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RADIOISOTOPE PROGRAM (8000) PROGRESS REPORT FOR MARCH 1973

A. F. Rupp

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ISOTOPES DEVELOPMENT CENTER

RADIOISOTOPE PROGRAM (8000) PROGRESS REPORT FOR MARCH 1973

A. F. Rupp

Work Sponsored by
AEC Division of Applied Technology

APRIL 1973

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RADIOISOTOPE PROGRAM (8000) PROGRESS REPORT FOR MARCH 1973

A. F. Rupp

RADIOISOTOPE PRODUCTION AND MATERIALS DEVELOPMENT

REACTOR-PRODUCED ISOTOPES — 08-01-01

Phosphorus-33 *(J. K. Poggenburg)*

The purpose of this project is to develop methods of preparing hundred-millicurie quantities of carrier-free phosphorus-33 containing ≤5% phosphorus-32. The two methods for producing phosphorus-33 being evaluated are based on the irradiation of highly enriched targets of sulfur-33 (>92 at. %) or chlorine-36 (approximately 63 at. %) in a fast neutron flux. Phosphorus-33 (25.2 days; 0.248-MeV β^-_{max}) has both a longer half-life and a lower energy beta than phosphorus-32 (14.3 days; 1.709-MeV β^-_{max}), which makes it advantageous for autoradiography, longer ecological and agricultural experiments than with phosphorus-32, synthesis of tagged complex organophosphorus compounds, and double labeling experiments.

We have received an order for 1.4 Ci of phosphorus-33 to be delivered over a three-month period. The lack of advance warning makes the target management a problem, but the application in fertilizer studies does not require <5% phosphorus-32, and it appears we can meet the orders under the relaxed phosphorus-32 specifications. The first shipment of 500 mCi was made at the end of March and was comprised of the phosphorus-33 recovered from the $K_2{}^{33}SO_4$ target last month plus that recovered from about 8.2 g of lower enrichment (∿48%) $K^{36}Cl$.

A yield of 1.2 Ci of phosphorus-33 (at discharge) was obtained from 1.0 g ∿65% $K^{36}Cl$ irradiated three cycles. It was processed about one month later and provided >500 mCi phosphorus-33 with <5% phosphorus-32, which is being used to fill orders for 300 mCi of specification quality phosphorus-33. This target was processed by adding dissolved $Fe_2(SO_4)_3$ to the aqueous solution of $K^{36}Cl$ target and making an expeditious $Fe(OH)_3$ precipitation with NH_4OH. The recovery of $K^{36}Cl$ then requires removal of $SO_4{}^{2-}$ ion by precipitation as $BaSO_4$, using $Ba(OH)_2$ reagent.

Among the $K^{36}Cl$ targets processed was one irradiated six months in the Control Rod Access Position (CRAP). It contained three ampuls of 44% $K^{36}Cl$. The center ampul was processed separately and the end ones together. While the yield of phosphorus-33 from these targets was small (∿150 mCi), a mass analysis indicated enrichment of the center target

to 55% chlorine-36 and the end targets to 50% during the irradiation. The HFIR flux spectra calculations do suggest that, while the fast flux is less in CRAP targets than Removable Beryllium (RB) targets, the thermal flux is generally higher in the CRAP targets. These positions are therefore most suited for the long-term production of chlorine-36 by thermal neutron capture.

Potassium-43 *(J. K. Poggenburg)*

> *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 and to establish cooperative programs with medical institutions to evaluate it. Potassium-43 is potentially useful for metabolic and clinical studies of blood flow, rejection of transplanted organs, and kidney function. The half-life of potassium-43 is 22.5 hr; gamma-rays are 0.373 and 0.617 MeV.*

Three batches of potassium-43 were processed this month, and seven shipments totaling 26 mCi were made.

The ORELA has been down for repair of a broken water line to the building so the photoproduction experiment was not accomplished.

