shield consisted of a mild steel shell (1/4 in. thick) 18-in. outside diameter and 24-in. high with a cavity 14 in. long and 10 in. in diameter.

The remaining 68 sources (approximately 10,000 Ci) were loaded into a plaque and installed in a low-pressure beta irradiator. The low-pressure (atmospheric) beta irradiator consists of a source plaque 4 in. wide and 32 in. long, an inner stainless steel container, and a stainless steel-lined lead outer radiation shield. The source plaque is centrally positioned in the inner container (inside dimensions 2-9/16 in. by 5-1/2 in. by 38 in. long). Liquid sprayed from two manifolds located at the top is exposed to the radiation source as it flows down the side walls. The radiation shield consists of 4 in. of lead with a stainless steel lining and cladding. Provisions were made for heat transfer medium circulation around the inner container.

Final decontamination of the source tubes was accomplished by electrostripping the source surface in a conventional electropolishing bath. Electrostripping or polishing consists of a carefully controlled metal removal process in which the part to be polished is made the positive electrode in an electrolytic bath. Cleaning occurs because a small quantity of the metal is removed from the surface (less than 0.001 in.). The procedure used to clean the sources consisted of two electropolishing steps followed by fresh water rinses. In the first step the source was subjected to 10 Amp for 10 sec and in the second step to 10 Amp for 2 sec, which produced essentially contamination-free sources.

The conventional leak test procedure in which the source is placed in liquid nitrogen and then in hot water for bubble observation was used, but was considered to be unreliable because of the absence of voids in the source capsule. For this reason it was necessary to add a more conclusive test. The source was immersed in water for 24 hr with subsequent contamination check, heated to 400 °C for 4 hr, and immersed in water for 24 hr again with subsequent check for activity in the H_2O. This test proved to be very reliable, since the activity brought out of the leaking capsules by the heating step is very easily detected in the water used in the second immersion.

Two hundred grams of strontium silicate minibeads were produced with a specific activity of 0.5 Ci/g. The beads were classified to give a particle range of +40 -100 mesh and then washed to remove the fines. Attrition tests in connection with pebble bed irradiator work are currently being conducted with these beads by the Western New York Nuclear Research Center.

3. Characterization

Procedures for an experimental program have been written for the characterization studies of strontium silicate. These include the solubility of the strontium silicate minibeads and matrix in various acids and bases, the compatibility of molten aluminum and minibeads, thermal conductivity of matrix material and strontium silicate, and minibead attrition in gases and liquids in motion.

Future development plans include the substitution of graphitic carbon for aluminum as a matrix in the source capsule. The advantage of increased resistance to chemical attack and high-temperature corrosion of the capsule interior and reduced self-absorption of the beta particles make this possibility quite attractive. Two promising methods are coating the minibeads with pyrolytic carbon and producing the desired source form in a die using a resilient graphite which has recently become commercially available.

B. Fabrication of Carbon-14 and Technetium-99 Radiographic Sources

Nothing to report.

C. Alpha Source Fabrication Development

The purpose of this work is to develop a technique for fabricating alpha sources with the following characteristics:

1. Output in the millicurie range.

2. Encapsulated so that the source can be used safely in a laboratory environment.

3. Minimum energy degradation of the alpha particles.

4. Retain integrity in a vacuum.

To accomplish these objectives, a source design was developed which required a source capsule with a 0.0001-in. window (approximately 1-MeV degradation), a technique to deposit the source material onto a suitable backing, and a window support strong enough to prevent rupture of the window under vacuum, but with enough open space to allow alpha particle transmission. The electrodeposition of curium-244 was investigated as a means of depositing the alpha emitter. A uniform thickness is desired to provide a monoenergetic alpha output. It was determined that the deposition technique used by Ko[6,7] was useful when the total quantity of curium-244 contained in the electrolyte was less than 60 mCi. A solution that was 0.20 M HCOOH and 0.15 M $HCOONH_4$ was used as the electrolyte. At a current density of 450 mA/cm^2 a quantitative deposition of the metal hydroxide is possible. Attempts to deposit greater than 60 mCi of curium-244 resulted in a decreasing yield. When 150 mCi was used, only 8.8% of the material was deposited. A system using dimethylsulfoxide (DMSO) was investigated as a method of preparing sources containing greater than 60 mCi. The initial experiments resulted in erratic yields. It was determined that the presence of nitrates was causing this problem. The curium-244 was converted to a chloride and deposits containing greater than 80% of the starting material were obtained. The depositions were successful using nickel, beryllium, carbon, and copper as the backing materials.

Because of the interaction between alpha particles and air, it is necessary to use a metal window that is not subject to rapid corrosion. Several metals were considered and stainless steel or nickel appeared to be suitable for this application. Thin nickel foils prepared by electroplating nickel on a copper backing which is then removed is readily available; however, it was necessary to roll the required stainless steel foils. The windows were covered with stainless steel screens that provided physical strength. Nickel, stainless steel, copper, and organic films were used as source windows. The copper and organic films (approximately 25-mCi sources) deteriorated within two weeks. The sources fabricated with stainless steel foils leaked. Nickel windows on sources containing less than 30 mCi of curium-244 remained intact for several months, but nickel windows on sources containing greater than 30 mCi slowly deteriorated and after a few weeks leakage occurred.

Most of these experiments were pilot experiments that point out several areas requiring additional work. The most important task to be accomplished is the development of a suitable window.

D. Sources for Medical Applications

Two blood irradiators, each containing from 75 to 100 Ci of strontium-90, have been fabricated to be used in extracorporeally irradiating the blood stream so that radiation effects can be evaluated in medical research of blood disorders. These units will be sent to Vanderbilt University Hospital, Nashville, Tennessee, and to East Tennessee Baptist Hospital, Knoxville, Tennessee.

[6]Roy Ko, Nucleonics 14(7): 74 (1956).
[7]Roy Ko, Nucleonics 15(1): 72-7 (1957).

Strontium-90-yttrium-90 radioactivity is fixed as a matrix of strontium silicate spheres and aluminum metal and encapsulated inside an annular stainless steel source capsule. The solution flows through plastic tubing of various diameters which has been threaded through the outer lead shielding, through the highly radioactive annular source, and back to the outside of the irradiator. When the unit is used for animal or human blood irradiation, the tubing is a shunt between an artery and a vein, and the blood is pumped by the heart action through the irradiator.

These irradiators were designed to give an absorbed dose of slightly greater than 500 rads when flowing at a rate of 100 ml/min. The dose rate and hence the absorbed dose is dependent upon the velocity of the solution through the irradiator, the inside diameter of the tubing, and the thickness of the tubing wall. The amount of beta energy which is absorbed by solutions flowing through the irradiator is determined by flowing Fricke solution (ferrous ammonium sulphate in 0.4 M H_2SO_4) and measuring the oxidation of Fe(II) and Fe(III). From the measured absorbed dose rate information, a family of curves was drawn (Fig. 4) which relates radiation absorbed dose (rad) to flow rate and tubing size for a particular blood irradiator.

From the radiation absorbed dose rate studies, the efficiency for transfer of energy from the source capsule to the inside volume of the irradiator was calculated to be from 6 to 8%.

Beta radiation is not homogeneously absorbed; the first layers receive the greatest amount of absorbed energy. In these irradiators, there may be a 4:1 ratio between the amount of energy absorbed in the outside surface and the center section of the flowing solution.

Further studies will be made to determine variation of energy deposition with depth in the irradiated material. This depth effect is to some extent offset by mixing of the solution within the tubing during flow through the irradiator. Dose measurements on such solutions tend to give average energy absorbed, which may suffice for the blood irradiation studies.

