LOCKHEED MARTIN ENERGY RESEARCH LIBRARIES
3 4456 0514439 5

CENTRAL RESEARCH LIBRARY
DOCUMENT COLLECTION

OAK RIDGE NATIONAL LABORATORY
operated by
UNION CARBIDE CORPORATION • NUCLEAR DIVISION
for the
U. S. ATOMIC ENERGY COMMISSION

ORNL - TM - 3529
cy 52

ISOTOPE PROGRAM (5000) PROGRESS REPORT FOR QUARTER ENDING JUNE 30, 1971

J. H. Gillette

OAK RIDGE NATIONAL LABORATORY
CENTRAL RESEARCH LIBRARY
DOCUMENT COLLECTION
LIBRARY LOAN COPY
DO NOT TRANSFER TO ANOTHER PERSON
If you wish someone else to see this document, send in name with document and the library will arrange a loan.
UCN-7969
(3 3-67)

NOTICE This document contains information of a preliminary nature and was prepared primarily for internal use at the Oak Ridge National Laboratory. It is subject to revision or correction and therefore does not represent a final report.

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

ORNL-TM-3529

Contract No. W-7405-eng-26

ISOTOPES DEVELOPMENT CENTER

ISOTOPE PROGRAM (5000) PROGRESS REPORT FOR QUARTER ENDING JUNE 30, 1971

J. H. Gillette

Isotopes Division

AUGUST 1971

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee
operated by
UNION CARBIDE CORPORATION
for the
U.S. ATOMIC ENERGY COMMISSION

LOCKHEED MARTIN ENERGY RESEARCH LIBRARIES

3 4456 0514439 5

CONTENTS

	Page
TARGET FABRICATION AND DEVELOPMENT	1
Measurement of Film Thickness and Uniformity	1
Reduction of $^{243}AmO_2$ to Metal	2
Parting Agent Studies	2
Standard Targets for Cross-Section Measurements	2
Ion-Implanted Targets	3
ISOTOPIC SEPARATIONS	3
RADIOISOTOPIC SEPARATIONS	3
Uranium-238	3
Uranium-235	4
KRYPTON-85 RECOVERY	4
180° SECTOR ISOTOPE SEPARATOR	4
CHEMICAL PROCESSING	5
Material in Process	5
Material Added to Inventory	5
ISOTOPES PREPARATION AND SALES	6
ADMINISTRATIVE	8

ISOTOPE PROGRAM (5000) PROGRESS REPORT FOR QUARTER ENDING JUNE 30, 1971

J. H. Gillette

TARGET FABRICATION AND DEVELOPMENT

Measurement of Film Thickness and Uniformity

Determination of the number of atoms of a nuclide and their distribution in a target sample is of considerable importance to the accelerator physicist. Atom count can be determined by measuring the dimensional thickness of a sample, and, assuming that the target material is theoretically dense, the product of the calculated volume of the sample and its density should yield the total mass which can then be easily converted into an atom count. Measurement of the sample thickness can be performed by interferometry with high accuracy, but the method is destructive. Furthermore, for films of thickness <2000 Å, the density is not theoretical in many cases, especially if the sample was produced by vacuum evaporation-condensation. Errors in this procedure can be as great as 20% depending upon sample size and the material.

A superior method of thickness measurement is that of determining the energy attenuation or absorption of fission fragments, alpha particles, beta particles, or gamma radiation in the foil or film. This has the advantage of being a nondestructive technique. Choice of the radiation used in the process is dependent upon sample thickness; e.g., fission fragments would be used to measure the thickness of films in the 50-800 Å range, while beta particles would be more appropriate when measuring foils of 10,000-100,000 Å thickness. Energy attenuation of the incident radiation is a function of the initial energy of the particle and the material being measured. This same process can be used to measure thickness in different portions of a target film and thus yield a measure of the atom uniformity or distribution. Being directly related to the number of atoms in the sample, the accuracy of this process is not affected by deviations from theoretical density. This process yields data exactly in the form suitable for the physicist's application.

