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RADIOISOTOPE PROGRAM (8000)
PROGRESS REPORT FOR APRIL 1973

A. F. Rupp

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ORNL-TM-4229

Contract No. W-7405-eng-26

ISOTOPES DEVELOPMENT CENTER

RADIOISOTOPE PROGRAM (8000) PROGRESS REPORT FOR APRIL 1973

A. F. Rupp

Work Sponsored by
AEC Division of Applied Technology

MAY 1973

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RADIOISOTOPE PROGRAM (8000) PROGRESS REPORT FOR APRIL 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.

An RB target containing 3.48 g of low enrichment (∿48%) $K^{36}Cl$ was processed after a 4-cycle irradiation. All three ampuls were found broken upon opening. At time of processing, the yield was 0.8 Ci phosphorus-33. This product had 15% phosphorus-32 content but was used to fill an order for 400 mCi for fertilizer applications. Two 100-mCi orders were also shipped from material processed last month.

A great deal of effort went into recovering various targets, and four target assemblies were prepared and inserted at the last shutdown; two of these assemblies are intended for long-term chlorine-36 enrichment.

Two other target assemblies were opened and are being processed. One had all three enclosed quartz ampuls broken; but the pressed plugs of aluminum foil used to separate them appears to have kept most of the three targets separate. The product does have a higher phosphorus-32 content due in part to the irradiation for only two cycles and, probably, to partial mixing of the lower enrichment $K^{36}Cl$ from the end ampuls.

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 four shipments totaling 14 mCi were made.

The ORELA is again operating and an experiment using short pulses is scheduled the first week in May. The photoproduction experiment will be run at this time.

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.

Another target of 96.06% erbium-170 was irradiated in HFIR to supply erbium-171 under our medical cooperative with ORAU. It was irradiated for 20 hr, and we will determine how much erbium-172 is produced by double neutron capture.

A target of 97.46% neodymium-146 was irradiated 11.6 days in the HFIR hydraulic tube. At discharge the specific activity of neodymium-147 was 50 mCi per mg of neodymium. No chemical purification was made and the product also contained an undetermined amount of promethium-147 daughter activity and 2.5% promethium-148 activity from neutron capture by the daughter promethium-147. This material was also supplied to ORAU.

Iodine-123 *(J. K. Poggenburg)*

AEC requested that we examine the iodine-123 available from commercial suppliers. Orders for 1 mCi each were placed with two vendors. One was received the following day, while the second required several weeks. Analysis of the products was performed using computer resolution of gamma spectra taken with a germanium detector. After approximately four weeks' decay, a sample was counted on a calibrated silicon x-ray detector to determine iodine-125. The accuracy of the iodine-125 measurement is estimated at ±30% due to greater uncertainties in the detector calibration and the photon intensity of the 35-keV transition. The results of the analysis are presented in Table 1 with activities corrected to the vendor's calibration time.

Table 1. Analysis of Commercial Iodine-123

	Half-Life	Vendor A[a]		Vendor B[b]	
		mCi at Calibration	% of ^{123}I	mCi at Calibration	% of ^{123}I
Iodine-123	13.2 h	1.02	—	1.04	—
Iodine-124	4.17 d	0.0049	0.49	0.13	12.5
Iodine-125	60.14 d	0.00081	0.08	0.0031	0.30
Iodine-126	13.0 d	0.0027	0.27	0.00087	0.084
Iodine-130	12.4 h	0.016	1.6	—	—
Iodine-131	8.06 d	0.0026	0.26	—	—

[a]Method of preparation not stated.
[b]Prepared by $^{124}Te(p,2n)^{123}I$ on enriched target. Calibration time was appreciably long after end of bombardment, resulting in relatively high iodine-124.

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

Processed Units	
Radioisotope	Amount (mCi)
Zinc-69m	165.6
Calcium-47	29.02
Copper-67	60.72

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.