RADIOISOTOPE SOURCE SAFETY TESTING - 08-01-04

The safety testing program is concerned with determining how reliably radioactive sources will contain the radioactive material under the conditions of normal use as well as in the event of an accident. In addition to the sources, shipping casks used to transport sources and radioactive material are tested under normal conditions of transport as well as hypothetical accident conditions. The investigations produce data which enable source manufacturers and regulatory agencies to evaluate existing and future source and shipping container designs from the standpoint of safety to the users and the general public. Simple tests are developed that can be used by manufacturers for quality control purposes.

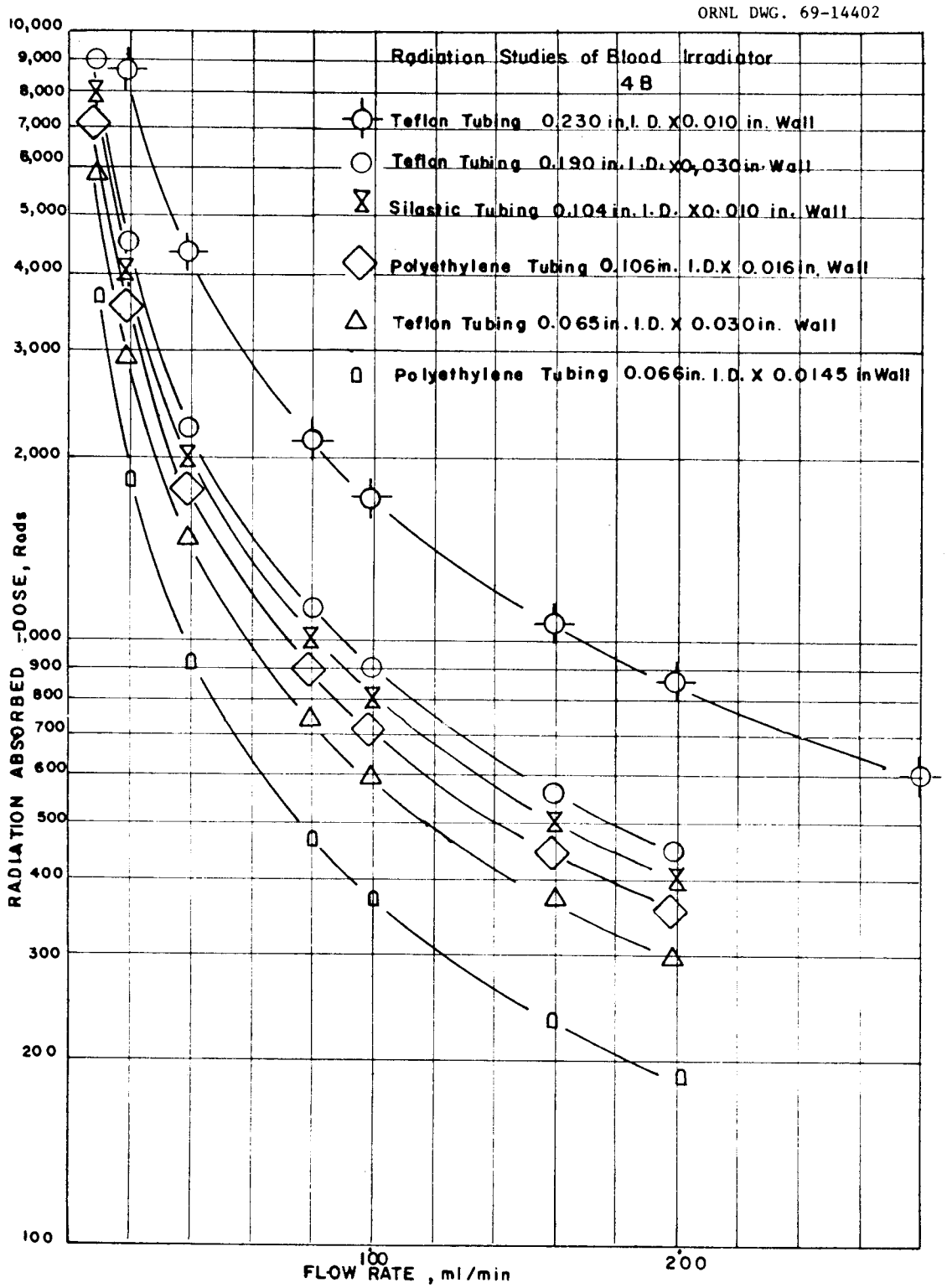


Fig. 4. Radiation Absorbed Dose vs Flow Rate for a Blood Irradiator.

A. Special Form Prototype Testing

Prototype sources are subjected to the tests prescribed for "special form material." On the basis of these tests, sources of similar design may be judged to pass the requirements for "special form material." No sources were tested. This is a continuing program.

B. Testing Sources with Manufactured Leaks

Sources with manufactured leaks are tested to determine the relationship between leak hole size and the quantity of activity which escapes under various normal and accident conditions. These sources are also used to evaluate the various leak test methods. No sources were tested. This is a continuing program.

C. Surveillance of Regulations and Cask Evaluations

Up-to-date information on radioisotope regulations is maintained by review of the Federal Register. Comments approving the proposed USA DOT Certificate of Compliance for radioactive material packaging were submitted to the Oak Ridge Operations Office. This certificate is proposed to replace special permits, which according to law are issued as exceptions to regulations when USA DOT feels it is in the public interest. Radioactive material packaging is permitted by regulations; therefore, the legal branch of the DOT feels the certificates should be issued.

A preliminary survey of radioactive material regulations has been completed and forwarded to DID. The efforts of approximately 150 organizations were reviewed and areas of conflict were pointed out.

An ORNL Isotopes Division Guide for the Packaging of Radioactive Material has been prepared and will be published as a TM report. This guide presents packaging requirements for radioactive material in a straight forward, simplified form for use by personnel not familiar with the many regulatory changes regarding radioactive material.

Large quantity radioactive material shipping packages are evaluated prior to application for AEC certification and petitions for USA DOT approval. The SNAP-7E Thermoelectric Generator, the Canadian Transfer Case F-143, the ORNL Lithium-Shielded Curium Shipping Cask, and the ORNL Neutron-Shielded Fire and Impact Shield have been evaluated to determine compliance with the Safety Standards of AEC Manual Chapter 0529. The SNAP-7E device is a strontium-90 powered thermoelectric generator presently operating a sonar reference point in the Atlantic Ocean and is scheduled for return to ORNL for examination. The Canadian Transfer Case F-143 is a Canadian-approved lead-shielded cask with a wooden fire and impact shield to be used to transport cobalt-60 from ORNL to Puerto Rico Nuclear Center.

The ORNL Lithium-Shielded Curium Shipping Cask was designed and is being built to transport and store Savannah River Operations 100-g curium-244 capsules. This cask, with 13-1/2 in. of lithium hydroxide monohydrate

as fast-neutron shielding equivalent to a foot of water, complies with all transport regulations. It has a heat dissipation capability of 250 W. The shielding material with its free moisture content and water of hydration effectively meets the requirements of the hypothetical thermal accident through evaporative cooling. The cask, 36 in. in diameter by 58 in. in height, weighs 3500 lb.

The ORNL Neutron-Shielded Fire and Impact Shield was designed and is being built to replace the Sugarman Cask Wooden Fire and Impact Shield. This package is composed of two concentric stainless steel cylinders which form the cladding for 4-3/4 in. of lithium hydroxide monohydrate. This package will furnish equivalent protection as the wooden shield but with reduced maintenance and increased thermal loading capacity. A request for a broad coverage certification has been made in the AECM 0529 submission to include all USA DOT Specification 55 containers with weights up to 2500 lb. The outer packaging weighs 1550 lb and will dissipate 500 W of decay heat.

A lightweight package for the transport of californium-252 and other fast neutron-emitting radionuclides is being designed and will be evaluated. It is essentially a 10-gal drum of paraffin centered in a 55-gal drum of lithium hydroxide monohydrate. Shielding will be equivalent to 6 in. of paraffin plus 5 in. of water. The package will be under the 800-lb limit set by regulations.