Platinum-195m *(J. K. Poggenburg)*

> *Recent medical research has shown that some platinum compounds act as chemical therapeutic agents towards certain kinds of tumors. Platinum radioisotopes are needed for investigation of the therapeutic mechanism involved. Platinum-195m (4.0 d) has gamma emissions of 99 keV (11%) and 129 keV (2.8%) that make it suitable for whole-body scanning.*

Thirty-eight mCi of platinum-195m was shipped to University of Southern California under our medical cooperative program.

A 9.9-mg sample of 95.95% enriched platinum-194 was irradiated for nine days in the HFIR hydraulic tube. At discharge the main activities were:

	mCi/mg
Platinum-195m	0.96
Platinum-197	0.86
Gold-199	0.75
Copper-64	0.24

The flux monitor and trace activity analyses are in progress.

Rare-Earth Radionuclides *(J. K. Poggenburg)*

Ytterbium-169 has been found to exhibit high tumor-to-nontumor uptake ratios, comparable to the uptake ratios of gallium-67. 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.

Samarium-153, erbium-171, and neodymium-147 were selected as nuclides suitable for preliminary localization studies. A 99.18% enriched $^{152}Sm_2O_3$ target was irradiated 29 hr in the HFIR hydraulic tube to produce samarium-153 with a specific activity of 4.63 Ci/mg and >99.9% radionuclidic purity. This 46.8-hr nuclide has one principal gamma emission of 103 keV in 28.4% abundance.

A 96.06% enriched $^{170}Er_2O_3$ target was irradiated 7.9 hr in the HFIR hydraulic tube to produce a product with a specific activity of 0.76 Ci/mg at discharge with a radionuclidic purity greater than 99%. This 7.5-hr nuclide emits gammas of energy 296 keV and 308 keV, with abundances of 28% and 63%, respectively.

Distribution and imaging studies are in progress at ORAU.

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

Processed Units	
Radioisotope	Amount (mCi)
Calcium-47	23.79
Copper-67	21.6
Zinc-69m	2.3

ACCELERATOR-PRODUCED ISOTOPES — 08-01-02

Bismuth-206 *(L. C. Brown)*

The objectives of this program are to evaluate the pertinent production parameters and to provide bismuth-206 (6.24 d) for evaluation as a diagnostic radionuclide in medicine. Bismuth-206 has been used to delineate brain tumors in patients with known brain tumors which are poorly visualized by more common radionuclides. Even though very favorable tumor-to-background activity ratio has been obtained by earlier workers, the lack of appropriate scanning

systems suitable for the high-energy emissions of this nuclide has retarded the evaluation of the isotope. Cooperative programs have been established for the evaluation of bismuth-206 as a diagnostic radionuclide using high-energy photon scanning systems.

Development work on a high-yield, electrodeposited lead-on-copper target design has been extended to the test irradiations of enriched lead-207 targets (92.4 atom % lead-207) in order to optimize irradiation conditions and target integrity and to provide high-purity bismuth-206 to medical collaborators for evaluation. A 0.005-in.-thick lead-207 target was irradiated with a 22-MeV proton beam at an average current of 150 μA without failure. No visible signs of irradiation damage, either to the outer surface or at the lead-copper interface were observed. The measured cyclotron yield of ∿710 mCi/mA-hr for this target configuration was within 10% of the value estimated from the $^{nat}Pb(p,2n)^{205}Bi$ data reported earlier.[1] Unacceptably high losses, ∿25%, of carrier-free bismuth-206 were encountered when 3 *M* HCl was used to back-extract the carrier-free bismuth-206 from the pregnant organic during processing. Tracer level experiments will be undertaken to determine how the chemical separations process can be effectively altered to minimize these losses. Development of a lead target recovery procedure, compatible with the spent electrolyte and aqueous process raffinate solutions generated during the production cycle, and suitable for efficient target material recycle, has been studied. Two different chemical systems have been investigated, but additional development work must be performed before reporting an optimal procedure. Additional enriched lead-207 target optimization irraditions will be performed during the next period. This work could be done using natural lead targets, but enriched targets will be used to provide high-purity bismuth-206 to medical cooperative participants so the evaluation of this radionuclide with humans can begin.

