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MASTER

ISOTOPE PROGRAM (5000) PROGRESS REPORT FOR QUARTER ENDING JUNE 30, 1970

J. H. Gillette

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ISOTOPES DEVELOPMENT CENTER

ISOTOPE PROGRAM (5000) PROGRESS REPORT
FOR QUARTER ENDING JUNE 30, 1970

J. H. Gillette

Isotopes Division

AUGUST 1970

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ISOTOPE PROGRAM (5000) PROGRESS REPORT
FOR QUARTER ENDING JUNE 30, 1970

J. H. Gillette

TARGET FABRICATION AND DEVELOPMENT

The Isotope Target Laboratory experienced its largest volume of sample preparation and work for others during this period as compared with all previous years. Much of the increased volume was due to the development and preparation of neutron dosimeters used for reactor flux and spectrum measurement and two development projects sponsored by Lawrence Radiation Laboratory, Livermore, California.

Reactor Neutron Dosimeters

Nearly 500 specimens of encapsulated isotopes were fabricated for use in reactor flux and neutron energy measurements at Argonne National Laboratory, Pacific Northwest Laboratories, and the Naval Research Laboratory. Most of the samples consisted of accurately weighed submilligram quantities of ^{237}Np , ^{238}U , ^{235}U , ^{239}Pu , and ^{238}Pu . The isotopes used and the number of dosimeters prepared are shown below:

<u>Isotope</u>	<u>Number of Dosimeters</u>
Cobalt	7
Lutetium	5
Neptunium-237	121
Plutonium-238	34
Plutonium-239	40
Potassium Iodide	40
Scandium	45
Silver	3
Uranium-235	127
Uranium-238	76

About 50 dosimeters were loaded with Al-Co, Al-Ag, and Al-Sc alloys. As described previously,¹ all samples were encapsulated in high-purity vanadium containing <15 ppm tantalum. Since commercial, high-purity vanadium contains 300-700 ppm tantalum, special zone-refined material, which was drawn into 0.050-in.-diam wire, was obtained from the ORNL Solid State Division. Since the local supply of this high-purity metal is very limited, an intensive search is in progress to locate similar or better purity vanadium. The low tantalum content is mandatory to minimize the production of tantalum radionuclides whose gamma emissions would interfere with the accurate determination of emissions from the dosimeter target material.

¹J. H. Gillette, Isotope Program (5000) Progress Report for Quarter Ending March 31, 1970, ORNL-TM-2985, Oak Ridge National Laboratory, p. 6.

Decontamination of all dosimeters containing α -active isotopes was performed by ultrasonic cleaning. This procedure, combined with a mild acid etch of the vanadium capsule, was sufficient to achieve smear tolerances of <50 dis/min in all instances except for ^{238}Pu -containing samples. To achieve low smear and probe activity levels with ^{238}Pu samples, double encapsulation in vanadium was necessary.

Techniques for extrusion of dosimeter isotopes as oxides are being studied so that repetitive, rapid measurement of quantities to be loaded into the capsules can be performed, especially where high activity level materials are involved. At present oxide powders are being loaded with a hypodermic syringe — a satisfactory technique, but not compatible with routine metering into hundreds of capsules within a glove box.

Fabrication of Thick Targets of Large Area by Rolling

A major shift in sample size has been noted over the past year. Frequent requests and orders for radio and stable isotope metals in flat, large area geometries have been received. As much as 5 kg of metal may be required in processing a sample (e.g., 4- x 8- x 0.25-in. plates of ^{238}U and ^{235}U). Not only are the areas requested increasing, but average sample thickness has increased. Processing large quantities of isotopic material in high purity form is not compatible with existing equipment used routinely for rolling metals to <0.001 in. It has been found necessary to employ large 18-in. (2-high) rolling mills for this purpose and to solicit services and equipment from the Technical Division of the Oak Ridge Gaseous Diffusion Plant (ORGDP). On the average, five samples are fabricated per week using this equipment.

The fabrication technique consists of arc melting thick forms which are suitable for rolling; however, kilogram amounts must be cast into slabs prior to rolling. Each sample is encapsulated in stainless steel and hot rolled at temperatures between 500 and 1000°C until thickness is commensurate with the final requirement or the material can be further reduced by cold rolling without cracking. Satisfactory samples of Mg, Sc, Ti, Co, Ni, Y, Zr, Nb, Mo, Hf, U, Pu, and rare-earth isotopes have been fabricated by these methods. The Dow Chemical Company, Rocky Flats Division, has assisted in the sample preparation of uranium and plutonium isotopes as no facilities for this work exist at ORNL or ORGDP. It is hoped that in the near future additional facilities can be made available at ORNL to fabricate all such samples.