New designs of shipping containers are tested to determine their effectiveness during transport. These casks presently being designed or constructed will be tested to determine shielding effectiveness and thermal dissipation capability prior to use in transport. Testing will begin in December.

D. Source Leak Test Guide

The various leak test methods are evaluated to determine their reliability and sensitivity for various source designs.

A guide for leak testing encapsulated radioactive sources is being edited for publication. This guide discusses each of the common leak test methods from the viewpoint of reliability and sensitivity. A considerable portion of the report is given to the presentation of laboratory data obtained by leak testing sources having known leak hole diameters.

E. Sealed Source Testing

Sealed sources are tested to evaluate their ability to contain the radioactive material under normal and abnormal conditions.

No sources were tested during October and November. This is a continuing program.

F. Unsealed Source Testing

Unsealed sources are tested to evaluate their ability to contain the radioactive material under both normal and abnormal conditions.

Testing of commerical tritium foils was continued. Tests were made to determine the amount and the chemical form of tritium released from the foils at various temperatures over the range of 100 to 600°C. The results of these tests are given in Table 3. Most of the losses were in the form of tritium oxide.

Table 3. Tritium Losses at Elevated Temperatures[a]

Temperature, °C	Tritium Loss, % of initial activity			
	1 Ci/in.^{2} b	4 Ci/in.^{2} b	8 Ci/in.^{2} b	15 Ci/in.^{2} b
100	0.01	0.00043	0.00037	0.00017
200	1.26	0.064	0.0199	0.00445
250	5.28	0.561	0.0891	0.021
300	18.4	1.25	0.265	0.120
350	38.4	3.25	0.846	0.563
400	47.7	5.61	2.03	0.926
500	99.3	24.9	10.4	5.81
600	100	71.0	56.0	32.5

[a]Over a 2-hr period of flowing dry air.
[b]Nominal activity of foil.

G. Examination of Commercial Sources

Commercial sources that have been involved in fires or that may fail in use are examined to determine their condition. Other commercial sources are tested as previously stated under sealed sources and unsealed sources.

A commercial krypton-85 beta gage source which leaked in use has been received from the gage manufacturer and will be tested to determine where the leak hole is located and if possible whether the leak is a puncture type leak or an opening in one of the seal areas.

TECHNOLOGY UTILIZATION - 08-01-05

A. Information Center

As a result of intensive indexing and accessioning effort, the IIC now has 15 thousand items available for searching by keywords. The backlog that accumulated while the IIC Thesaurus was being revised and while computer compatibility with the MT/ST was being worked out is now less than 1000 items. Shortly after the first of the year we expect to be accessioning on a current basis.

During October and November, 170 requests for information were filled. Several articles and reports published the last two months, The Isotope User's Guide, the JARI article on Tagging of Small Animals for Tracking Purposes, and the machine translation of Nuclear Geophysical Methods of

Analyses, have proved very popular, and we have distributed 900, 150, and 130 copies of these, respectively. Requests for Special Sources of Information on Isotopes continue to come in, with the total number distributed now at 4000. Another hundred copies of the neutron radiography review were sent to the New York Operations Office for use at a small-business workshop. A list of the reviews in process is given below:

Title	Author(s)	Status, % complete
Cement-Polymer Combinations	Martha Gerrard, P. S. Baker, R. E. Greene	60
DID Research and Developments - 1969	Isotopes Information Center Staff	80
Indium-113m: Preparation and Uses	Martha Gerrard	10
Flow Measurement	R. H. Lafferty, Jr.	5
Iodine-125	P. S. Baker, Martha Gerrard	25
Isotopic Methods of Examination and Authentication in Art and Archaeology	F. J. Miller, E. V. Sayre, B. Keisch	90
List of AEC Radioisotope Customers with Summary of Radioisotope Shipments, FY 1969	Ruth Curl, compiler	90
Milk Preservation by Ionizing Radiation: Literature Review	Martha Gerrard	In Reproduction
Oceanography	Isotopes Information Center Staff	90[a]
Patent Review - Process Radiation Development	R. E. Greene, P. S. Baker, Helen Warren	70
Potato Sprout Inhibition by Radiation	F. E. McKinney	10
Radiation vs. Glass	D. N. Hess	20
Radioisotopes in the Pharmaceutical Industry	P. S. Baker, Martha Gerrard	5
Radioisotopes in the Steel Industry	Republic Steel Corporation	10
Radioisotopes in X-Ray Fluorescence Analysis	R. H. Lafferty, Jr.	5
Selected Abstracts of World Literature on Production and Industrial Uses of Radioisotopes	Martha Gerrard, P. S. Baker	In Reproduction
Self-Diffusion in Liquids	F. J. Miller	75
Strontium-90	Roberta Shor, R. H. Lafferty, Jr., P. S. Baker	80
Technetium-99m: Preparation and Uses	Martha Gerrard, P. S. Baker	20
Radioisotopes in the Textile Industry	F. J. Miller	90
Use of Radioisotopes in Sedimentology	V. Romanovsky[b]	10
Wheat Disinfestation - A Worldwide Review	F. E. McKinney	5

[a]Draft completed; being held at DID.
[b]Translated and edited by Martha Gerrard.

In connection with the program established to enable the USAEC Divison of Reactor Development and Technology (RDT) and regulatory staffs, Commission contractors, and the nuclear industry to follow accomplishments on nuclear safety research and development contracts sponsored by RDT, the IIC was asked to prepare project summary information for the Program and Project Information File (PPIF). Ten contracts from the Environmental and Ocean Sciences category of DID-sponsored research were picked as being appropriate to the overall interest in environmental nuclear safety.

B. Isotopes and Radiation Technology

Issue No. 1 of Vol. 7 of Isotopes and Radiation Technology was received from the Government Printing Office (GPO) and distributed. Page proof for issue No. 2[a] was checked and released to GPO; most of the galley proof for issue No. 3 was received from DTIE, the copyeditors, and was checked and returned to them. Writing and editing for issue No. 4 were continued and are now about 60% complete.

C. Publications

Martha Gerrard, Tagging of Small Animals for Tracking Purposes: A Literature Review, Int. J. Appl. Rad. Isotop. 20: 671-6 (1969).

F. E. McKinney, S. A. Reynolds, and P. S. Baker, Isotope User's Guide, ORNL-IIC-19 (September 1969).

Nuclear Geophysics, Abstracts of Reports, machine translation, ORNL-TM-2668, part 1 (October 1969).

Nuclear Geophysical Methods of Analyses, Abstracts of Reports, machine translation, ORNL-TM-2668, part 2 (October 1969).

P. S. Baker, Robert H. Lafferty, Jr., Martha Gerrard, R. E. Greene, F. E. McKinney, F. J. Miller, and D. N. Hess (Eds.) and A. F. Rupp (Adviser), Isotopes and Radiation Technology 7(1) (Fall 1969).

The following articles appeared in Isotopes and Radiation Technology 7(1):

P. S. Baker, Idaho Nuclear Energy Commission
IAEA Sponsors Engineering Compendium on Radiation Shielding
ORNL Stable Isotope Inventory Augmented

Martha Gerrard, Miniature Iodine-125 X-ray Source
Sweden's Radioisotope-powered Navigational Light
Radioisotope Use in France---1966
Australian Radioisotope Production and Services---FY 1968
South African Radioisotope Production and Services---1967
Transportable Beta Irradiator
Statistics of Radiation Use in Japan---1968
Radiation Vulcanization of Natural Rubber in the Latex Phase: Study of a 1-Tonne/hr Pilot Plant
New Radiometric Viscometer
Russian 20-year Isotope Production Anniversary
Mutations in Plant Breeding
Radioisotope Production in the IAN-R1 Reactor
Isotopes in Chemistry (book review)

---

[a]A special Heart-Assist Devices issue (2500 extra copies will be distributed to a special mailing list supplied by Dr. Richard E. Hurley, President of the American Heart Association).

