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ISOTOPE PROGRAM (5000) PROGRESS REPORT FOR QUARTER ENDING MARCH 31, 1971

J. H. Gillette

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ISOTOPES DEVELOPMENT CENTER

ISOTOPE PROGRAM (5000) PROGRESS REPORT FOR QUARTER ENDING MARCH 31, 1971

J. H. Gillette

Isotopes Division

MAY 1971

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee
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CONTENTS

Page

TARGET FABRICATION AND DEVELOPMENT 1

Radioisotope Preparations 1

Curium-244 Cross-Section Samples 1
Multiple Uranium Oxide Targets 2
Large Area Tritium Targets 2
Shipping Container for Thin Film Targets 3

ISOTOPIC SEPARATIONS 3

RADIOISOTOPIC SEPARATIONS 3

Uranium-238 3

Uranium-235 4

Thorium-230 4

THE 180° SECTOR ISOTOPE SEPARATOR 4

EXPLORATORY MERCURY SEPARATIONS 5

SOURCE STUDIES 5

CHEMICAL PROCESSING 5

Material in Process 5

Material Added to Inventory 5

ISOTOPES PREPARATION AND SALES 6

ADMINISTRATIVE 9

ISOTOPE PROGRAM (5000) PROGRESS REPORT FOR QUARTER ENDING MARCH 31, 1971

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TARGET FABRICATION AND DEVELOPMENT

Most samples produced during this period were rather routine in nature, but the volume increased over the preceding period by 89% for stable isotope research materials and 23% for radioisotope sample preparations.

Radioisotope Preparations

Curium-244 Cross-Section Samples

Approximately 80 mg of high isotopic purity $^{244}CmO_2$ was made available by TRU for preparation of two cross-section targets to be used at ORELA. One sample contained 10 mg and the other ~60 mg of the isotope. This isotope, enriched to >99% by the calutrons, was purified by ion exchange (TRU) to remove the decay daughter product, $^{240}PuO_2$.

Because of the precise nature of the measurements to be performed, a special double capsule was designed so that the isotope could be exactly located in the neutron beam. Capsule dimensional tolerances of ±0.0001 in. on the internal and external diameters of each component were required, and since the capsule material was 1100-series aluminum, the machining of these containers was critical.

Each of the two curium aliquots was dissolved and diluted with aluminum nitrate before the two elements were coprecipitated as their respective oxalates. The exact concentration of aluminum atoms was preserved in each curium sample, although the curium content varied by a factor of 6. This was to permit direct data subtraction. Aluminum dilution made a more voluminous sample available for capsule loading which would result in a more uniform distribution in depth and cross section of the curium atoms.

Each curium-aluminum sample was pressed into the 0.125-in.-diam cylindrical hole in the cylindrical inner capsule. End plugs of 0.010-in.-thick aluminum metal were formed on each side of the Cm-Al oxide compact by pressing a carefully weighed amount of high-purity aluminum powder in each side. The composite pressing locked the curium compact into the sample tube such that even severe handling could not dislodge the sample. Compaction of the various layers was performed so that no dimensional variation in the wall thickness or outer diameter of the aluminum inner capsule occurred.

A slight amount of stearic acid was applied to the outer wall of the inner capsule to facilitate loading into the outer shell. Both inner and outer capsules were sealed on each end with a 0.015-in.-thick aluminum window joined to the assembly using radiation-resistant epoxy cement. After capsule assembly and cleaning, external surfaces were found to be contaminated

to <10 dpm alpha activity. The capsule assembly was estimated to be capable of withstanding temperatures up to 200°C and was experimentally found to retain structural integrity to the temperature of boiling liquid nitrogen.

Multiple Uranium Oxide Targets

Fifteen duplicate deposits of $^{235}UO_2$ on stainless steel (304L) substrates were produced, each having an 0.8-in.-diam deposit. These samples were specified by LRL-Livermore to be uniform in thickness to ±10%, when comparing one sample with all others, and to have an individual coating uniformity of ±5%.