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 gallium-67. 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, and their production development will be undertaken in the initial stages of this project. The neutron-deficient isotopes

[1] L. C. Brown, "Bismuth-206," pp. 3-4 in *Radioisotope Program (8000) Progress Report for February 1973*, ORNL-TM-4161, Oak Ridge National Laboratory.

of interest are terbium-155 (5.4 d), dysprosium-157 (8.1 h), and thulium-167 (9.3 d). The recent availability of 99.5+% isotopically enriched dysprosium-156 target material will allow the reactor development of high-purity dysprosium-157 suitable for human use, which will be reported under the 08-01-01 project category. Terbium-155 and thulium-167 will be prepared via the (p,n) reaction path by 15-MeV proton bombardment of highly enriched (Z-1,A) target material.

A new, improved design, high-pressure ion-exchange chromatographic apparatus has been fabricated during this reporting period for use in the rapid separation of adjacent rare-earth elements. The design is similar to the apparatus employed for the final purification of gadolinium-153 and has been designed for both in-cell operation and servicing. This equipment will be tested initially on a Eu-Sm-Pm separation of 265-day promethium-143 for Battelle Pacific Northwest Laboratories. A cooperative arrangement has been undertaken with Battelle to prepare high-purity promethium-143 via the $^{144}Sm(p,2n)^{143}Eu \xrightarrow{\beta^+,\ EC} {}^{143}Sm \xrightarrow{\beta^+,\ EC} {}^{143}Pm$ production path as reported earlier.[2] This radionuclide will be used for excretion experiments to better evaluate the medical hazard that promethium might present. Since normal human volunteers are used, highest radionuclidic purity and minimum radiation dose are essential and the extra cost associated with this production path is warranted.

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

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

Table 1. Cyclotron Irradiations and Runs for March 1973

Date	Customer	Product	Target	Total Time (hr:min)	Total Charges
		ORNL Programs			
3-1-73	ORAU and Others	Gallium-67	Zinc-68	9:00	$ 862
3-6-73	Isotopes Sales	Cobalt-57	Nickel	42:45	4179
3-8-73	ORAU and Others	Gallium-67	Zinc-68	9:25	901
3-12-73	Isotopes Division	Bismuth-206	Lead-207	2:15	221
3-14-73	Work for others	Promethium-143	Samarium-144	25:15	2412
3-16-73	ORAU and Others	Gallium-67	Zinc-68	4:50	466
			TOTAL	93:30	$9041

In addition to routine maintenance, the dees were removed and cracks which had developed were welded. Current leakage from the radiofrequency field impressed on the dees to the liner had concentrated in some areas resulting in holes being burned through the liner wall. These were also repaired.

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[2] A. F. Rupp, *Radioisotopes Program (8000) Progress Report for March 1970*, ORNL-TM-2965, Oak Ridge National Laboratory, p. 9.

FISSION PRODUCTS — 08-01-03

Krypton-85 Enrichment

Three column bundles have been removed from Cell 1 and are in the shop being cleaned and the tubes removed for inspection. Purchase orders have been placed for additional calrods and Inconel tubing to rebuild all six columns and three spares. These materials are scheduled for delivery in May.

Specifications are being prepared for the purchase of a 130-ton water chiller unit to permit a closed loop cooling system for the columns when they are re-installed.