Martha Gerrard, Iodine-123: New Production Method
Canadian Biological Irradiator
Determination of Wear with Radioisotopes (book review)
Wear in Floor Covering Detected with Radioisotopes
Californium-252: Its Use and Market Potential
New French Isotope Catalog
New Technique for Rapid Preparation of ^{99}Mo from ^{99m}Tc Generator
Korean Iridium-192 Radiography Handbook
Radioisotopic Control for Pipeline Crawler
New British Radioisotope Catalog

Ralph Greene, Extracts from a Report on Concrete-Polymer Materials

D. N. Hess, A Summary of Recent Work by the French in the Development of Isotopic Thermoelectric Generators

F. E. McKinney, Measuring Moisture with Gamma Radiation
Nucleonics in Aerospace

F. E. McKinney and P. S. Baker, Reader Interest Check Sheet Summary

RADIOISOTOPE APPLICATIONS DEVELOPMENT

BASIC TECHNOLOGY DEVELOPMENT - 08-03-01

A. Liquid Scintillation Development

1. Alpha Radiation Damage in Phosphors

A detailed study of the emission spectrum of two phosphor-isotope sources has been completed. Two phosphors (CdS:Ag and YVO_4:Eu) were mixed with known amounts of $^{241}Am_2O_3$ and sealed in special holder assemblies. The isotope loading of the CdS was 0.5 Ci/g, while the YVO_4 was prepared at 3.0 Ci/g. The integrated intensity and the spectral distribution of the sources were measured over a 5-week period. The radiation dose for the CdS:Ag ranged from 0 to 5×10^9 rads and for YVO_4:Eu the range was 0 to 4×10^{10} rads. Although the spectral distribution of each phosphor has remained essentially unchanged, the luminous intensity has dropped markedly as shown in Table 4.

The data clearly show the greater radiation sensitivity of the sulfide phosphor over the vanadate material. These results will be considered in greater detail before further tests are started.

Table 4. Luminous Intensity as a Function of Radiation Dose

Radiation Dose, rads	Relative Luminous Intensity	
	CdS:Ag	YVO_4:Eu
0	1.0	1.0
7×10^8	0.40	-
1.4×10^9	0.22	-
2.1×10^9	0.15	0.89
2.8×10^9	0.11	-
3.5×10^9	0.085	-
4.2×10^9	0.070	0.79
8.4×10^9	-	0.64
1.26×10^{10}	-	0.53
1.68×10^{10}	-	0.44
3.36×10^{10}	-	0.37

The emission spectrum for the CdS was broad band in nature and exhibited peak emission at 680 nm. Although radiation damage with this phosphor was already severe at an exposure of 1.4×10^8 rads, no significant change in the spectral shape was observed. Even when the total emission had dropped to 7% of the original value, the spectral shape remained unchanged. The YVO_4:Eu phosphor exhibits sharp line emission; the maximum response is at 618.3 nm. Here also, no spectral shift could be observed with exposures as high as 4×10^{10} rads. However, some small changes in the relative intensities of the emission lines were noted. This response could be caused by the different reflectivity and/or absorbtivity of the damaged phosphor.

Two new phosphor-isotope samples are being prepared for similar studies.

2. Cerenkov Counting

During the process of the formation of Cerenkov radiation, a majority of the photons are produced in the ultraviolet region of the spectrum. Much of the available energy is thus lost to the counting system because of the limited sensitivity of the phototubes in this spectral region. A few reports suggest that a wave-shifter can be used to convert some of the ultraviolet photons to visible photons and therefore increase the observed counting efficiency; however, detailed studies of possible wave-shifting compounds are not available.

An initial study of 12 likely wave-shifting materials was recently completed. Of these, three compounds (4-methylumbelliferone, 2-naphthol-3,6-disulfonic acid disodium salt, 7-amino-1,3-naphthalenedisulfonic acid) have shown up to a 40% increase in the counting efficiency of ^{90}Sr-^{90}Y. These tests will be continued with lower energy emitters and the results will be prepared for publication.

SYSTEMS ENGINEERING APPLICATIONS - 08-03-02

A. Oceanographic Systems Study

One of the major goals of the RIST program is to develop techniques to determine volume of sediment transport. To accomplish this, several tasks must be completed: 1) development of a new injection technique, 2) improvement of stationary detector designs, and 3) development of computer programs to analyze the data.

One of the problems associated with determining sand volume transport is the lack of a technique to relate the gold-198 tagged sand to the total quantity of sand being transported. Injection techniques previously employed placed the gold-198 tagged sand on the surface of the ocean bottom. This material is subject to immediate dispersion and can present a false indication of sand transport. A possible solution to this problem is to physically mix the gold-198 tagged sand with a known volume of sand on the bottom in such a manner that the bottom topography is not seriously altered. A prototype device (Fig. 5) for injecting sand in this fashion has been designed and constructed. Laboratory tests are being conducted and the device will be field tested at Oceanside, California, in February 1970. Basically, the device is a steel cylinder with an electric stirrer mounted in the center. While the stirrer is in operation the cylinder is lowered into the sand by utilizing two air-operated rams. The gold-198 tagged sand will be introduced when the cylinder has been inserted to the proper depth. After mixing, the cylinder is withdrawn and leaves a well defined core. During an experiment this core will be monitored continuously with a stationary detector.

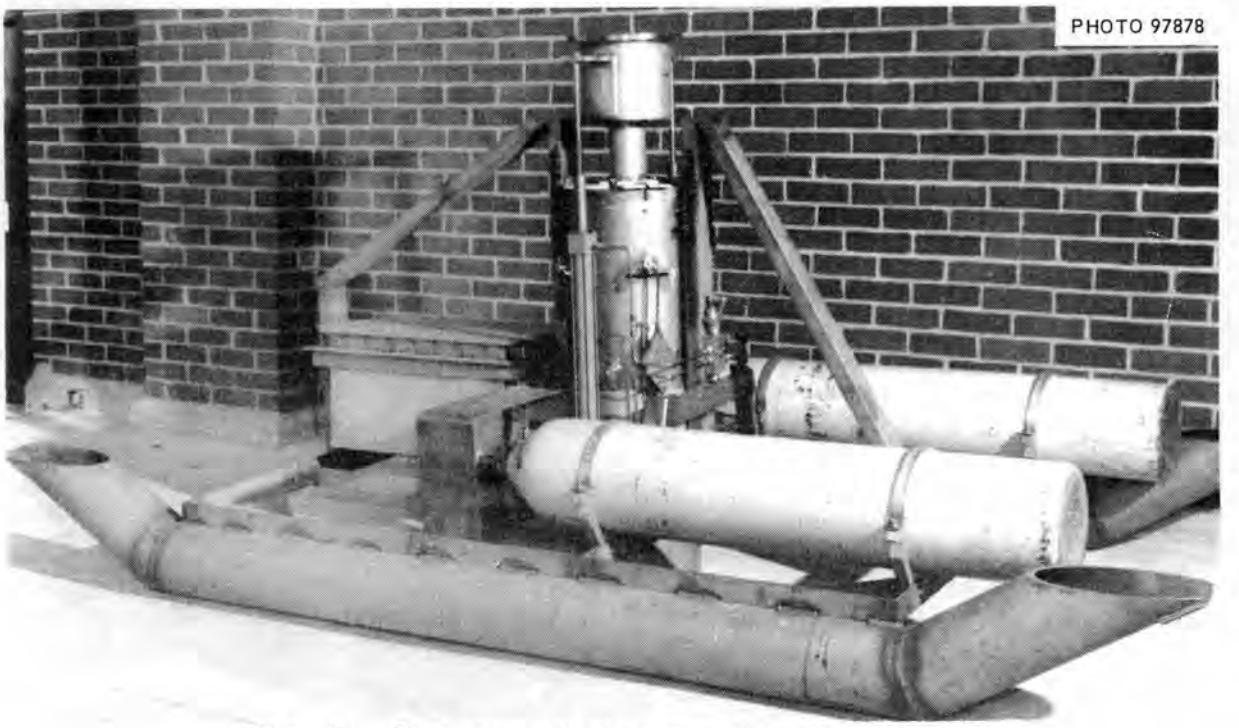


Fig. 5. Prototype Device for Injecting Sand.

