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RADIOISOTOPE PROGRAM PROGRESS REPORT FOR JULY 1973

J. H. Gillette

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ISOTOPES DEVELOPMENT CENTER

RADIOISOTOPE PROGRAM PROGRESS REPORT FOR JULY 1973

J. H. Gillette

Work Sponsored by
AEC Division of Biomedical and
Environmental Research

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RADIOISOTOPE PROGRAM PROGRESS REPORT FOR JULY 1973

J. H. Gillette

RADIOISOTOPE PRODUCTION AND MATERIALS DEVELOPMENT

REACTOR-PRODUCED ISOTOPES

Promethium-147 *(J. K. Poggenburg)*

A large quantity of ^{147}Pm is available as a byproduct of fission reactor operations. This nuclide, with a 2.6-year half-life and low beta energy ($E_{\beta\ max}$ = 0.225 MeV), has a gamma emission of 0.12 MeV in very low abundance (0.004%). These properties make ^{147}Pm potentially useful for biomedical applications as a power source if the level of ^{146}Pm (5.5 year) can be reduced to less than 0.1 part per million from the typical levels of 0.2 to 0.5 ppm found in byproduct materials. The object of this project is to determine the practicality of reducing the ^{146}Pm by thermal neutron irradiation.

The ^{146}Pm contaminant is produced by the $^{147}Pm(n,2n)^{146}Pm$ and $^{147}Pm(\gamma,n)^{146}Pm$ reactions. An effective thermal neutron cross section of ∿8400 barns was determined by Pinajian and Allen[1] while a value as low as 3500 barns has also been reported. Calculations both at ORNL and BNWL have indicated that ^{146}Pm can be "burned out" by neutron irradiation because of the relatively larger cross section of ^{146}Pm than the cross section of 182 barns for ^{147}Pm. The problem resolves to irradiating the material in a site with a small fast neutron component of the flux and a low gamma flux. In practice, the presence of aluminum as a major structural component in reactors creates a problem, because the 7.72-MeV capture gammas in aluminum produce ^{146}Pm. An ideal facility for this irradiation is provided by the HFIR in the beryllium reflector. For such an irradiation, a capsule fabricated of beryllium is also indicated.

As a preliminary effort in this project, the cross-section measurement of Pinajian[1] will be repeated with a modification of experimental design to resolve the effective cross section into its thermal and resonance integral components. This may help to resolve the variation in reported values. Knowledge of the contribution of resonance capture is important in the design of a target configuration for a practical test of ^{146}Pm burnout. The experiment will consist of irradiating ^{146}Pm produced on the ORNL 86-Inch Cyclotron with and without cadmium shielding. During this stage of the project, the analytical techniques for determining low-level changes in the ^{146}Pm content in ^{147}Pm will be investigated.

---

[1] J. J. Pinajian and J. F. Allen, *J. Inorg. Nucl. Chem.* 29, 2117 (1967).

A second phase of this project would be the design and irradiation of a practical sample of ^{147}Pm in the HFIR to determine the net burnup of ^{146}Pm.

Reactor Products Pilot Production *(R. W. Schaich)*
(Production and Inventory Accounts)

Processed Units	
Radioisotope	Amount (mCi)
Copper-67	18.72
Calcium-47	20.91

ACCELERATOR-PRODUCED ISOTOPES

Potassium-43 *(L. C. Brown and J. K. Poggenburg)*

Potassium-43 has been shown to be an excellent tool for studying blood perfusion in the heart and for diagnosing myocardial infarctions. Potassium-43 is currently produced in millicurie quantities in the HFIR by the $^{43}Ca(n,p)^{43}K$ reaction. The objective of this project is to investigate the large-scale photonuclear production of this radionuclide using the Oak Ridge Electron Linear Accelerator (ORELA). The ultimate goal is to develop the technology required to produce curie quantities of ^{43}K at costs that will allow its widespread clinical application as a primary agent for the early detection and diagnosis of heart disease.

The marble ($CaCO_3$) slab experiment described last month[2] was irradiated at a low power level, ≤12 W, for 46 min at the 20° port position of the ORELA beam energy spectrometer. This irradiation, which was originally scheduled for 24-hr duration, was prematurely terminated when the spectrometer switching magnet arced and failed. As a result, the induced ^{43}K activity was insufficient for the extensive rectilinear scanning originally envisaged. All slabs were scanned with a hand-held radiation monitor as the target was disassembled. The induced activity was found to be highly localized in the front end slabs proximal to the electron beam entry and spread only slightly with depth — consistent with the calculated photon beam divergence angle of ∿3°.[3] The first and fifth slabs of the 22-slab target assembly were scanned with a rectilinear scanner, and the results will be reported after the data analysis has been completed. Sufficient activity was produced to allow determination of the decrease of activity with depth and the total induced ^{43}K activity. An exponential decrease of the relative ^{43}K activity was observed with depth

[2]L. C. Brown and J. K. Poggenburg, "Potassium-43," in *Radioisotope Program (8000) Progress Report for June 1973*, ORNL-TM-4309, Oak Ridge National Laboratory.
[3]J. A. Harvey, Oak Ridge National Laboratory, private communication.