So as to achieve maximum accuracy and precision in thickness measurements, the energy shift of the incident charged particle must be measured with great accuracy. For this purpose an NDA-12 multichannel analyzer (Nuclear Data, Inc.) is being used which has a 3072-channel capacity that can yield a minimum energy variation between channels of ~0.25 keV. With this instrument a 20-$\mu g/cm^2$ film can be measured with an error of ±5% or less. At present, measurements of stopping power for many elements in a variety of thicknesses commonly used as target materials are being made so that thickness calibrations will be available.

Reduction of $^{243}AmO_2$ to Metal

Using the high temperature reduction-distillation method, 2.5 g of ^{243}Am, as oxide, was converted to metal. Thorium metal was used as the reductant. A stoichiometric amount of thorium was used with an ~10% excess. Collection of the vapor condensate in a quartz thin-wall hemisphere resulted in a 94% yield of highly malleable metal.

Parting Agent Studies

By electron microscopy, the structure of vapor deposited NaCl, Na_3PO_4, "Victawet," CsCl, and $BaCl_2$ was studied. These materials are frequently used as an intermediate layer between a thin film (usually vapor deposited) and a metal or glass substrate. When the sandwich-like structure is immersed in distilled water, the intermediate layer is dissolved and the film overcoating is released from the heavy substrate, hence the name "parting agent" is ascribed to these materials. In addition, several water-soluble organic-base materials, i.e., detergents, were studied by similar methods.

In this study, carbon replicas of the parting-agent-coated surface were examined after the surfaces were shadowed with chromium. Effects of parting agent deposition techniques, substrate temperature, atmospheric exposure, storage, and other environmental conditions were studied in this manner. Optical microscopy of the dissolving parting agent-water interface proved to be of considerable value in judging the relative merits of each agent.

As the result of this effort, the choice of parting agent to be used in preparing thin film isotope targets was well defined. Much of the "inexplicable" black magic of this portion of target fabrication has been eliminated, and considerable progress toward selecting appropriate parting agents in the formation of single crystal thin films has been made. This work will be reported at the "International Symposium on Research Materials for Nuclear Measurements," October 5-8, 1971, in Gatlinburg, Tennessee.

Standard Targets for Cross-Section Measurements

Evaporation-condensation of highly enriched isotopes (^{10}B, $^{235}UO_2$, $^{238}UO_2$) is being used to form well defined thin layers of these materials for use in cross-section measurements at ORELA. In this process, vapors are condensed onto a planar substrate of aluminized mylar film (~0.001 in. thick). Both sides of the plastic substrate require coating to a thickness of 10 $\mu g/cm^2$ of the respective isotope. Although the coating technology is not unique, definition of such thin coatings to an accuracy of ~1% is difficult at best and requires the application of in-situ thickness monitoring, thickness measurement after deposition, film uniformity measurement, and chemical and isotopic analyses.

In-situ thickness monitoring is performed using a quartz resonating crystal located within a short distance of the vapor source as compared to

substrate location. In this manner, more material is condensed on the crystal per unit time than on the substrate, which improves the accuracy of the thickness measurement. The crystal coating thickness is roughly proportional to that on the plastic substrate according to the inverse ratio of the squares of distances from the vapor source.

Post deposition, thickness, and uniformity measurements are performed by charged particle energy attenuation on the sample. Since each attenuation measurement subtends an area of about 0.1 cm, many thickness measurements and a topographical mapping of thickness over the circular 20-cm^2 area can be made. Other thickness measurement techniques involving optical density are being investigated to improve the accuracy of the measurements.

Ion-Implanted Targets

Twenty-five targets of ^{191}Ir, ^{193}Ir, ^{185}Re, and ^{187}Re were prepared by implanting approximately 10 $\mu g/cm^2$ of isotopically separated ions at approximately 40 keV on both iron and nickel substrates. Rhenium ions were obtained in the sector separator from the internal charge system, rhenium metal plus oxygen, while iridium ions were produced by electron bombardment of the metal in a beta calutron.