Test irradiation of the high-yield, electrodeposited lead-on-copper target design continued using enriched lead-207 targets (92.4 atom % lead-207). A target of 0.0058-in. average thickness lead-207 was irradiated with a 22-MeV proton beam at an average current of 200 μA without failure. No signs of irradiation damage were observed on the outer surface by visual examination, but upon dissolution a small abnormality was observed on the substrate surface in the region of the maximum beam energy deposition. It cannot be ascertained at present whether this was due to beam damage, intermetallic alloy formation at the Pb-Cu interface, or an imperfection in the copper substrate material. The measured cyclotron yield of 736 mCi/mA-hr for this target was approximately 15% lower than the value estimated from data obtained using natural lead targets. An autoradiogram of the target substrate after removal of the lead-207 target material showed the area of maximum beam concentration to be off-center approximately 1/4 to 3/8 in. Another irradiation at this power level will be performed on another target of approximately the same thickness in order to obtain additional experience regarding a possible breakdown of the lead-copper interface and to remeasure the cyclotron yield after the cyclotron beam has been realigned. Unacceptably high bismuth-206 losses, ∿25%, were encountered again during chemical processing. The loss of carrier-free bismuth-206 in the organic phase after back-extraction was reduced to ∿10% by reducing the organic/aqueous phase volume ratios, but an approximately 15% loss was encountered when an attempt was made to redissolve the carrier-free bismuth-206 final product — which had been evaporated to dryness from a dilute HNO_3 solution in a glass beaker — with 0.05 *M* HNO_3. Later attempts to recover the activity by leaching with more concentrated HNO_3 and HCl solutions removed very little additional activity. The bismuth-206 seemed to be irreversibly bound to the glass surface, possibly due to excessive "baking" of the beaker during the evaporation. Additional experience will be obtained regarding this phenomenon during future production runs. Development of a lead-207 target material recovery procedure has continued. Precipitation of $^{207}PbSO_4$ from both the spent HBF_4 electrolyte and the aqueous raffinate followed by metathesis to $^{207}PbCO_3$ with Na_2CO_3 appeared to work well during the initial inactive experiments. Traces of Cu^{2+}, introduced during target dissolution, are not precipitated by this sulfate system. The $^{207}PbCO_3$ is then reused directly for preparation of the new HBF_4 plating electrolyte. Traces of $^{207}PbSO_4$, which are not converted to the carbonate, can be easily separated from the plating electrolyte before use and recycled in the next recovery. Preliminary experiments indicate that >95% recovery yields can be anticipated.

An additional enriched lead-207 target optimization irradiation and process run will be performed during the next reporting period. A monthly development run schedule seems to harmonize well with the needs of the medical cooperative participants, now totaling three, who are doing the applications evaluation of this radionuclide.

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.

As part of a cooperative arrangement with Battelle Pacific Northwest Laboratories, a 200-mg sample of $^{144}Sm_2O_3$ which had been irradiated in the ORNL 86-Inch Cyclotron to prepare approximately 1 mCi of high-purity promethium-143 (265 d) via

$$^{144}Sm(p,2n)^{143}Eu \xrightarrow{\beta^+,\ EC} {}^{143}Sm \xrightarrow{\beta^+,\ EC} {}^{143}Pm$$

was separated using the new design high-pressure ion-exchange apparatus fabricated last month. This apparatus performed well and yielded a final promethium-143 product of >99.9% radionuclidic purity. Since the yield for this cyclotron irradiation was only approximately 35% of an earlier run,[1] an autoradiogram of the aluminum tube used to contain the target material in the window-type target holder[2] was obtained and indicated that the beam was off-center during the irradiation. As a result, only approximately 350 μCi (as of end-of-bombardment) of promethium-143 was prepared, and an additional production run will have to be scheduled to prepare the remainder of the desired 1 mCi.

Thulium-167 (9.3 d) will be prepared via the $^{167}Er(p,n)^{167}Tm$ reaction, separated, and then evaluated by the Medical Division of ORAU as a tumor-localizing diagnostic agent. In addition to the ^{143}Pm-^{145}Sm-^{146}Eu high-pressure ion-exchange separation, an ^{171}Er-^{171}Tm-^{175}Yb separation, using a total sample of approximately 20 mg rare-earth oxides, was performed to gain experience with the separation conditions required for a rapid and efficient Er-Tm separation. The complete ion-exchange separation of Er-Tm-Yb was performed in approximately 2 hr, with no erbium or ytterbium activities observed in the combined thulium fractions. A chemical yield of >98% was achieved. Future work will be directed towards scale-up of this separation to handle the 400-500 mg $^{167}Er_2O_3$ cyclotron targets that will be required. It is hoped that distribution of this radionuclide to ORAU can be started next reporting period.

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

April 1963 ORNL 86-Inch Cyclotron runs for ORNL and non-ORNL programs are given in Table 2.

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[1]A. F. Rupp, *Radioisotope Program (8000) Progress Report for March 1970*, ORNL-TM-2965, Oak Ridge National Laboratory, p. 9.
[2]J. J. Pinajian, Chapter 9 in *Radioactive Pharmaceuticals*, CONF-65111 (1966), p. 149.