(note: the marble slabs were large enough, and the area of ^{43}K production localized enough, to disregard any possible losses perpendicular to the beam axis), and a "half-thickness" value (i.e., the thickness within which the ^{43}K yield decreased by a factor of two) of ∿12.3 cm was determined for this target material ($CaCO_3$ with a density of ≈2.7 g/cm^3). The absolute amount of ^{43}K produced and the saturation production rate will be determined next period when an accurate counting efficiency can be determined for the counting geometry employed.

Due to the problems associated with the spectrometer switching magnet and the corroboration of the narrow photon beam dimensions gleaned from this experiment, future effort will be directed toward low power (1 to 2 kW) irradiations in the direct beam position that will be used ultimately for the large-scale ^{43}K production. The next set of experiments are presently in the formative design stage, and it is anticipated that they will be finalized and fabrication started during the next reporting period.

Rare-Earth Radionuclides *(L. C. Brown)*

> *Ytterbium-169 has been found to exhibit high tumor-to-nontumor uptake ratios, comparable to the uptake ratios of ^{67}Ga. A cooperative project has begun with the Medical Division of ORAU to determine whether all the rare earths show this tumor localization, or whether it is limited to either the light (La-Gd) or heavy (Tb-Lu) members of this series, and to evaluate the most promising radionuclides for tumor localization in humans.*

A number of heavy mass rare-earth radionuclides, on both the neutron-excess and neutron-deficient side of the line of stability, have been identified as suitable for human use. The first neutron-deficient radionuclide to be investigated is ^{167}Tm (9.25 d) which decays *via* electron capture with a single predominate gamma ray of suitable characteristics (208 keV, 43% photon intensity) for efficient *in vivo* detection. Initial attempts[4] to prepare this radionuclide *via* the $^{167}Er(p,n)^{167}Tm$ reaction path resulted in concomitant production of large amounts of short-lived, high radiation dose ^{166}Tm (7.7 hr) — e.g., the end of bombardment ^{166}Tm: ^{167}Tm activity ratio was approximately 50:1. In order to avoid the necessity of using shielded "hot-cells" for chemical processing and to minimize the enriched Er_2O_3 target size [e.g., the (p,n) target configuration requires ∿900 mg of enriched target material versus only ∿165 mg for the (p,2n) target configuration], investigation of the $^{168}Er(p,2n)^{167}Tm$ reaction path was initiated.

Two (p,2n) production path irradiations have been performed to date utilizing the internal beam window target assembly described elsewhere.[5] Thick target production rates of ∿60 mCi/mA-hr (machine yield ≈5.5 mCi/hr)

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[4]L. C. Brown, "Rare-Earth Radionuclides," in *Radioisotope Program (8000) Progress Report for May 1973*, ORNL-TM-4281, Oak Ridge National Laboratory, p. 6.

[5]J. J. Pinajian, "ORNL 86-Inch Cyclotron," Chapter 9 in *Radioactive Pharmaceuticals*, CONF 65111, p. 148.

have been achieved with 22-MeV protons and this target configuration. These machine yields are comparable to those obtained for the (p,n) target configuration based on the "effective" ^{167}Tm yield after allowing for decay of the short-lived ^{166}Tm. A single pass, high-pressure ion-exchange (HPIE) separation has been developed to handle the 160- to 170-mg $^{168}Er_2O_3$ targets required for the window target. The 900-mg targets required for the (p,n) capsule target[5] required multiple HPIE passes — usually three — to separate the carrier-free ^{167}Tm from the target material. In order to follow the chromatographic behavior of the erbium target material, which forms no readily detectable erbium radionuclides during proton bombardment, 10- to 20-mg portions of the original enriched target material were neutron irradiated in the pneumatic tube of the Oak Ridge Research Reactor and added to the proton-irradiated target before starting the HPIE separation. Erbium-171 (7.52 hr) is produced from the unseparated ^{170}Er remaining in the target material, ∿0.4 atom %. In this way, elution profiles of both thulium and erbium can be obtained without adulterating the enriched ^{168}Er with undesirable higher mass ^{170}Er which would yield unwanted ^{170}Tm (129 d) upon re-irradiation during future production cycles.

Further work will be done on production parameter optimization, on HPIE separation development, and on product characterization and quality control.