ISOTOPIC SEPARATIONS

Isotopic separations of calcium, hafnium, mercury, and rhenium are currently underway in five 180° beta-type calutrons, three 255° modified calutrons, and the 180° sector separator. All calutron separations are being made on an 8-hr 5-day basis, while the sector is being operated on a 5-day around-the-clock basis. The calcium isotope separation is exploratory in nature in an effort to establish satisfactory operating conditions for medium temperature elements on an 8-hr operating schedule. The hafnium isotope separation was initiated on April 5, 1971, in the three 255° separators to fill both research and sales needs. An early evaluation of hafnium mass assay and retention data indicates that the hafnium charge system, $HfO_2 + CCl_4$, like other high vapor pressure charge materials that can react with the collected material, is adversely affecting both isotopic retention and mass assays. Trial preparations are underway in the charge preparation laboratory to prepare HfF_4 by wet chemistry methods. This compound, which is usually prepared by anhydrous gaseous reactions with F_2 or HF, has a lower vapor pressure than $HfCl_4$ and consequently may be a better charge material than the one presently being used.

RADIOISOTOPIC SEPARATIONS

Uranium-238

Chemical recovery, refinement, and mass spectrometric analyses have been completed on final samples of the recently completed ^{238}U separation series. Table 1 gives the analysis results.

Table 1. Uranium-238 Analysis Results

Sample Designation	Recovered Weight (g)	Mass Assay	
		^{235}U (ppm)	^{238}U (%)
as 271A	904	8.8	99.999
as 271B	330	6	99.999
as 271C	795	10	99.999
as 271D	17	87	99.991

Uranium-235

A ^{235}U enrichment program, which was begun in October 1970, interrupted on December 1, 1970, and resumed in March 1971, has been completed with a monitored equivalent of about 670 g of ^{235}U. Approximately 600 g of ^{235}U containing <0.1% ^{234}U and ≤0.5% ^{236}U will be available as soon as chemical recovery and refinement procedures are completed.

KRYPTON-85 RECOVERY

A sample of ^{85}Kr that has been enriched to 87.5% by ion bombarding a movable aluminum foil in an electromagnetic separator has been recovered for the ORNL Chemistry Division.

The krypton was recovered and simultaneously purified by heating the foil along with a quantity of calcium metal to ~800°C under vacuum. The released gas was allowed to pass through the calcium metal and was then condensed in a container cooled with liquid nitrogen. A crude pressure measurement showed a recovery of 25-30 cm^3, which is an indication that the krypton still retained considerable gaseous impurities.

180° SECTOR ISOTOPE SEPARATOR

The UDC8 equipment, which provides the interface between the PDP-8/I computer and the 180° ORSIS control equipment, is now being installed by the manufacturer. Checkout of the control action of this equipment on the separator functions will begin upon completion of this installation. When satisfactory control action is obtained on each parameter (through computer-controlled rheostats, relays, valves, etc.), the computer programs will be developed to provide computer-assisted 180° ORSIS operation. The programs, hardware, and experience gained with the existing limited control system will be applied to this expanded version.

Separation of rhenium is underway in order to provide ~500 mg of ^{185}Re at >99% for mass spectrometer abundance standards at the U. S. Bureau of Standards. Preliminary assays indicate a mass assay of ~99.5% ^{185}Re.

CHEMICAL PROCESSING

Material in Process

Elements in chemical processing during the quarter ending June 30, 1971, are: Cd, Fe, Gd, Ga, Hf, Hg, Re, Sr, Te, and Sn.