Table 2. Cyclotron Irradiations and Runs for April 1973

Date	Customer	Product	Target	Total Time (hr:min)	Total Charges
		ORNL Programs			
3-23-73	ORAU and others	Gallium-67	Zinc-68	9:20	$ 892
3-26-73	Isotopes Division	Lead-203	Thallium	2:30	269
3-29-73	ORAU and others	Gallium-67	Zinc-68	9:30	910
4-5-73	ORAU and others	Gallium-67	Zinc-68	9:00	862
4-10-73	Isotopes Division	Bismuth-206	Lead-207	1:55	189
4-12-73	ORAU and others	Gallium-67	Zinc-68	5:10	498
4-19-73	ORAU and others	Gallium-67	Zinc-68	9:00	862
		Totals		46:25	$4482
		Non-ORNL Programs			
4-2-73	International Chemical	Cobalt-56	Iron	9:15	$1487
4-23-73	and Nuclear Corporation	Yttrium-88	Strontium	11:15	1660
		Totals		20:30	$3147

FISSION PRODUCTS — 08-01-03

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

1. Process Status

All in-process cesium-137 solution (∿40,000 Ci) was discarded, and several full-volume water rinses of the cesium-137 processing and storage tankage were made. Chemical decontamination of the cesium-137 system has not been done yet, but no significant amounts of in-process cesium-137 remain. The solid residues from the cesium-137 glass processing were discarded to land burial. All in-cell processing equipment which could be dismantled by manipulator operations was discarded to land burial. Preliminary washing of the cesium-137 manipulator cells was started. It is planned to do only remote cleaning in this system until access has been gained to the strontium-90 manipulator cells; then direct decontamination of the cesium-137 cells will be done while maintenance is in progress in the strontium-90 cells. The current cesium-137 process status is as follows:

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

[a]Approximate; these values will be adjusted based on results of annual physical inventory next month.

2. Operational Summary

Item	April 1973 No.	April 1973 Ci	CY 1973 No.	CY 1973 Ci	FY 1973 No.	FY 1973 Ci
HAPO shipments received	0	0	0	0	0	0
Product batches prepared	0	0	17	225,000	47	532,000
Sources						
Fabricated	0	0	10	22,800	36	48,500
Shipped	0	0	9	14,700	35	40,400
Special form cans						
Fabricated	0	0	41	5,200	51	5,700
Shipped	0	0	6	950	10	2,400

3. Current Orders

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

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

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

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

1. Process Status

A small (∿2000 Ci) batch of calcined $^{90}SrCO_3$ is being prepared to supply some miscellaneous orders. The annual physical inventory of strontium-90 in-process material should be complete by May 1. Decontamination of the strontium-90 final processing cell was started. This will be a full-scale decontamination for personnel entry. All fixed equipment except the precipitator will be removed. The cell will probably be decontaminated sufficiently for top-of-cell work during the first week in May. A final decontamination schedule is difficult to predict. This cell was last opened in 1967, and even then only the upper area of the cell was entered. The current strontium-90 process status is as follows:

Item	Strontium-90 (Ci)[a]
In-process material	806,000
Strontium-90 products	426,000
Sources in fabrication	0
Returned SNAP sources	446,000
Completed sources awaiting shipment	221,000

[a]Approximate; the values will be adjusted based on annual physical inventory results next month.

2. Operational Summary

No entries except one 10-Ci special form can in FY 1973.

3. 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	May 1973
Radiochemical Centre	1,500	May 1973
General Radioisotope Products	10	May 1973

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

Isotope	Number of Batches	Amount (Ci)
Xenon-133	2	400
Iodine-131	1	8

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 May, July, October, and December 1973. W. Maier K.G., Germany, has placed an order for 200 Ci when material is available.

SOURCE DEVELOPMENT — 08-01-04

Gadolinium-153

Separation of europium from gadolinium to obtain very high radiochemical purity gadolinium-153 continued. Approximately 18 Ci of gadolinium-153 was processed to a radiochemical purity of greater than 99.99% using high-pressure ion exchange.

Extraction of europium from gadolinium using bis-2-ethylhexyl phosphate in di-isopropyl benzene was also carried out to produce a target for HFIR irradiation.

A medical cooperative program, investigating the use of gadolinium-153 sealed sources for determination of tungsten and tantalum in lungs, reports promising results in preliminary studies using a 2-Ci source to fluoresce the tungsten and tantalum x ray.

RADIOISOTOPE SALES

J. E. Ratledge

Requests for quotations have been received from the following: Sulzer Brothers Limited, Switzerland, for eight cesium-137 sources, each containing 3750 Ci; Radiochemical Centre, England, for promethium-147 in quantities of (1) 5000 Ci to be delivered 1000 Ci/year over the next five years and (2) 10,000 Ci to be delivered 2000 Ci every 2 to 3 months over a 12- to 15-month period. An order was received from IAEA for 500 mCi phosphorus-33 to be shipped in May 1973.