Cyclotron Products Pilot Production *(M. R. Skidmore)* (Production and Inventory Accounts)

July 1973 ORNL 86-Inch Cyclotron runs for ORNL and non-ORNL programs are given in Table 1.

Table 1. Cyclotron Irradiations and Runs for July 1973

Date	Customer	Product	Target	Total Time (hr:min)	Total Charges
		ORNL Programs			
6-21-73	ORAU and Others	Gallium-67	Zinc-68	4:10	$ 403
6-29-73	ORAU and Others	Gallium-67	Zinc-68	9:00	862
6-30-73	Isotopes Division	Potassium-43	Calcium-44	2:00	202
7-3-73	Isotopes Division	Thulium-167	Erbium-168	2:00	202
7-13-73	ORAU and Others	Gallium-67	Zinc-68	9:00	862
7-16-73	Isotopes Division	Thulium-167	Erbium-168	2:15	226
				28:25	$2757
		Non-ORNL Programs			
7-19-73	International Chemical & Nuclear Corporation	Cobalt-57	Nickel-58	49:15	$8185

FISSION PRODUCTS

Cesium-137 Pilot Production *(R. W. Schaich)*
(Production and Inventory Accounts)

1. Process Status

The ^{137}Cs process equipment is in stand-by condition.

2. Operational Summary

	July 1973		CY 1973		FY 1974	
	No.	Ci	No.	Ci	No.	Ci
Sources						
Fabricated	0	0	10	22,800	0	0
Shipped	0	0	10	22,800	0	0
Special Form Cans						
Fabricated	2	800	45	26,000	2	800
Shipped	0	0	40	23,100	0	0

$^{137}CsCl$ product inventory — 574,000 Ci.

3. Current Orders

Current orders for ^{137}Cs as sources or bulk powder are as follows:

Customer	Amount (Ci)	Estimated Shipping Date
J. L. Shepherd & Associates	7,614	September 1973
White Sands Missile Base	14,221	September 1973
Minnesota Mining & Manufacturing Co.	1,300	August 1973

Strontium-90 Pilot Production *(R. W. Schaich)*
(Production and Inventory Accounts)

1. Process Status

Two strontium titanate pellets containing 46,700 Ci of radioactivity and emitting 320 W were hot-pressed into a cylindrical shape.

The large furnace was pulled from cell 11 into a 4-in. steel shield which was then sent to the Burial Ground for disposal. Decontamination of cell 11 is continuing.

The current ^{90}Sr process is shown below and is in agreement with the annual physical inventory of May 1, 1973.

Item	Strontium-90 (Ci)
In-process material	766,800
Strontium-90 products	383,700[a]
Sources in fabrication	46,700
Returned sources	435,200
Completed sources awaiting shipment	214,400
	1,846,800

[a]Includes 30,500 Ci of $^{90}SrSiO_3$.

2. Operational Summary

Item	July 1973		CY 1973		FY 1974	
	No.	Ci	No.	Ci	No.	Ci
HAPO shipments received	0	0	0	0	0	0
Product batches prepared	0	0	0	0	0	0
Sources						
Fabricated	0	0	0	0	0	0
Shipped	0	0	0	0	0	0
Special Form Cans						
Fabricated	0	0	71	1700	0	0
Shipped	0	0	72	1700	0	0

3. Current Orders

Current orders for ^{90}Sr as sources or bulk powder are as follows:

Customer	Amount (Ci)	Estimated Shipping Date
U. S. Navy	221,000	a
New England Nuclear Corporation	9	When requested
Messerschmitt-Bolkow GmbH	46,000	August 1973

[a]Sources are complete and customer has been billed. Shipment awaiting receipt of DOT permit.

Short-Lived Fission Pilot Production (Production and Inventory Accounts) *(R. W. Schaich)*

Isotope	Number of Batches	Amount (Ci)
Xenon-133	2	500
Iodine-131	1	31
Barium/Lanthanum-140	1	89
Strontium-89	1	41

Promethium-147 Shipments and Current Orders

Current orders for ^{147}Pm as sources or bulk powder are as follows:

Customer	Amount (Ci)	Estimated Shipping Date
W. Maier KG, Germany	300	August and September 1973
3M Company	5400	August 1973

SOURCE DEVELOPMENT

Gadolinium-153 *(F. N. Case)*

Separation of $^{152-153}Gd$ from sections of the HFIR control rods was completed and 6.5 g of ^{152}Gd was recovered. This should provide target for the production of ∿200 Ci of ^{153}Gd after irradiation in the HFIR.

Four sources were prepared for the medical cooperative programs.

A 1.5-Ci and a 0.2-Ci source were prepared for the University of Wisconsin for use in bone mineralization studies.