Material Added to Inventory

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

Table 2. Material Added to Inventory

Isotope	Element Weight (g)	Assay (%)	Compound Form
Gadolinium-155	21.186	90.53	Gd_2O_3
Gadolinium-155	26.046	84.39	Gd_2O_3
Gadolinium-157	24.344	90.96	Gd_2O_3
Gadolinium-157	44.583	87.43	Gd_2O_3
Gadolinium-160	86.877	90.10	Gd_2O_3
Gallium-69	122.986	99.46	Ga_2O_3
Gallium-69	107.996	98.83	Ga_2O_3
Gallium-71	59.817	99.26	Ga_2O_3
Gallium-71	59.366	97.34	Ga_2O_3
Gallium-71	9.820	96.42	Ga_2O_3
Iron-57	29.264	91.23	Fe_2O_3
Iron-57	50.981	86.07	Fe_2O_3
Mercury-198	1.943	92.69	HgO
Mercury-198	3.677	82.12	HgO
Mercury-198	4.258	56.30	HgO
Mercury-201	2.974	96.45	HgO
Mercury-201	2.034	93.08	HgO
Mercury-201	1.886	83.55	Hg
Tellurium-125	70.702	95.46	Te
Tellurium-126	161.435	98.36	Te
Tin-118	163.077	95.75	SnO_2

ISOTOPES PREPARATION AND SALES

The radioisotope and stable isotope target and research material preparations for the quarter ending June 30, 1971, are given in Tables 3 and 4.

Table 3. Radioisotope Preparations

Isotope	Number of Preparations	Isotope	Number of Preparations
Americium-241	3	Promethium-147	4
Americium-243	1		
		Radium-226	4
Californium-252	21		
		Thorium-232	21
Carbon-14	2		
		Tritium-Titanium	4
Neptunium-237	23		
		Uranium-233	4
Plutonium-238	5	Uranium-235	50
Plutonium-239	35	Uranium-238	57
Plutonium-240	1		
Plutonium-242	1	Tritium	6

Table 4. Stable Isotope Preparations

Isotope	Number of Preparations	Isotope	Number of Preparations
Barium-136	2	Copper-63	1
Barium-138	1	Copper-65	1
Beryllium-9	3	Erbium-167	1
Boron-10	7	Gadolinium-155	1
Boron-11	1	Gadolinium-158	1
		Gadolinium-160	1
Cadmium-110	1		
Cadmium-112	1	Germanium-70	5
Cadmium-113	1	Germanium-72	2
Cadmium-114	2	Germanium-74	3
Cadmium-116	1	Germanium-76	1
Calcium-40	1	Hafnium-179	1
Calcium-48	1	Hafnium-180	1
Carbon - Normal	2	Iron-54	3
		Iron-57	3
Chromium-50	3	Iron-Beryllium	1

Table 4. Continued

Isotope	Number of Preparations	Isotope	Number of Preparations
Lead-204	1	Silicon-29	1
Lead-206	1	Silicon-30	1
Lead-207	2		
Lead-208	7	Strontium-86	1
		Strontium-88	3
Lithium-6	3		
Lithium-7	12	Tantalum-181	2
Magnesium-24	3	Tellurium-122	1
Magnesium-26	1		
		Thallium-203	2
Molybdenum-92	3	Thallium-205	2
Molybdenum-95	1		
		Tin-112	3
Neodymium-142	2	Tin-114	1
Neodymium-144	1	Tin-118	2
Neodymium-146	1	Tin-120	2
Neodymium-148	1		
		Titanium-46	3
Nickel - Normal	10	Titanium-47	3
Nickel-58	12	Titanium-48	2
Nickel-60	4	Titanium-49	1
		Titanium-50	3
Platinum-194	1		
Platinum-195	1	Tungsten-184	1
Platinum-196	1	Tungsten-186	1
Potassium-41	1	Zirconium - Normal	1
		Zirconium-90	6
Samarium-144	1	Zirconium-91	4
Samarium-154	1	Zirconium-92	2
		Zirconium-94	2
Silicon-28	1	Zirconium-96	2

The isotope sales and services for the quarter ending June 30, 1971, are given in Table 5.

Table 5. Isotope Sales and Services
April 1 Through June 30, 1971

	Shipments	Revenue
EM Isotope Sales	1,005	$258,943
Non-EM Isotope Sales	199	$ 58,579
Services:		
Stable Isotope Target Preparations		$ 46,794
Radioisotope Target Preparations		31,000
Special Services		14,051
Reprocessing Returned Loans		21,185
Work for Others		19,600
Total Services		$132,630
Total	1,204	$450,152

ADMINISTRATIVE

Visitors for the quarter ending June 30, 1971, are shown in Table 6.