The stationary detector assemblies that we have attempted to use in the past have proven to be too fragile. We have purchased a steel-armored cable which will eliminate the failures caused by rough handling during the deployment of the detectors and the stresses resulting from the water and sand in the breaker zone. The Geiger-Muller counter will be housed in a metal holder which will be connected to the steel cable. The holder will be filled with silastic to protect the counter from water and shock. Laboratory tests on this assembly should be completed within four weeks.

Although computer programs have been written that provide survey maps and radiation contours, several refinements need to be made. To express sediment volume rate of transport as a vector quantity, a logical sequence of data reduction must take place: 1) conversion of raw data to a meaningful form of maps and printed copy, 2) interpolation between data points to provide a grid of the radioactivity, and 3) calculation of the centroid of radioactivity and a material balance.

The first step of converting raw data to a meaningful form has been completed. The maps consist of a boat track, corrected radiation count rates along the detector track (not the same track as the tow vessel), significant count rates (count rate greater than background), and a composite of the significant count rages from any number of surveys at a given site. Maps are produced on a calcomp plotter from magnetic tapes written by the computer. Two thousand lines of data (14,000 data points) may be handled for any one survey and several surveys can be treated consecutively to obtain a composite map. Printed output information consists of line number, time, boat x-y location, detector x-y location, corrected radiation counts, and depth of water. Another output from this program is the storage of radiation data, water depth in feet, and detector location (x and y) on a data disk. The disk has random access with a storage capacity of five million bits of information. This allows all data from one experiment, which has received preliminary corrections, to be stored and permits very fast retrieval time without having the lengthy first program in the core of the computer.

The second requirement, interpolation between data points to provide a grid, has been worked on but the problem has not been solved. A program has been written as a stop-gap measure until a more sophisticated program can be developed. This program fills in data between data points acquired during a survey by double linear interpolation (x and y directions). The result is a complete map enclosed by the outer survey tracks. From these maps radiation intensity contours may be derived with some integrity. This program does not correct radiation data for burial and only does a linear interpolation. The distribution along a straight line position of a survey track is not linear; therefore, this interpretation technique is not valid. A program is being developed that will fit an equation to the actual distribution along a survey track and compute regularly spaced data points to form a grid. The new injection technique (core device) discussed above will provide additional burial information which will permit burial corrections. The third accomplishment, centroid and material balance calculations, can easily be performed once a valid grid of the radioactive tracer has been generated. Current efforts are being directed toward this goal.

1. Point Mugu Experiment

A field experiment was conducted at Point Mugu, California. Field operations began on September 22, 1969, and were completed on October 2, 1969. The experimental site was in the near vicinity of a groin (at this time the groin is a pier perpendicular to the beach) which is under construction by the U. S. Army Corps of Engineers. The purpose of the experiment was to gain additional knowledge concerning sand transport with specific emphasis on gaining information about sand transport through the groin before the structure is completed. After the groin has been completed, a second experiment will be conducted to determine the change in sand transport patterns.

Three injections of gold-198 were made and approximately one mile of beach was utilized as the survey area. Only minor equipment problems were experienced and they did not delay or compromise any of the radiation surveys. Weather was good and the sea conditions (4- to 5-ft breakers) were relatively calm throughout the test.

Since past observations indicated a southward movement of sand, the first injection (12.8 Ci) was made 1000 ft north of the groin. The tagged sand was distributed as a line source perpendicular to the beach, starting seaward of the low tide line and extending for approximately 360 meters. Before the injection, a drogue (a small buoy equipped with an underwater anchor that offers resistance to water current but does not touch bottom - the forces acting upon the anchor causes the buoy to travel in the general direction and at the approximate velocity of the underwater current) study was conducted and the results indicated a mixture of current movements, both north and south. Five hours after the injection of the tagged sand, a survey was conducted which established a definite northward movement. This northward movement continued for 2 days and then began to shift southward on the third day. Two conclusions, based on visual observations of the radiation detection equipment, were reached: 1) because of the northward dispersion of the gold-198 tagged sand, the sand passing through the groin would not contain sufficient radioactivity to produce an acceptable radiation pattern, and 2) most of the gold-198 tagged material transported came from that part of the line source that lay in the breaker zone.

Based on these conclusions, a second injection was made 300 ft north of the groin on September 28. This injection contained 3.9 Ci of radioactivity and was dispensed as a line source perpendicular to the shore. The line started seaward of the low tide mark and extended approximately 150 meters. The subsequent radiation surveys of this injection established a southward movement through the groin. The bulk of the tagged sand transported was from a short segment of the injection line that was in the breaker zone. Although the remainder of the injected material (approximately 80%) did not move, a suitable transport pattern through the groin was obtained.

A third injection was made on September 30, 1969, and was located approximately 1000 ft south of the groin. The injected material contained 2.4 Ci

of gold-198 and was placed as a point source in the breaker zone. After 24 hr, a survey indicated that all of the tagged sand was transported approximately 1000 ft southward of the injection site. In previous experiments, some tagged material remained at the injection site. This severely complicated the data treatment because in determining a transport rate it is necessary to calculate the centroid of the injected material and the centroid of the transported material. If tagged material remains at the injection site it is difficult to determine the quantity of material remaining since it is in a small concentrated area. If all of the material is transported, the centroid can easily be determined.

This discussion on the results of the Point Mugu experiment is preliminary in nature since only visual observations of the data displayed on the radiation detection equipment is available at this time.

B. National Security

Nothing to report.

RADIATION PROCESSING APPLICATIONS DEVELOPMENT

ORGANIC SYNTHESIS WITH ISOTOPIC SOURCES - 08-04-01

A. Woolen Mill Effluent

The objective of gamma radiolytic treatment of mill effluent was to accomplish what conventional treatment could not in a short period of time: 1) color removal, 2) destruction of dieldrin, and 3) significant lowering of biological oxygen demand (BOD). Conventional treatment of lagooning for several days to several months can lower BOD, which varies according to the organics present. Additives are introduced to promote efficiency. Slight color removal occurs. In our experiments dramatic color removal, destruction of dieldrin up to 99%, and lowering of BOD by 97% were effected by variations of oxygen pressure and total gamma dose. Table 5 lists the conditions under which the four woolen mill effluent samples were irradiated. The dose rate was 6.8×10^6 R/hr.

The BOD analysis on each experiment required at least 100 ml. Only 500 ml each of the four woolen mill effluents were received. Thus we were unable to work out optimum conditions for color removal at this time and had to rely on past experiments for dose and pressure. Color removal was accomplished in all but the scouring waste which consisted of high concentrations of lanolin and soap. Dieldrin and BOD analyses will be done by Southeast Water Laboratory in Athens, Georgia.

Table 5. Radiation Conditions for Woolen Mill Effluent Samples

Volume, ml	Oxygen Pressure, psi	Time Irradiated, min	Total Dose, R
	Yellow-Gold Dye Vat Solution		
100		12	1.36×10^6
100	1600	6	6.8×10^5
100	1600	12	1.36×10^6
100	1600	12	1.36×10^6
	Bright Orange Dye Vat Solution		
100		12	1.36×10^6
100	1600	10	1.13×10^6
100	1600	12	1.36×10^6
100	1600	12	1.36×10^6
	Scouring Waste		
100		71	8×10^6
100	2000	71	8×10^6
100	2000	71	8×10^6
100	2000	283	3.2×10^7
	Total Woolen Mill Waste		
100		71	8×10^6
100	2000	71	8×10^6
100	2000	71	8×10^6

RADIOISOTOPE SALES

Orders were received from Radiochemical Centre, England, for 90,000 Ci of tritium; from U. S. Radium Corporation for 30,000 Ci of tritium; from Dhom Products, Inc. for 10,001 Ci of tritium; from General Nucleonics Corporation for 200 Ci of krypton-85 enriched to 14%; and from United Aircraft Corporation for 225 Ci of krypton-85 enriched to approximately 24%.