A 2-Ci source for Vanderbilt University for use in secondary x-ray scanning of stable gallium in the brain and a 500-mCi source for the Franklin McLean Memorial Research Laboratory for use in the detection of lead in tissue and blood were prepared.

The 2-Ci source sent to the University of California, Laboratory of Nuclear Medicine, earlier this year has been reported to be less intense than desired, and a 10-Ci source will be prepared as a replacement for their tomographic brain-scanning research.

Twenty curies of ^{153}Gd is now being purified for the fabrication of a source for the Instrumentation and Controls Division's position sensitive detector research. This source is expected to be delivered during August.

RADIOISOTOPE SALES

J. E. Ratledge

An order was received from Aktiebolaget Atomenergi, Sweden, for ∿294 Ci of ^{133}Xe to be shipped ∿25 Ci weekly beginning July 13 through September. An order was received from J. L. Shepherd and Associates for 7614 Ci of ^{137}Cs to be encapsulated into seven sources. An order was received from 3M Company for 1300 Ci of ^{137}Cs as bulk powder. A request was received from Leybold-Heralus GmbH and Company, Germany, for information concerning the ^{137}Cs to be available from Hanford Waste Encapsulation Plant.

Shipments made during the month that may be of interest are listed below:

Customer	Isotope	Amount
Large Quantities		
Western Electric Company	Krypton-85	160 Ci
Radiochemical Centre, England	Krypton-85	100 Ci
New England Nuclear Corporation	Tritium	8000 Ci
Withdrawn Items		
University of Chicago	Copper-67	7 mCi
Mayo Clinic	Copper-67	10 mCi
George Washington University	Copper-67	2 mCi
University of Pittsburgh	Iodine-131	50 mCi
University of Rochester	Iodine-131	50 mCi
Cleveland Metropolitan General Hospital	Iodine-131	50 mCi
ORNL, Biology Division	Chromium-51	25 mCi
Items Used in Cooperative Programs		
Johns Hopkins University	Bismuth-206	1 mCi
Oak Ridge Associated Universities	Bismuth-206	2 mCi
Oak Ridge Associated Universities	Gallium-67	520 mCi
Oak Ridge Associated Universities	Thulium-167	6 mCi
Vanderbilt University	Bismuth-206	10 mCi
Unusual Items		
Donald W. Douglas Laboratories	Promethium-147	∿1200 Ci[a]
Nagoya University, Japan	$^{154}Sm_2O_3$	1 sample[b]

[a]1.139 g ^{147}Pm oxide that had been calutron separated to remove ^{146}Pm.
[b]For Vanderbilt University; irradiated in HFIR.

The radioisotope sales proceeds and shipments for FY 1972 and FY 1973 are given in Table 2.

Table 2. Radioisotope Sales and Shipments

Item	7-1-71 thru 6-30-72	7-1-72 thru 6-30-73
Inventory items	$ 507,745	$ 548,166
Major products	65,133	58,796
Radioisotope services	309,026	256,604
Cyclotron irradiations	143,230	104,667
Miscellaneous processed materials	62,225	60,143
Packing and shipping	72,280	67,852
Total	$1,159,639	$1,096,228
Number of shipments	2,468	2,120

A summary of radioisotopes shipped during July 1973 is given in Table 3.

Table 3. Summary of Radioisotopes Shipped During July 1973

Isotope	Number of Shipments	Quantity (mCi)
Barium-140	1	1
Calcium-47	7	5
Cerium-144	3	120
Cesium-137	10	30,147,000
Copper-67	4	14
Gallium-67	13	3,176
Iodine-129	4	2,700 mg
Iodine-131	5	300
Krypton-85, normal	26	1,039,700
Niobium-95	5	9
Phosphorus-33	1	100
Potassium-43	3	11
Promethium-147	2	5,050
Ruthenium-103	1	5
Strontium-89	3	17
Strontium-90	7	222,642,700
Technetium-99	3	7,712
Tritium	18	19,470,013
Xenon-133	53	133,850
Zirconium-niobium-95	2	10
Miscellaneous preparations and targets	5	14
Cyclotron irradiations: Pacific Northwest Labs	1[a]	1
Totals	177	273,442,096 10,412 mg

[a]Promethium-143.

ADMINISTRATIVE

J. K. Poggenburg made a trip to the National Heart and Lung Institute in Washington, D. C., to discuss ^{43}K program.

PUBLICATIONS

REPORTS

E. Lamb, *Isotopic Power Fuels Monthly Status Report for June 1973*, ORNL-TM-4310, Oak Ridge National Laboratory.

A. F. Rupp, *Radioisotope Program (8000) Progress Report for June 1973*, ORNL-TM-4309, Oak Ridge National Laboratory.

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