A substrate holder suitable for containing all 1-in.-diam circular substrates was designed, and a rotation mechanism was developed to permit each substrate to reside in the UO_2 vapor for equal time intervals during evaporation condensation. By using electron-beam heating of a pressed pellet of UO_2, uniform evaporation of the oxide was achieved such that the evaporation rate could be made constant over a time interval of 30 min, and the vapor beam geometry was reasonably constant. With these vacuum chamber fixtures, it was possible to produce the targets so that each sample was uniform to ~±2% (as determined by radiation counting), and intercomparison among the 15 samples showed a uniformity of ±5% as determined by mass measurements. All deposits were very adherent (could not be removed by scotch tape or mild abrasion) since the oxide was condensed at a substrate temperature of >350°C.

Large Area Tritium Targets

For many years, preparation of tritium-titanium targets for use in generating 14-MeV neutrons by the reaction $^{3}H(^{2}H,n)^{4}He$ has been plagued with irreproducibility. Target stability in terms of neutron yield and lifetime has not been routinely predictable. Recently LRL-Livermore has required twenty 6-in.-diam targets of this type that can produce ~4 x 10^{12} n/sec. Success in producing such targets has been achieved by using electron-bombardment-heated titanium vapor sources in a vacuum of ~10^{-7} Torr followed by immediate transfer to the tritium sorption system.

Each target is formed using a 6-in.-diam dish-shaped metal substrate of Cu-0.1% Zr alloy (Amzirc). Film thickness of the annular titanium deposit was specified to be 4 mg/cm^2; this thickness produced a total sorbed tritium content of ~300 Ci. When three different targets were run in the accelerator, neutron production and target life did not vary more than 10%. Moreover, the uniformity of tritium in the target face was found by bremsstrahlung counting to be at least as good as ±10%. Unfortunately, no reproducible correlation between radiation readings and neutron output could be made with certainty because of the rather crude counting technique employed. At present twenty similar targets are being produced on contract.

Shipping Container for Thin Film Targets

A container has been designed and constructed to protect thin self-supporting targets from fracture during shipment from ORNL. Tests of the container under conditions of mechanical shock and finally an actual cross-country shipment of carbon films (35 $\mu g/cm^2$) to California and back proved the success of the suspension system.

The present design involves suspension of a target holder plate among springs (eight total) attached to an internal steel framework. The entire framework and spring assembly can be removed from the container housing for ease of mounting or removal of targets. So that reactive or radioactive targets (e.g., barium metal or radium bromide) can be shipped, the outer container can be sealed with a gasketed lid and can contain an inert atmosphere (argon) or can be evacuated. Either Plexiglas (for chemically inert targets) or steel outer containers can be used depending on the environment in which the target must be housed for maximum preservation.

Use of one of these shipping containers will be made in transporting targets of 100 $\mu g/cm^2$ $RaBr_2$ on 35-$\mu g/cm^2$ carbon substrates to Germany by ordinary commercial carriers. If this shipment is successful, customers will no longer need to hand carry targets or be bothered with the removal of films from plates having a water-soluble layer between the desired film and a rigid support. Many targets can be offered to the research community that heretofore were not possible to provide because of almost sure fracture during shipment.

ISOTOPIC SEPARATIONS

Isotopic separations of cadmium, gadolinium, and strontium are currently underway in 14 calutrons on a 5 day around-the-clock basis. All four isotopes of strontium and five of the seven isotopes of gadolinium are being collected in 180° beta-type calutrons, while all eight isotopes of cadmium are being separated in the three modified 255° separators. During this quarter, the gallium separation series was terminated after it reached the stated purity and quantity requirements and was replaced by the strontium isotope separation. The major objective in the strontium separation is the recovery of $\geq$20 g of ~80% ^{84}Sr, which is needed to replenish the sales inventory. An iron isotope separation and a mercury collection with emphasis on ^{198}Hg and ^{201}Hg were both discontinued during the quarter due to budgetary restrictions.

RADIOISOTOPIC SEPARATIONS

Uranium-238

A ^{238}U separation series was initiated on December 1, 1970, to obtain ~2 kg of ^{238}U containing <30 ppm ^{235}U. The separation was completed in early March with ion currents equivalent to 2116 g ^{238}U monitored. In this separation, double-arc equipment was utilized in four of the eight alpha-contained calutrons and UO_2 plus CCl_4 was used for charge material.