An authorization was received from the USAEC to supply American Atomics Corporation with 450 Ci of krypton-85 enriched to approximately 27% for a contract with the Air Force. American Atomics Corporation requested that 300 Ci be shipped to them during December 1969 and 150 Ci be shipped during June 1970.

A request was received from the Australian Embassy, Washington, D. C., concerning the price and availability of fission-produced molybdenum-99. Further telephone conversations with them indicate that they will be placing an order for weekly shipments, if possible, of approximately 2 Ci or biweekly shipments of approximately 10 Ci.

A verbal request was received from J. L. Shephard and Associates inquiring about the possible supply of a cesium-137 source having active dimensions of 12 x 12 x 1 in. and containing approximately 250,000 Ci of cesium-137. We agreed to send them details of the strip-type cesium-137 source for their consideration and asked them to furnish us more precise information on their requirements.

Shipments made during October and November included 11,500 Ci of cesium-137 to Lockheed-Georgia Nuclear Laboratory (material is in the form of sources - activity has been billed in previous fiscal year), 10,000 Ci tritium to U. S. Radium Corporation, 1,750 mg of iodine-129 to McDonnell Douglas Astronautics Company, and 30,000 Ci of tritium to Radiochemical Centre, England.

A list of outstanding large orders is shown below:

Isotope	Amount, Ci	Customer
Cesium-137	300,000	Brookhaven National Laboratory[a]
Cesium-137	~87,000	Lockheed-Georgia Nuclear Laboratory[a]
Cesium-137	~180,000	American Nuclear Corporation[a]
Cesium-137	~207,000	U. S. Army, Natick Laboratories
Cesium-137	205,000	Industrial Reactor Laboratories, Inc.
Cesium-137	1,180,000	Radiation Machinery Corporation
Cesium-137	~194,000	Atomic Energy of Canada Limited
Cesium-137	200,000	General Nuclear, Inc.
Cesium-137	40,000	Radiation Resources, Inc.
Cesium-137	20,000	Radiochemical Centre
Cesium-137	200,001	CEA, France
Cesium-137	200,001	Radiation Resources, Inc.
Cesium-137	~5,000	Miscellaneous orders
Strontium-90	300,000	CEA, France
Strontium-90	~32,000	Aerojet General
Strontium-90	~532,000	Isotopes Inc.
Strontium-90	211,323	Isotopes Inc.[a]
Strontium-90	~41,000	Brookhaven National Laboratory
Strontium-90	~2,000	Miscellaneous orders
Strontium-90	4,951	Radiation Machinery Corporation
Promethium-147	500	Minnesota Mining and Manufacturing Co.
Promethium-147	5,100	Reactor Centrum Nederland
Carbon-14	1,445 mCi	Catholic University

[a]Activity has been filled in previous fiscal years; material is being fabricated into sources or has been fabricated into sources and stored for request for shipment.

Tables 6 and 7 give the internal and external customers and runs for October and November 1969.

Table 6. Internal Cyclotron Customers and Runs for October and November 1969

Customer	Product	Target	Time, hr:min			Total Charges
			Beam	Misc	Total	
ORAU	Gallium-67	Zinc	3:00	1:15	4:15	$438.85
	Gallium-67	Zinc	1:00[a]	1:15	2:15	248.85
	Gallium-67 (stock)	Zinc	2:00 :10	1:00	3:00	320.10
	Gallium-67 (stock)	Zinc	1:00 :15	1:00	2:00	225.10
Physics	Cobalt-61	Nickel-64	2:45	2:05	4:50	469.17
	Cobalt-61	Nickel-64	1:15	1:00	2:15	215.83
	Cobalt-61	Nickel-64	1:15	1:15	2:30	247.50
	Cobalt-61	Nickel-64	1:15	1:15	2:30	247.50
Y-12	Cobalt-56	Iron	4:00	:50	4:50	542.96
Isotopes	Bismuth-206	Lead-206	1:00	1:05	2:05	217.37
	Indium-111	Cadmium-111	2:00	1:15	3:15	308.75
Stock	Bismuth-207	Lead	8:00	1:15	9:15	

[a]Actual innage was 1 hour and 20 min; additional time to correct for decay.

Table 7. External Cyclotron Customers and Runs for October and November 1969

Customer	Product	Target	Time, hr:min Beam	Misc	Total	Total Charges
Mound Laboratory	Cobalt-56	Iron	4:00	:50	4:50	$804.22
New England Nuclear Corporation	Yttrium-87	Strontium-87	2:00[a]	1:15	3:15	540.10
	Rubidium-84	Krypton	1:30	:25	1:55	341.67
	Yttrium-87	Strontium-87	3:30	1:15	4:45	750.10
	Yttrium-87	Strontium-87	3:30	1:15	4:45	750.10
	Rubidium-84	Krypton	1:30	:25	1:55	341.67
	Yttrium-87	Strontium-87	3:30	1:15	4:45	750.10
	Cobalt-57	Nickel-58	50:00	1:15	51:15	b
N. V. Philips-Duphar	Rubidium-84	Krypton	2:00	:36	2:36	473.99
International Chemical & Nuclear Corporation	Cobalt-57	Nickel-58	48:00	1:15	49:15	b
Huntington Hospital	Rubidium-84	Krypton	8:00	:50	8:50	1,383.32
Winnipeg Hospital	Rubidium-84	Krypton	1:00	:25	1:25	271.67
Boston University	Bismuth-205	Lead-206	1:00	1:15	2:15	335.50
Stock	Rubidium-84	Krypton	2:30			

[a]Actual innage was 3.5 hours; adjustment made for shipment September 20, 1969.
[b]Purchased but not shipped.

ISOTOPES SALES AND SHIPMENTS

The radioisotope sales proceeds and shipments for the first four months of fiscal years 1969 and 1970 are given in Table 8.

Table 8. Isotopes Sales and Shipments

Item	7-1-68 thru 10-31-68	7-1-69 thru 10-31-69
Inventory items	$161,002	$172,095
Major products	36,017	39,222
Radioisotope services	73,201	63,500
Miscellaneous processed material	25,342	32,182
Packing and shipping	26,909	24,202
Cyclotron irradiations	56,719	40,933
Total Radioisotope Sales	$379,190	$372,134
Total Radioisotope Shipments	930	803

RADIOISOTOPE PRODUCTION

CESIUM-137 PROCESSING

All ^{137}CsCl product has been blended, assayed, and removed from the cells to various storage carriers. A total of 236,000 Ci was transferred to Building 3029 for fabrication of BNL-type ^{137}CsCl sources.

STRONTIUM-90 PROCESSING

Disadvantages of the NH_4OH-NH_4CO_3 procedure are: (1) It requires approximately 4 hr of processing time to adjust the pH with the potassium compound procedure as compared with an average of 8 hr for the NH_4OH system. (2) The response of the pH on the addition of NH_4OH is much slower than with the potassium compounds. (3) It often requires the back-addition of 1 M HNO_3 to get the pH in the range of 3-5 for proper precipitation with NH_4CO_3. (4) The fumes from the NH_4OH are very obnoxious to operating personnel, and the time required for the handling of this material supplements this problem. (5) Plugging of the off-gas filtration system is encountered throughout the NH_4OH-NH_4CO_3 precipitation procedures. The filters become plugged at least once per hour during the NH_4OH addition and during any solution transfer operation. The problem has been completely eliminated through the use of the new technique.