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

1. Process Status

One oxalate precipitation cycle using NH_4^+ carrier was run and yielded 25,000 Ci. This material along with the 50,000 Ci processed in February was converted to $^{137}CsCl$. Final assay and storage should be completed by April 1. All cesium-137 processing is now complete. A residual batch of 40,000 Ci resulting from final recycling, tank rinses, etc., remains in the process system. This solution is being held temporarily in the crystallizer system until a decision is made on its final disposition. The current cesium-137 process status is as follows:

Item	Cesium-137 (Ci)
In-process material	40,000
$^{137}CsCl$ products	554,000
Sources in fabrication	0
Completed sources awaiting shipment	190,600

2. Operational Summary

Item	March 1973		CY 1973		FY 1973	
	No.	Ci	No.	Ci	No.	Ci
HAPO shipments received	0	0	0	0	0	0
Product batches prepared	9	75,000	17	225,000	47	532,000
Sources						
Fabricated	2	20	10	22,800	36	48,500
Shipped	2	20	9	14,700	35	40,400
Special Form Cans						
Fabricated	0	0	41	5,200	51	5,700
Shipped	1	50	7	1,000	10	2,400

3. Current Orders

Current orders for cesium-137 as sources or bulk powder are listed below:

Customer	Amount (Ci)	Estimated Shipping Date
J. L. Shepherd & Associates, Inc.	10,000	March & May 1973
Radiochemical Centre	1,500	May 1973

An order is on hand from AECL for ∿63 kCi of cesium-137 as CsCl powder. The powder has been canned and stored and is awaiting release by customer.

4. Source Fabrication

Fifty curies of cesium-137 powder was shipped to Brookhaven National Laboratory.

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

1. Process Status

All strontium-90 source-grade material was stored in weldable cans and removed from the FPDL cells. These cans will be welded for long-term storage. The current strontium-90 process status is as follows:

Item	Strontium-90 (Ci)
In-process material	805,900
Strontium-90 products	426,200
Sources in fabrication	0
Returned SNAP sources	446,600
Completed sources awaiting shipment	221,000

2. Current Orders

Current orders for strontium-90 as sources or bulk powder are as follows:

Customer	Amount (Ci)	Estimated Shipping Date
U. S. Navy	221,000	FY 1973
Radiochemical Centre	1,000	May 1973

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

Isotope	Number of Batches	Amount (Ci)
Xenon-133	2	700
Iodine-131	1	34
Cerium-144	1	168

Promethium-147 Shipments and Current Orders

Donald W. Douglas has placed an order for 200,000 Ci of promethium-147. Shipments of 50,000 Ci will be made in April, July, October, and December 1973.

SOURCE DEVELOPMENT — 08-01-04

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

Three grams of gadolinium-152 was recovered from the HFIR europium control plates using solvent extraction for separation of europium. This synthetic gadolinium-152 will be used as target material for HFIR irradiations to produce gadolinium-153.

The ion chamber used to monitor column effluents was redesigned to permit the chamber to be placed outside the process hot cell. This change was made to reduce radiation background and to provide better control of the column operation.

A 2-Ci source of gadolinium-153 containing <10 ppm total radiochemical impurities was sent to the University of California at San Francisco as a part of our cooperative program with this institution. This project deals with the clearance of heavy metals, such as tantalum, cerium, and barium, from the lungs, and a study of ways to modify the mechanisms involved. It has application to the study of basic physiology, the investigation of pollution-related problems, and chronic lung disease. It is expected that the presence of heavy metals in lungs can be detected and their concentration determined by transmission radiography and by detection of their excited characteristic x rays.

A series of high-pressure ion-exchange runs is in progress to purify 20 Ci of gadolinium-153 for fabrication of a single source to be used in single-wire position-sensitive detector development at ORNL.

Two sources are being prepared for our cooperative program with the U. S. Public Health Service, HEW, Los Angeles, to be used for studies of bone mineral content.

Americium-241 *(F. N. Case)*

The americium-241 sources, prepared for Vanderbilt University Medical Center for use in a thyroid scanner that makes use of the fluorescent x ray from stable iodine (reported last month), were tested and gave a threefold increase of signal-to-noise ratio when compared to the annular source previously used. This work was reported at a nuclear medicine meeting in San Francisco in March.