Preliminary mass analyses of composited samples show that the majority of the material collected will contain 6 to 12 ppm ^{235}U.

Uranium-235

A ^{235}U enrichment program begun in October 1970 was interrupted on December 1, 1970, in order to collect the ^{238}U mentioned above. The ^{235}U collection was resumed after the ^{238}U collection, and at this writing a monitored equivalent of ~330 g ^{235}U has been accumulated. Approximately 600 g ^{235}U containing <0.1% ^{234}U and ≦0.5% ^{236}U will be available when the separation is completed.

Two samples have been recovered, chemically refined, and made available to the Research Materials Collection to satisfy immediate needs. Analytical data in Table 1 show the quality of the material.

Table 1. Analysis Results of ^{235}U Samples

Sample Designation	Recovered Weight (g)	Mass Assay (%)		
		^{234}U	^{235}U	^{236}U
ar 265A	14.649	0.024	99.934	0.013
ar 265B	198.41	0.0298	99.912	0.0165

Thorium-230

A special high-purity ^{230}Th separation has been completed in one of the alpha-contained calutrons using 1 g of thorium that contained 69% ^{230}Th. This material was obtained along with ~2 g of ~90% ^{230}Th. The separation yielded about 70 mg of >99% ^{230}Th.

THE 180° SECTOR ISOTOPE SEPARATOR

The PDP-8/I minicomputer data system has been successfully used to gather operational data from the 180° Oak Ridge Sector Isotope Separator, and, in addition, it has exercised a degree of operational control. The collected data include vacuum levels, high voltage power supply drains and voltages, analyzing magnetic field, beam currents, and ion-source parameters as reflected back through to the primary of the 40-keV isolation transformers. Several ion source and power supply functions have been successfully controlled by the computer during isotope collection runs.

Mercury isotope separations have begun in the ORSIS to determine whether ^{198}Hg, ^{199}Hg, and ^{201}Hg of >99% purity can be obtained in one pass. Mercury-196 is, or will be, collected simultaneously with each of these.

EXPLORATORY MERCURY SEPARATIONS

The ion source vaporizing metallic mercury feed from the water-heated constant-temperature reservoir has been equipped with various arc chambers in search of improved focal properties. Preference to date lies with an extremely shallow chamber (~0.090 in. from back of chamber to ion exit slit) used in conjunction with an ion exit slit having a height of 0.062 in. Accompanying these source changes are receiver evaluations in which the Hg^+ is collected in copper, in oil-filled copper pockets, and in the oil-filled recessed pocket.

SOURCE STUDIES

Work in evaluating conditions which reduce multiple ion formation in calutron-type sources faces power supply problems in that controls which compensate for filament heating by ion bombardment are lacking. Run time at high power inputs is often limited to minutes because the ionization filament melts.

CHEMICAL PROCESSING

Material in Process

Elements in chemical processing during the quarter ending March 31, 1971, are Cd, Fe, Ga, Gd, Hg, Sb, Sn, Sr, and Te.

Material Added to Inventory

Isotopic material released to the Sales Department during the first quarter of calendar year 1971 is shown in Table 2.

Table 2. Material Added to Inventory

Isotope	Element Weight (g)	Assay (%)	Compound Form
Antimony-123	48.627	99.05	Sb
Gadolinium-155	3.069	99.82	Gd_2O_3
Gadolinium-157	2.666	99.70	Gd_2O_3
Iron-57	16.128	67.90	Fe_2O_3
Iron-58	18.469	65.09	Fe_2O_3
Iron-58	3.203	54.29	Fe_2O_3
Mercury-201	0.448	98.11	HgO
Strontium-84	0.0046	99.32	$Sr(NO_3)_2$
Strontium-84	0.0056	99.31	$Sr(NO_3)_2$