A large number of 2- x 0.5- x 0.001- to 0.005-in. strips of all Zr, Mo, Zn, Sn, Fe, Co, Cu, and Ti isotopes (nonradioactive) have been fabricated. All samples could have been formed on existing equipment at ORNL except that the uniformity requirement of $\pm 1\%$ could not be attained using small quantities of starting material. To attain these desired specifications, it was necessary to first prepare a large area (4 x 6 in.) sample from which a smaller specimen having the proper uniformity can be cut. As described above, multigram samples are consolidated by arc melting and casting followed by sandwiching and hot rolling using 18-in. mills. Many samples were impossible to cold roll even after thickness reductions of 95% because the large area increased cracking probability.

Accurate uniformity measurement of the finished samples was accomplished by α -, β -, or γ -ray energy attenuation measurements. With suitable beam collimation, a thickness topograph was obtained. Unfortunately, stopping power coefficients for various radiations in many of the sample elements are not available; measurements of these parameters are being made.

Tritium Targets

Although they comprise a minor part of the Isotope Target Program, custom-fabricated tritium targets have been produced for more than ten years. Such samples are designed for use in special neutron generators yielding flux levels of 10^{12} n/sec or greater. Although much development has been carried out in this country and in European laboratories, predictable targets (in terms of neutron yield and target life) have not been routinely produced. The consensus is that contamination of sorption sites in the metal layer causes unpredictable tritium uptake with corresponding variations in neutron yield.

Far more reproducible neutron yields were attained by applying the Temescal electron bombardment evaporation system to the formation of vapor-condensed films of titanium and zirconium which are used to sorb tritium. Cleanliness of the system (residual gases) and the gettering action of the more intense vapor beam attainable with the 10-kW power supply probably were responsible for this result. Previously, evaporations were performed either with resistance heating (tungsten filaments) or by 3-kW electron bombardment heating, both of which required long evaporation times.

With the Temescal system, high purity films of most rare earths, yttrium, scandium, and other hydrogen-sorbing metals can be formed. Future plans include evaluation of neutron yield, target life, and physical and chemical characteristics of targets produced from the above-noted materials.

Two 6-in.-diam tritium targets were produced by Temescal electron bombardment evaporation of titanium onto copper substrates. The copper was given a flash coating of chromium ($100 \mu\text{g}/\text{cm}^2$) to improve adherence of the titanium layer to the substrate. A protective atmosphere of argon gas was used to inhibit poisoning of sorption sites in the $4\text{-mg}/\text{cm}^2$ titanium layer during transfer of the specimen to the tritium sorption system and during shipment. Under deuteron bombardment, the time required for the neutron output to drop to one-half the initial yield was ~ 400 mA-hr and an average neutron yield of 1.9×10^{10} n/sec·mA·steradian was experienced over two hours of bombardment.

High Purity Rare-Earth Isotopes

Large quantities (~ 50 g) of ultrahigh purity Dy, Sm, and Er metal isotopes have been and are being produced for the Solid State Division at ORNL. The purpose of this program is to convert calutron-separated nuclides from the inventory sesquioxide form to metal containing a minimal amount of oxygen and nitrogen (as well as metal impurities) so that single crystals can be grown by the strain-anneal technique. Use of the

reduction-distillation method² of metal preparation has yielded products ideally suited for this purpose with associated collection efficiencies of >90% of theoretical. In each case, thorium metal was employed to reduce the oxide and the product metal was distilled through a tantalum column and was condensed in hemispherical quartz receivers. Approximately 25 g of isotope per distillation were converted, and the product metal was consolidated by arc melting. Thus far, successful crystals of dysprosium and samarium have been grown by a member of the Solid State Division. Erbium metal distillations are still in progress.

ISOTOPIC SEPARATIONS

Isotopic separations of ^{198}Hg , ^{201}Hg , and the stable isotopes of Fe, Nd, and Sn are currently underway in separation facilities now being operated on a 5-day/week rotational shift basis. The isotopic separations program was severely limited during this quarter, inasmuch as all separations were curtailed from April 15 through June 21, 1970 due to a plant-wide work stoppage. Major emphasis during this interval of downtime was placed on technical studies to improve the efficiency of the process and identify the causes of isotopic contamination. Particular emphasis was placed on problems resulting from the use of high pressure complex charge materials. The backlog of unprocessed samples in chemistry was also reduced during this period.

RADIOISOTOPIC SEPARATIONS

Uranium-238

The chemical recycle of depleted ^{238}U , used as feed material in series (an), was completed with the recovery and purification of 2311 g of uranium as U_3O_8 . Chemical refinement of the enriched products was also completed with the recovery of the following materials:

<u>Lot Number</u>	<u>Weight (g)</u>	<u>^{235}U Content (ppm)</u>
250A	858.9	2.4
250B	130.2	5.0
250C	105.6	10.4

Uranium-233

A ^{233}U collection series (ao) is currently underway to obtain approximately 100 g of uranium containing <52 ppb of ^{232}U . This product, when recovered, will be used as the "cow" in generating ^{229}Th relatively free from ^{228}Th . Thus far approximately 70 g have been collected. More than 550 g of the ^{233}U feed has been recovered from calutron wash solutions, purified to U_3O_8 , and is now available for subsequent runs in the calutron.