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

Customer	Isotope	Amount
Large Quantities		
New England Nuclear Corporation	Tritium	8,000 Ci
Radiochemical Centre	Krypton-85	100 Ci
Sumitomo Shoji America, Inc.	Krypton-85	100 Ci
General Nucleonics, Inc.	Krypton-85, enriched	99.66 Ci
Minnesota Mining and Manufacturing Company[a]	Promethium-147	5,001 Ci
Withdrawn Items		
Mayo Clinic	Copper-67	∿13 mCi
George C. Marshall Space Flight Center, NASA	Copper-67	∿6.5 mCi
Brookhaven National Laboratory	Iodine-125	∿75 mCi
Cleveland Metropolitan General Hospital	Iodine-131	50 mCi
Items Used in Cooperative Programs		
Oak Ridge Associated Universities	Gallium-67	520 mCi
	Gadolinium-153	1 mCi
	Neodymium-147	50 mCi
	Erbium-171	50 mCi
	Bismuth-206	0.1 mCi
Vanderbilt University	Bismuth-206	5 mCi
John Hopkins University	Bismuth-206	5 mCi
Union Carbide Corporation, Y-12 Plant	Platinum-195m	0.5 mCi

Unusual Items

Carbon-13 specimens irradiated in HFIR were shipped to BNL. Approximately 0.25 mCi promethium-143 recovered from cyclotron irradiation of samarium-144 was shipped to PNL. A 1-Ci source of tellurium-123 was shipped to Bell Telephone Laboratories.

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[a]Shipped directly from PNL, Richland, Washington.

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

Table 3. Summary of Radioisotopes Shipped During March 1973

Isotope	Number of Shipments	Quantity (mCi)
Argon-37	2	4
Barium-140	3	10
Calcium-47	7	6
Cerium-144	14	20,000
Cesium-137	5	11,902,200
Copper-67	3	20
Europium-152-154	1	2
Gadolinium-153	3	101
Gallium-67	14	7,367
Iodine-129	2	350 mg
Iodine-131	1	50
Krypton-85	12	435,200
Krypton-85, enriched	6	70,100
Niobium-95	2	11
Phosphorus-33	4	610
Potassium-43	7	26
Promethium-147	2	5,001,050
Ruthenium-103	1	2
Ruthenium-106	3	24
Samarium-151	1	10 mg
Strontium-89	3	25
Strontium-90	4	200
Tritium	23	20,962,400
Xenon-133	51	57,710
Zirconium-Niobium-95	3	365
Miscellaneous preparations and targets	15	400
Cyclotron irradiations: Philips-Duphar, Netherlands	1[a]	400
Totals	193	38,458,283 360 mg

[a]Cobalt-57 as solution.

The radioisotopes sales proceeds and shipments for the first nine months of FY 1972 and FY 1973 are given in Table 4.

Table 4. Radioisotope Sales and Shipments

Item	7-1-71 thru 3-31-72	7-1-72 thru 3-31-73
Inventory items	$325,743	$362,135
Major products	47,900	42,411
Radioisotope services	223,807	150,105
Cyclotron irradiations	103,250	84,450
Miscellaneous processed materials	46,627	42,805
Packing and shipping	54,230	51,888
Total	$801,557	$733,794
Number of shipments	1,844	1,621

ADMINISTRATIVE

Visitors to the IDC are given in Table 5.

Table 5. Visitors to IDC

Visitor	Affiliation	Subject Discussed
Hirotake Kaheki	Shiba University Hospital, Japan	Nuclear medicine work
Bruce Wieland	University of Washington, St. Louis	Production of isotopes on cyclotron and tour ORNL 86-Inch Cyclotron
Henry N. Wagner, Jr.	Johns Hopkins Medical Institutions	
Gordon L. Brownell	Massachusetts General Hospital	
David E. Kuhl	University of Pennsylvania	
Robert L. Straube	Division of Research Grants, NIH	
R. L. Hayes	Oak Ridge Associated Universities	
L. Washburn	Oak Ridge Associated Universities	
C. L. Edwards	Oak Ridge Associated Universities	
Björn Dreher	Institut für Kernphysik der Universität Mainz, Germany	Radioisotope targets
Hans Dieter Wohlfahrt		
Kenji Motojima	Japan Atomic Energy Research Institute	Studies of radioisotope production and solidification of strontium and cesium

PUBLICATIONS

REPORTS

E. Lamb, *Isotopic Power Fuels Monthly Status Report for March 1973*, ORNL-TM-4215, Oak Ridge National Laboratory.

A. F. Rupp, *Radioisotope Program (8000) Progress Report for March 1973*, ORNL-TM-4214, Oak Ridge National Laboratory.

A. N. Tse, *Si-Au Schottky Barrier Nuclear Battery*, Doctorate of Engineering Thesis, Thayer School of Engineering, Dartmouth College, Hanover, New Hampshire.

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