Table 2. continued

Isotope	Element Weight (g)	Assay (%)	Compound Form
Tin-112	12.358	82.70	SnO_2
Tin-114	20.099	64.12	SnO_2
Tin-115	6.835	32.72	SnO_2
Tin-115	8.377	26.79	SnO_2
Tin-115	2.256	19.95	SnO_2
Tin-117	276.716	84.23	SnO_2
Tin-119	222.478	84.48	SnO_2
Tin-120	261.097	98.05	SnO_2
Tin-122	152.735	92.20	SnO_2
Tin-124	35.036	96.96	SnO_2
Tin-124	152.371	96.71	SnO_2

ISOTOPES PREPARATION AND SALES

The radioisotope and stable isotope target and research material preparations for the quarter ending March 31, 1971, are given in Tables 3 and 4.

Table 3. Radioisotope Preparations

Isotope	Number of Preparations	Isotope	Number of Preparations
Americium-243	9	Plutonium-241	34
Californium-252	17	Protactinium-231	2
Curium-244	4	Thorium-230	2
Curium-244—Carbon-13	1	Thorium-232	1
Neptunium-237	126	Tritium	4
Plutonium-238	51	Uranium-235	35
Plutonium-239	59	Uranium-238	6
Plutonium-240	33		

Table 4. Stable Isotope Preparations

Isotope	Number of Preparations	Isotope	Number of Preparations
Barium-138	1	Mercury-196	1
		Mercury-198	1
Cadmium-111	1	Mercury-199	1
Cadmium-114	1	Mercury-201	1
		Mercury-202	1
Calcium – Normal	2	Mercury-204	1
Calcium-44	1		
Calcium-48	1	Molybdenum-92	1
Caldium-40, 42, 44, 48	1	Molybdenum-95	2
		Molybdenum-96	1
Cobalt – Normal	2	Molybdenum-97	2
		Molybdenum-98	1
Copper-63	1	Molybdenum-100	2
Chromium-52	1	Neodymium-142	2
Chromium-53	1	Neodymium-150	2
Dysprosium-164	1	Nickel – Normal	1
		Nickel-58	5
Erbium-166	1		
Erbium-167	1	Phosphorus – Normal	2
Erbium-168	1		
		Potassium – Normal	2
Germanium-72	1		
Germanium-74	1	Samarium-144	1
Germanium-76	1	Samarium-148	1
		Samarium-152	2
Hafnium-177	1	Samarium-154	1
Iron – Normal	1	Scandium – Normal	20
Iron-54	1		
Iron-56	2	Silicon-28	3
Iron-57	1	Silicon-30	1
Iron-57 – SS 310	1		
		Sodium – Normal	2
Lanthanum – Normal	1		
		Strontium-86	2
Lead-206	1	Strontium-87	3
Lead-207	1	Strontium-88	5
Lead-208	2		
		Tellurium-128	1
Lithium-6	36		
Lithium-7	4	Thallium-205	1
Magnesium-24	5	Tin-114	1
Magnesium-26	1	Tin-116	1

Table 4. continued

Isotope	Number of Preparations	Isotope	Number of Preparations
Tin-118	2	Ytterbium-170	1
Tin-120	1	Ytterbium-173	1
Tin-124	2		
		Zirconium-90	6
Titanium-46	3	Zirconium-91	4
Titanium-48	4	Zirconium-92	4
		Zirconium-94	1
Tungsten-182	1	Zirconium-96	3

The isotope sales and services for the quarter ending March 31, 1971, are given in Table 5.

Table 5. Isotope Sales and Services
January 1 Through March 31, 1971

	Shipments	Revenue
EM Isotope Sales	993	$194,483
Non-EM Sales	44	$ 43,467
Services:		
Stable Isotope Target Preparations		$ 39,180
Radioisotope Target Preparations		52,301
Special Services		15,064
Reprocessing Returned Loans		10,525
Work for Others		27,134
Total Services		$144,204
Total	1037	$382,154

ADMINISTRATIVE

Visitors for the quarter ending March 31, 1971, are shown in Table 6.