Radioisotope Characterization, Quality Control, and Standards *(S. A. Reynolds)*

1. Radioisotope Characterization

A report[3] on specific activities and other characteristics of over 170 common radionuclides was completed.

In a review[4] on "Perturbation of Nuclear Decay Rates," G. T. Emery states that beta decay rates should be little affected by chemical environment and cites our recent findings on iodine-131 and copper-64.

The draft of a paper on the half-life of cesium-137 was reviewed, and the indicated value was 30.21 ± 0.08 years, in excellent agreement with our 30.18 years and the 30.17 years of Dietz (KAPL).

2. Radioisotope Special Analysis and Quality Control

A review[5] of more than a quarter-century of radioisotope measurement methodology was prepared. We have been considering preparation of "weightless" planchet sources of carbon-14, to be used as reference sources for counters and survey instruments, for example. A survey of needs and potential suppliers was initiated some time ago. Seventy-four organizations were contacted by questionnaire, and 39 responded. Twenty-six checked "not interested." Nine indicated interest in sources somewhat like those proposed, and their estimated annual needs totaled 32. Four companies reported willingness to supply such sources. It was accordingly decided not to pursue the matter further at ORNL.

An argon-37 standard from NBS was measured by internal proportional counter. Assuming 100% efficiency for counting the radiations (mostly Auger electrons), our measurement was 3.5% below that of NBS, which was certified to ±4.3%.

Inquiries were received concerning analysis of samples from multiple-tracer experiments, determination of plutonium in environmental samples, and possible causes for erratic results in analysis of traces of cesium-137 in water.

RADIOISOTOPE SALES

(J. E. Ratledge)

Requests for quotations have been received from the following: Atomic Energy of Canada, Limited, for six cesium-137 sources (each containing 8500 Ci and using their cesium-137 presently in storage at ORNL) and

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[3]S. A. Reynolds, *Theoretical and Practical Specific Activities and Other Properties of Common Radionuclides*, ORNL-TM-4167, Oak Ridge National Laboratory, 1973.
[4]G. T. Emery, *Ann. Rev. Nucl. Sci.* 22, 165 (1972).
[5]S. A. Reynolds, *Some Radioisotope Assay Methods*, ORNL-4788, Oak Ridge National Laboratory (1973).

for 50, 100, and 200 kCi of cesium-137 as bulk powder to be stored at ORNL for fabrication into sources as requested; CEA, France, for 2 MCi cesium-137 as cesium chloride bulk powder and for 1.5 MCi strontium-90 as titanate bulk powder; AERE, England, for 640 sources, each containing 10 kCi of strontium-90 as titanate, to be shipped beginning February 1974 and ending March 1977; and Gamma Industries, Inc., for molecular sieve pellets impregnated with cesium-137.

Orders were received from the following: University of California, LLL, for a 1000-Ci cobalt-60 source having specific activity of ∿1000 Ci/g; IAEA, Austria, for 500 and 400 mCi of phosphorus-33 to be shipped in March and April 1973, respectively; and Messerschmitt-Bolkow-Blohm, Germany, for a 315-W (∿46 kCi) strontium-90 source.

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

Customer	Isotope	Quantity
Large Quantities		
New England Nuclear Corporation	Tritium	8,000 Ci
Radium-Chemi A. Zeller & Company	Tritium	10,000 Ci
Lovelace Foundation	Cerium-144	20 Ci
Radiochemical Center	Krypton-85	100 Ci
J. L. Shepherd & Associates	Cesium-137	11,754 Ci
3M Company	Promethium-147	5,001 Ci
Withdrawn Items		
Gamma Industries, Inc.	Cesium-137	9 Ci[a]
Items Used in Cooperative Programs		
Oak Ridge Associated Universities	Gallium-67	∿520 mCi
Oak Ridge Associated Universities	Erbium-171	∿60 mCi
Vanderbilt University	Bismuth-206	∿17 mCi
Vanderbilt University	Americium-241	16 Ci
University of California, San Francisco	Gadolinium-153 source	2 Ci
University of Southern California	Platinum-195m	∿30 mCi
Oak Ridge Associated Universities	Samarium-153	∿50 mCi
Union Carbide Corporation, Y-12 Plant	Samarium-153	∿10 mCi

[a]Impregnated in glass cones.