As a result a KOH-$KHCO_3$-K_2CO_3 technique was investigated and incorporated in $^{90}SrTiO_3$ precipitation process. The pH of the feed $^{90}Sr(NO_3)_2$ is adjusted to 1.0 with 10 M KOH and the TiO_2 slurry added to the pH-adjusted feed. The quantity of TiO_2 is determined by the desired activity of the final SrTiO product. The $^{90}Sr(NO_3)_2$-TiO_2 mixture is reacted with 3 M $KHCO_3$ (to pH 3.0) and with 5 M K_2CO_3 (to pH 10) to form the $^{90}SrCO_3$-TiO_2 mixture. The precipitate is cooled, filtered, rinsed, and then calcined to the final $^{90}SrTiO_3$ product. Several processing advantages inherent in the KOH-$KHCO_2$-K_2CO_3 procedure are the ease of pH adjustment with the potassium compounds versus the use of ammonium compounds, the elimination of the troublesome fumes of NH_4OH, and the total elimination of the formation of NH_4NO_3 in the building off-gas system.

Two 50,000-Ci batches of $^{90}SrTiO_3$ were processed during this period using the new procedure. The material balance and analytical results for these two runs are shown below:

	Batch No. 8SR-69	Batch No. 9SR-69
Feed		
Strontium-90 concentration, mCi/ml	1,572	1,327
P-511 volume, liters	34.5	37.5
Total batch activity, Ci	54,234	49,762
TiO_2 Addition, g	855	743
pH Adjustment		
3 M $KHCO_3$, liters	3.2	1.0
5 M K_2CO_3, liters	11.0	10.0
Product - $^{90}SrTiO_3$		
Net weight, g	1,943	1,519
Specific activity, Ci/g (by calorimetry)	27.8	32.3
Specific activity, Ci/g (RCA)	31.7	32.8
Strontium, wt %	33.6	39.5
Calcium, wt %	2.0	2.4
Potassium, wt %	0.5	0.7
Total activity, Ci	54,015	49,064

Waste		
Strontium-90 concentration, mCi/ml	0.54	0.068
Volume waste, liters	1,021	140
Total activity, Ci	557	10
Total Strontium-90 Activity Balance, Ci		
Feed	54,234	49,762
Product	54,015	49,064
Waste	557	10
Unaccountable	+338	-668

The results indicate that an average strontium-90 yield of 99.1% was obtained for the two runs. It also indicates that the residual potassium concentration is less than 1.0% of product $^{90}SrTiO_3$.

MAINTENANCE

The agitator in the C38 crystallizer vessel was replaced. It is planned to check out the processing lines in cell 8 next month for possible leaks.

The inner safety pane was replaced in the cell 10W $ZnBr_2$-filled window. A cylindrical welding fixture was installed in this cell for the welding of $^{90}SrTiO_3$ sources and powder cans.

Ten loads of solid waste were transferred to the Solid Waste Disposal Facility during this period. The program of waste removal prior to the decontamination of cell 14 for general cell maintenance and modification is complete. A major decontamination program has been initiated on this cell and completion of the required modifications is scheduled for February 1, 1970. Cell 14 has been in operation for seven years without decontamination and repair.

There were a total of 22 manipulator repairs during the month. Of these, 15 were required because of boot failure and the remaining seven were required because of mechanical failure.

SOURCE FABRICATION

A. Cesium-137

Fabrication of BNL-strip rectangular $^{137}CsCl$ sources was placed on a routine three-shift operation at Building 3029. Due to the large number of these units scheduled for fabrication, all three source fabrication cells were set up for this work so that one cell is used for all $^{137}CsCl$ powder and pellet handling, one is used for welding and leak-testing, and one is used for finished source testing and decontamination. With this arrangement, the high contamination work is isolated from the source handling operation, resulting in less decontamination effort on completed sources.

To eliminate problems experienced previously with the deformation of sources of this type, modifications were made in assembly procedures in order to minimize contacts of the fuel or the encapsulation materials with organic matter or water, either of which could cause deformations if included in the sealed source. A more rigorous heat test (400°C for 4 hr instead of 200°C for 4 hr) was initiated for both welded liners and completed sources. Another modification was the inclusion of a pellet-baking cycle (400°C for 4 hr in air) prior to source loading. The original practice of baking loaded, uncapped liners before welding led to poor results in welding, probably due to formation of oxides in the weld zone. This was abandoned in favor of baking the pellets in separate containers then loading the baked pellets into liners and welding them immediately. The pellets are measured before loading to detect any changes in dimensions. The pellet-baking step serves to identify bad fuel batches before they are used for source fabrication. To date, two such batches have been identified. In the first case, the pellets swelled uniformly about 15%. The $^{137}CsCl$ was analyzed and was found to contain approximately 600 ppm of carbon, although the original analysis of this batch showed only 75 ppm of carbon. The second bad $^{137}CsCl$ batch has not yet been analyzed. Fractions of powder from both of these batches were baked at 400°C for 4 hr. The swelling of the pellets from the first batch was about one-half of that experienced with unbaked powder from the same batch, but there was no detectable swelling of the pellets made from baked powder from the second batch. Neither of these materials has been used for source fabrication.

Experience to date with the revised procedures has been encouraging. Of 46 liners fabricated there was one reject (a leak) and of 36 outer encapsulations there were three rejects, all due to deformation of the outer capsule. The three liners which were removed from the rejected outer capsules were rebaked and tested; then they were re-encapsulated in new outers with good results. At present there are 33 completed sources made by the new procedures on shelf test, 12 welded liners ready for final encapsulation, and pellet source loads for five more units being baked. The next 15 units scheduled for fabrication are "patterned" sources, requiring pellets of various lengths fabricated to close tolerances.

A group of 31 BNL-strip sources made by the earlier procedures was heat-tested at 400°C and then leak-tested and checked for dimensional changes. All of these sources passed the tests and will be shipped to Natick Laboratories. A cylindrical source containing 11,500 Ci of $^{137}CsCl$ as packed powder was tested, loaded into a customer-supplied irradiator, and shipped to Lockheed Corporation.

B. Strontium-90

Preparation of the four 55-W $^{90}SrTiO_3$ pellets was started. These pellets will be encapsulated in welded stainless steel cans supplied by the customer, Aerojet General Nucleonics. The fueled cans will become the liners for four sources prepared by AGN. Two pellets have been completed; the other two are in progress. After all pellets have been made, the canned pellets will be assayed by calorimetry, welded, and decontaminated for shipment.

SHORT-LIVED FISSION PRODUCT OPERATIONS

On November 21, 1969, Isotopes Sales received an order from the Australian Embassy (AAEC) requesting 12 Ci of fission-produced molybdenum-99 to be shipped on November 25, 1969. This material was produced from a uranium ring removed from the reactor on November 18, 1969. The following analytical results were obtained from the product having a volume of 200 ml.

Concentration, mCi/ml	700
Total activity, mCi	140,000
Radiochemical purity, %	>99.99
Specific activity, mCi/g	5,500,000

All analytical data is corrected to 8:00 AM November 25, 1969 (day of shipment). The required amount of activity was loaded onto a 10-g alumina column and shipped to the customer in a returnable container.

SPECIAL FORM MATERIALS

Packaging of 300,000 Ci of $^{90}SrTiO_3$ powder for shipment to the CEA (France) was started. This material will be packaged in approximately 22 welded stainless steel shipping cans, each 2-1/4 in. in diameter by 8 in. long. The cans will be shipped to France in two casks. Fifteen cans have been loaded, and six of these have been welded.

Source encapsulation and special form packaging operations planned for December are essentially continuations of work already in progress. The patterned BNL-strip sources will be completed; then encapsulation of a cycle of 40 fully loaded BNL strips will be started. Packaging of the 300,000-Ci bulk $^{90}SrTiO_3$ order and the four $^{90}SrTiO_3$ pellets will be completed. After completion of current strontium-90 encapsulation work, a miscellaneous $^{137}CsCl$ source run will be started.