Table 6. Visitors

Affiliation	Purpose of Visit
AECL	Discuss isotope separation
Rocky Flats	Discuss ^{241}Am shipments
LRL-Livermore	Discuss LRL special projects

INTERNAL DISTRIBUTION

1. A. L. Allen
2. M. A. Baker
3. P. S. Baker
4. E. E. Beauchamp
5. W. A. Bell
6. D. S. Billington
7. G. E. Boyd
8. T. A. Butler
9. F. N. Case
10. J. W. Cleland
11. T. F. Connolly
12. F. L. Culler
13. W. C. Davis
14. J. S. Drury
15. J. L. Fowler
16. J. H Frye, Jr.
17. J. H. Gibbons
18. J. H. Gillette
19. W. F. Good
20. H. R. Gwinn
21. J. A. Harvey
22. R. F. Hibbs
23. J. E. Keeton
24. O. L. Keller
25. Lynda Kern
26. E. H. Kobisk
27. R. H. Lafferty, Jr.
28. E. Lamb
29. R. E. Leuze
30. R. S. Livingston
31. L. O. Love
32. W. S. Lyon
33. F. C. Maienschein
34. F. M. O'Hara
35. W. K. Prater
36. J. J. Pinajian
37. M. E. Ramsey
38. S. A. Reynolds
39-40. R. A. Robinson
41. A. F. Rupp
42. R. W. Schaich
43. M. R. Skidmore
44. A. H. Snell
45. P. H. Stelson
46. H. F. Stringfield
47. D. A. Sundberg
48. A. M. Weinberg
49. J. C. White
50. A. Zucker
51-52. Central Research Library
53. Document Reference Section
54-59. Laboratory Records Department
60. Laboratory Records - RC

EXTERNAL DISTRIBUTION

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

80. G. A. Kolstad, Div. of Research, AEC, Washington, D. C.
81. J. Lawrence, Lawrence Radiation Laboratory, Berkeley, California
82. J. L. Lerner, ANL, Argonne, Illinois
83. J. S. Lichliter, Lawrence Radiation Laboratory, Berkeley, California
84. J. A. Maddox, DID, AEC, Washington, D. C.
85. W. B. Mann, National Bureau of Standards, Washington, D. C.
86. R. B. Martin, Laboratory and University Division, AEC, ORO
87. S. J. Marsalis, Mississippi State College for Women, Columbus, Mississippi
88. P. W. McDaniel, Division of Research, AEC, Washington, D. C.
89. J. W. Meadows, Lawrence Radiation Laboratory, Livermore, California
90. M. C. Michel, Lawrence Radiation Laboratory, Berkeley, California
91. D. R. Miller, Division of Research, AEC, Washington, D. C.
92. R. A. Neumann, Princeton University, Princeton, New Jersey
93. H. W. Newson, Physics Department, Duke University, Durham, N. C.
94. A. O. C. Nier, University of Minnesota, Minneapolis, Minnesota
95. G. C. Phillips, Rice University, Houston, Texas
96. D. M. Richmond, Div. of Research, AEC, Washington, D. C.
97. R. B. Roberts, Carnegie Institute of Washington, Washington, D. C.
98. G. L. Rogosa, Division of Research, AEC, Washington, D. C.
99. H. M. Roth, Laboratory and University Division, AEC, ORO
100. B. C. Rusche, SRL, Aiken, South Carolina
101. W. D. Sandberg, AEC, Aiken, South Carolina
102-103. J. Spaepen, CBNM-CEA, Geel, Belgium
104. L. G. Stang, BNL, Upton, New York
105. J. A. Swartout, Union Carbide Corporation, New York
106. W. L. Talbert, Jr., Iowa State University, Ames, Iowa
107. G. Taplin, Dept. of Biophysics & Nuclear Medicine, University of California, Los Angeles, California
108. G. R. Tilton, University of California at Santa Barbara, Santa Barbara, California
109-114. D. H. Turno, SRL, Aiken, South Carolina
115. A. R. Van Dyken, Division of Research, AEC, Washington, D. C.
116. L. E. Wahlin, University of Colorado, Boulder, Colorado
117. M. Williams, Lawrence Radiation Laboratory, Livermore, California
118-119. Division of Technical Information Division