²E. H. Kobisk and W. B. Grisham, Application of Reduction-Distillation Method for Preparing High-Purity Rare-Earth Isotope Metals, Mater. Res. Bull. 4: 651-62 (1969).

Uranium-Palladium Alloy

A special project is underway to reclaim >900 g of natural abundance palladium from approximately 1 kg of U-Pd alloy. The uranium is enriched in ^{235}U and for the palladium to be useful, the uranium content should be reduced to <100 ppm. Two different alloy compositions were received: the first consisting of approximately 500 g of 1% uranium by weight and the second approximately 500 g but containing 10% by weight uranium. After some initial exploratory work, the 1% uranium-composition alloy was started first and the recovery is about 90% completed.

Miscellaneous Projects

The consolidation and re-assay of three batches of high isotopic purity ^{233}U , which is to be reduced to UO_2 and pelletized by ORNL Ceramics Division for a project at Knolls Atomic Power Laboratory, is about 90% completed.

Several batches of radiogenic ^{208}Pb , enriched to approximately 72% in ^{208}Pb , have been combined and chemically processed to approximately 1.8 kg of pure lead carbonate.

180° SECTOR ISOTOPE SEPARATOR

Our PDP-8 computer has been tested and is being connected to the 180° Oak Ridge sector isotope separator. This Computer Data System will be used to collect experimental run data during separator operation that is now being recorded periodically by the sector operator. The system will be used later to assess the possibility of operating the sector with computer assistance.

The most recent exploratory strontium isotope collections in the Oak Ridge sector isotope separator have resulted in the recovery of 4 mg of 99.54% and 5 mg of 99.72% ^{84}Sr . Samples of this isotopic purity are ideal for mass spectrometry earth science research and in the production of high purity $^{85\text{m}}\text{Sr}$. The recovery of ^{84}Sr from the last run, using a platinum-lined isotope collector, was 55% of the monitored ion current. This collection technique is a decided improvement over the use of conventional isotope collectors and will be used in all subsequent strontium collections to facilitate the chemical recovery and refinement of the small samples involved.

CHEMICAL PROCESSING

Material in Process

Elements in chemical processing during the quarter ending June 30, 1970 are: Sb, Ce, Fe, Hg, Nd, Re, Te, and Sn.

Material Added to Inventory

Isotope material released to the Sales Department during the second quarter of calendar year 1970 is shown in Table 1.

Table 1. Material Added to Inventory

Isotope	Element Weight (g)	Assay (%)	Compound Form
Cerium-138	2.244	26.0	CeO ₂
	2.782	17.61	CeO ₂
	2.435	11.35	CeO ₂
Cerium-142	66.850	92.11	CeO ₂
	19.932	83.10	CeO ₂
Chromium-50	2.785	88.05	Cr ₂ O ₃
Mercury-198	1.203	92.26	HgO
	1.969	82.20	HgO
	3.899	71.68	HgO
	1.779	54.43	HgO
Mercury-201	2.572	95.56	HgO
	6.299	92.34	HgO
	6.545	85.53	HgO
	6.472	74.12	HgO
	2.739	65.58	HgO
Rhenium-185	68.193	96.25	Re
Rhenium-187	74.905	99.22	Re
	54.460	96.91	Re
Tin-116	0.090	99.25	Sn
Tin-117	0.068	98.81	Sn
Tin-118	0.214	99.684	Sn
Tin-119	0.081	99.10	Sn

ISOTOPES PREPARATION AND SALES

The radioisotope and stable isotope target and research material preparations for the quarter ending June 30, 1970 are given in Tables 2 and 3, respectively.

Table 2. Radioisotope Preparations

Isotope	Number of Preparations	Isotope	Number of Preparations
Americium-243	4	Thorium-232	7
Californium-252	11	Tritium	9
Curium-244	5	Uranium-234	2
Neptunium-237	5	Uranium-235	9
Plutonium-239	21	Uranium-235- Thorium-232 Alloy	1
Plutonium-240	2		