OPERATIONAL SUMMARY

	Oct.-Nov. 1969		1969 Totals	
Cesium-137	Number	Amount, Ci	Number	Amount, Ci
HAPO shipments received	0	0	0	0
Powder batches prepared	0	0	20	231,000
Sources fabricated	34	155,000	296	535,000
Special form containers	0	0	0	0
Total shipments	3	12,000	50	368,000
Strontium-90				
HAPO shipments received	0	0	0	0
Powder batches prepared	2	100,000	9	307,000
Sources fabricated	0	0	0	0
Special form containers	0	0	5	2,000
Total shipments	2	100	53	919,000

PROCESS STATUS

Item	Cesium-137, Ci	Strontium-90, Ci
In-process solution	220,000	1,800,000
Product powder	52,000	780,000
Sources in fabrication	114,000	218,000
Completed sources	342,000[a]	674,000[b]
Total	728,000	3,472,000

[a]Includes cesium-137 borosilicate source (approximately 27,000 Ci).

[b]Includes the following:

Type Source	Ci
SNAP-7C	30,000
Weather Bureau	13,300
RCA	68,400
SNAP-23	163,700
Sentinel-25	209,000
SNAP-7B	189,600
Total	674,000

MISCELLANEOUS RADIOISOTOPE PROCESSING

Short-Lived Fission Products			Processed Units			Service Irradiations	
Radioisotope	No. of Batches	Amount, Ci	Radioisotope	No. of Batches	Amount, mCi	Type	No. of Irradiations
Xenon-133	2	1,100	Copper-67	6	70	Lunar soil	11
Iodine-131	2	200	Calcium-47	3	40	$^{168}Y_2O_3$	3
Tellurium-132	1	3	Barium-131	2	80	Nickel-64	1
Strontium-89	1	18	Rubidium-86	1	5	Germanium metal	1
Barium-140	1	70	Elemental iodine-131	2	400	Platinum-196	3
			Nickel-63	1	10,000	Zinc-68	1
			Bismuth-210	1	30	Vanadium metal	1
Total	7	1,391		16	10,625		21

ADMINISTRATIVE SECTION

Travel by IDC personnel and visitors to the IDC are given in Tables 9 and 10, respectively.

Table 9. Travel of IDC Personnel

Site Visited	Purpose of Visit
Palo Alto, California	Attend National Academy of Science Subcommittee meeting
San Diego, California	Attend meeting on sediment transport studies
Paducah, Kentucky	Invited lecturer at ACS local meeting

Table 9. continued

Site Visited	Purpose of Visit
Nashville, Tennessee	Attend Southeastern Chapter Society of Nuclear Medicine meeting
Argonne, Illinois	Attend AEC Safeguards Course
Seattle, Washington	Present paper at 16th National Vacuum Symposium
Los Alamos, New Mexico	Attend AEC Symposium on Nuclear Safeguards
Sewannee, Tennessee	Attend Tennessee Academy of Science meeting
Dayton, Ohio	Attend Isotopic Fuels and Materials Committee Meeting at Mound Laboratory
Knoxville, Tennessee	Attend training session on use of MT/ST
Washington, D. C.	Meeting with AEC-SNS concerning recovery of SNAP-7E generator
	Program discussions with AEC-DID
	Attend DID Advisory Committee (ACIRD) meeting
	Discussions on isotope kilowatt program with AEC and 3M Company
	Attend Research Materials Meeting at AEC Headquarters
	Aid in writing and editing DID project summaries

Table 10. IDC Visitors

Visitors (affiliation)	Purpose of Visit
Battelle Northwest Laboratory	Visit hot cells and discuss cesium-137 encapsulation
AECL, Canada	Discuss purchase of cesium sources

Table 10. continued

Visitors (affiliation)	Purpose of Visit
Dow Chemical Company Rocky Flats	Inspect our cells and learn how to handle work in a remote facility
Transnuclear, Inc. France	Strontium-90 and cesium-137 separations for CEA, France
Stanford University	Cooperative on use of isotopes
Missile Intelligence Library Redstone Arsenal	Tour Isotopes Information Center
USAMICOM Redstone Arsenal	Tour Isotopes Information Center
University of Munchen West Germany	Price and availability of curium-244, americium-243, and californium-252
Massachusetts Institute of Technology	Tour of 86-Inch Cyclotron
National Physical Research Laboratory, Petoria, South Africa	Discuss high beam currents on production targets and other phases of research program at the ORNL 86-Inch Cyclotron
University of Miami	Technical discussion of operation of ORNL 86-Inch Cyclotron
Isotopes, Westwood Laboratories Westwood, New Jersey	Discuss plans for using the high current proton beam on the ORNL 86-Inch Cyclotron as a source for producing high energy neutrons in order to study the high energy neutron fission cross section of xenon-133 and xenon-135 from uranium-235 and plutonium-239

INTERNAL DISTRIBUTION

1. A. L. Allen
2. M. A. Baker
3. P. S. Baker
4. E. E. Beauchamp
5. G. E. Boyd
6-7. T. A. Butler
8. F. N. Case
9. J. A. Cox
10. W. C. Davis
11. J. S. Drury
12. J. H. Gillette
13. H. R. Gwinn
14. R. F. Hibbs
15. K. E. Jamison
16. Lynda Kern
17. C. V. Ketron
18. E. H. Kobisk
19. E. Lamb
20. R. E. Leuze
21. J. L. Liverman
22. L. O. Love
23. W. S. Lyon
24. H. G. MacPherson
25. R. E. McHenry
26. F. M. O'Hara
27. W. W. Parkinson
28. J. J. Pinajian
29. M. E. Ramsey
30. S. A. Reynolds
31-32. R. A. Robinson
33. D. A. Ross
34. A. F. Rupp
35. R. W. Schaich
36. K. A. Spainhour
37. M. R. Skidmore
38-39. H. F. Stringfield
40. A. M. Weinberg
41. J. C. White
42-43. Central Research Library
44. Document Reference Section
45-49. Laboratory Records Department
50. Laboratory Records - RC

EXTERNAL DISTRIBUTION

51. D. S. Ballantine, AEC, Washington, D. C.
52-55. R. F. Barker, AEC, Washington, D. C.
56. N. F. Barr, AEC, Washington, D. C.
57. O. M. Bizzell, AEC, Washington, D. C.
58. C. R. Buchanan, AEC, Washington, D. C.
59. R. L. Butenhoff, AEC, Washington, D. C.
60. T. D. Chikalla, PNL, Richland, Washington
61. D. F. Cope, AEC Site Representative, ORNL
62. D. C. Davis, AEC, Oak Ridge
63. J. C. Dempsey, AEC, Washington, D. C.
64. W. K. Eister, AEC, Washington, D. C.
65. E. E. Fowler, AEC, Washington, D. C.
66. J. D. Goldstein, AEC, Washington, D. C.
67. F. D. Haines, AEC, Washington, D. C.
68. J. W. Irvine, MIT (Consultant)
69. J. E. Machurek, AEC, Washington, D. C.
70. J. N. Maddox, AEC, Washington, D. C.
71. J. C. Malaro, AEC, Washington, D. C.
72. W. E. Mott, AEC, Washington, D. C.
73. J. A. Powers, AEC, Washington, D. C.
74. J. G. Rotariu, AEC, Washington, D. C.
75. B. A. Ryan, AEC, Richland, Washington
76. W. D. Sandberg, AEC, Aiken, S. C.
77. S. J. Seiken, AEC, Washington, D. C.
78. W. F. Sheely, PNL, Richland, Washington
79. R. W. Shivers, AEC, Washington, D. C.
80. L. G. Stang, Jr., BNL, New York
81-86. D. H. Turno, SRL, Aiken, S. C.
87. A. R. Van Dyken, AEC, Washington, D. C.
88. Laboratory and University Division
89-103. Division of Technical Information Extension