A summary of radioisotopes shipped during February 1973 is given in Table 2.

Table 2. Summary of Radioisotopes Shipped During February 1973

Isotope	Number of Shipments	Quantity (mCi)
Barium-140	2	20
Calcium-47	9	6
Cesium-137	65	706,776
Copper-67	3	21
Gadolinium-153	2	600
Gallium-67	11	457
Hafnium-181	1	10
Iodine-131	1	200
Krypton-85	12	316,000
Krypton-85, enriched	1	500
Niobium-95	2	11
Phosphorus-33	2	190
Potassium-43	5	20
Promethium-147	2	676,800
Ruthenium-103	2	6
Ruthenium-106	1	2
Samarium-151	3	1,080 mg
Strontium-89	4	16
Tritium	19	21,556,000
Xenon-133	51	51,020
Yttrium-91	2	24
Zirconium-95	4	46
Misc. compound preparations and targets	8	818
Cyclotron irradiations:		
New England Nuclear Corporation	Cobalt-57 as target	∿2,000
Philips-Duphar, Netherlands	Cobalt-57 as solution	321
TOTALS	214	23,311,864 1,080 mg

The radioisotopes sales proceeds and shipments for the first eight months of FY 1972 and FY 1973 are given in Table 3.

Table 3. Radioisotopes Sales and Shipments

Item	7-1-71 thru 2-29-72	7-1-72 thru 2-28-73
Inventory items	$293,280	$317,736
Major products	42,435	37,646
ORNL 86-Inch Cyclotron	88,711	84,450
Radioisotope services	205,386	120,608
Miscellaneous processed materials	41,340	34,323
Packing and shipping	48,285	45,893
Total	$719,437	$640,656
Number of shipments	1,648	1,428

ADMINISTRATIVE

Visitors to the IDC are listed in Table 4.

Table 4. Visitors to IDC

Visitor	Affiliation	Purpose of visit
Milton Lewis	Donald W. Douglas	Discuss promethium-147 separation
A. B. Brill R. L. Hayes J. E. Machurek W.W.H. Weyzen	Vanderbilt University ORAU AEC-DAT/ID AEC-DBER	Discuss proposed medical radioisotope program
Carl W. Seidel Paul Tyree Richard Garniewicz	New England Nuclear Corporation	Discuss radioactive source encapsulation, cyclotron target fabrication and irradiation, and enriched stable isotope production
B. N. Bhot	NASA, Marshall Space Flight Center	Discuss target production

PUBLICATIONS

REPORTS

T. A. Butler and J. K. Poggenburg (Compilers), *Technical Feasibility of Using Thulium-170 Oxide for Aerospace Radioisotope Heat Sources*, Lynda Kern and Robert H. Lafferty, Jr. (eds.), ORNL-CF-73-2-12, Oak Ridge National Laboratory, Feb. 9, 1973.

E. Lamb, *Isotopic Power Fuels Monthly Status Report for February 1973*, ORNL-TM-4162, Oak Ridge National Laboratory.

E. Lamb, *ORNL Isotopic Power Fuels Quarterly Report for Period Ending December 31, 1972*, ORNL-4857, Oak Ridge National Laboratory.

A. F. Rupp, *Radioisotope Program (8000) Progress Report for February 1973*, ORNL-TM-4161, Oak Ridge National Laboratory.

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