Table 3. Stable Isotope Preparations

Isotope	Number of Preparations	Isotope	Number of Preparations
Bismuth-209	1	Nickel-60	2
Calcium - Normal	1	Nickel-61	1
Calcium-42	2	Niobium-93	1
Cobalt-59	13	Platinum-195	1
Cadmium-112	1	Samarium - Normal	3
Cadmium-114	1	Samarium-144	1
Chromium-52	1	Scandium-45	3
Dysprosium-160	1	Silver-107	1
Erbium - Normal	1	Silver-109	3
Gold-197	1	Strontium-87	2
Indium-115	1	Strontium-88	2
Iron-54	3	Tantalum - Normal	4
Iron-56	1	Terbium-159	3
Iron-57	5	Tin-116	2
Iron-58	3	Tin-122	1
Lead - Normal	3	Tin-124	1
Lead-206	2	Titanium-46	1
Lead-208	1	Titanium-47	2
Magnesium-24	1	Titanium-49	1
Magnesium-26	1	Titanium-50	1
Molybdenum-92	1	Tungsten - Normal	3
Molybdenum-94	1	Yttrium-90	1
Molybdenum-96	1	Ytterbium-173	1
Molybdenum-98	1	Zirconium-90	3
Molybdenum-100	1	Zirconium-91	2
Neodymium-142	2	Zirconium-92	3
Nickel-58	20	Zirconium-94	2
Manganese-55	1	Zirconium-96	2

The stable isotopes sales and services for the previous quarter are given in Table 4.

Table 4. Stable Isotope Sales and Services
April 1 through June 30, 1970

	Shipments	Revenue
EM Isotope Sales	997	\$326,161
Non-EM Isotope Sales	86	41,327
Services:		
Target Preparations		117,912
Work for Others		43,000
Special Services		12,126
Reprocessing Returned Loans		<u>7,715</u>
Total Services		\$180,753
Total Stable Isotopes	1083	\$548,241

ADMINISTRATIVE

Visitors for the quarter ending June 30, 1970 are given in Table 5.

Table 5. Visitors

Affiliation	Purpose of Visit
Dragon Project, England	Discuss electromagnetic separation techniques
New England Nuclear Corporation	Tour EM separation facility
AEC, Washington	Tour EM separation facility
General Electric	Tour EM separation facility
Atomic Industrial Forum, Inc.	Tour EM separation facility
Lawrence Radiation Laboratory Livermore, California	Discuss fabrication projects

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18. J. H. Gillette	45. D. A. Sundberg
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EXTERNAL DISTRIBUTION

59. D. Axelrod, National Institute of Health, Bethesda, Maryland
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61. G. W. Barton, Jr., Lawrence Radiation Laboratory, Livermore, California
62. K. E. Chellis, Florida State University, Tallahassee, Florida
63. J. T. Christy, AEC-RL00, Richland, Washington
64. D. C. Davis, AEC-ORO, Oak Ridge, Tennessee
65. D. R. de Boisblanc, Phillips Petroleum Company, Idaho Falls, Idaho
66. J. C. Dempsey, DID, AEC, Washington, D. C.
67. B. J. Dropesky, Los Alamos Scientific Lab, Los Alamos, New Mexico
68. E. E. Fowler, DID, AEC, Washington, D. C.
69. L. Friedman, BNL, Upton, New York
70. J. D. Goldstein, AEC, Div. of Biology and Medicine, AEC, Washington, D.C.
71. R. N. Goslin, Oglethorpe University, Atlanta, Georgia
72. K. A. Gschneidner, Jr., Dept. of Metallurgy and Institute for Atomic Research, Iowa State University, Ames, Iowa
73. C. C. Harris, Medical Department, Duke University, Durham, N. C.
74. Max Harris, Lawrence Radiation Laboratory, Livermore, California
75. R. Herber, Rutgers University, New Brunswick, New Jersey
76. I. A. Hobbs, AEC-SRO, Aiken, South Carolina
77. T. R. Jones, Div. of Research, AEC, Washington, D. C.
78. J. S. Kirby-Smith, Div. of Biology and Medicine, AEC, Washington, D. C.

79. G. A. Kolstad, Div. of Research, AEC, Washington, D. C.
80. J. Lawrence, Lawrence Radiation Laboratory, Berkeley, California
81. J. L. Lerner, ANL, Argonne, Illinois
82. J. S. Lichliter, Lawrence Radiation Laboratory, Berkeley, California
83. J. A. Maddox, DID, AEC, Washington, D. C.
84. W. B. Mann, National Bureau of Standards, Washington, D. C.
85. R. B. Martin, Laboratory and University Division, AEC, ORO
86. S. J. Marsalis, Mississippi State College for Women, Columbus, Mississippi
87. P. W. McDaniel, Division of Research, AEC, Washington, D. C.
88. M. C. Michel, Lawrence Radiation Laboratory, Berkeley, California
89. D. R. Miller, Division of Research, AEC, Washington, D. C.
90. R. A. Neumann, Princeton University, Princeton, New Jersey
91. H. W. Newson, Physics Department, Duke University, Durham, N. C.
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