Table 6. Visitors

Affiliation	Purpose of Visit
Radionuclides Group, EURATOM Central Bureau of Nuclear Measurements Geel, Belgium	Discuss isotope separations
Kyoto University, Kyoto, Japan	Discuss ion implantation
Arm Control and Disarmament Agency Washington, D. C.	Facility inspection
Centre D'etudes Nucleaires de Saclay Saclay, France	Discuss calutron separations
Ames Laboratory, Ames, Iowa	Discuss isotope separations

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. W. R. Casto
11. J. W. Cleland
12. T. F. Connolly
13. F. L. Culler
14. W. C. Davis
15. J. S. Drury
16. J. L. Fowler
17. J. H Frye, Jr.
18. J. H. Gibbons
19. J. H. Gillette
20. W. F. Good
21. H. R. Gwinn
22. J. A. Harvey
23. R. F. Hibbs
24. J. E. Keeton
25. O. L. Keller
26. Lynda Kern
27. E. H. Kobisk
28. R. H. Lafferty, Jr.
29. E. Lamb
30. R. E. Leuze
31. R. S. Livingston
32. L. O. Love
33. W. S. Lyon
34. F. C. Maienschein
35. F. M. O'Hara
36. J. J. Pinajian
37. W. K. Prater
38. M. E. Ramsey
39. S. A. Reynolds
40-41. R. A. Robinson
42. A. F. Rupp
43. R. W. Schaich
44. M. R. Skidmore
45. A. H. Snell
46. P. H. Stelson
47. H. F. Stringfield
48. D. A. Sundberg
49. A. M. Weinberg
50. J. C. White
51. A. Zucker
52-53. Central Research Library
54. Document Reference Section
55-60. Laboratory Records Department
61. Laboratory Records - RC

EXTERNAL DISTRIBUTION

62. D. Axelrod, National Institute of Health, Bethesda, Maryland
63. N. F. Barr, Office of General Manager, AEC, Washington, D. C.
64. R. M. Brugger, Aerojet Nuclear Corporation, P. O. Box 1845, Idaho Falls, Idaho
65. K. E. Chellis, Florida State University, Tallahassee, Florida
66. J. T. Christy, AEC-RLOO, Richland, Washington
67. D. C. Davis, AEC-ORO, Oak Ridge, Tennessee
68. J. C. Dempsey, DID, AEC, Washington, D. C.
69. B. J. Dropesky, Los Alamos Scientific Lab, Los Alamos, New Mexico
70. E. E. Fowler, DID, AEC, Washington, D. C.
71. L. Friedman, BNL, Upton, New York
72. J. D. Goldstein, AEC, Div. of Biology and Medicine, AEC, Washington, D.C.
73. R. N. Goslin, Oglethorpe University, Atlanta, Georgia
74. K. A. Gschneidner, Jr., Dept. of Metallurgy and Institute for Atomic Research, Iowa State University, Ames, Iowa
75. C. C. Harris, Medical Department, Duke University, Durham, N. C.
76. Max Harris, Lawrence Radiation Laboratory, Livermore, California
77. R. Herber, Rutgers University, New Brunswick, New Jersey
78. I. A. Hobbs, AEC-SRO, Aiken, South Carolina
79. R. W. Hoff, Lawrence Radiation Laboratory, Livermore, California

80. J. W. Irvine, 2-325 Massachusetts Institute of Technology, Cambridge, Massachusetts 02139
81. J. S. Kirby-Smith, Div. of Biology and Medicine, AEC, Washington, D. C.
82. G. A. Kolstad, Div. of Research, AEC, Washington, D. C.
83. J. Lawrence, Lawrence Radiation Laboratory, Berkeley, California
84. J. L. Lerner, ANL, Argonne, Illinois
85. J. S. Lichliter, Lawrence Radiation Laboratory, Berkeley, California
86. J. A. Maddox, DID, AEC, Washington, D. C.
87. W. B. Mann, National Bureau of Standards, Washington, D. C.
88. R. B. Martin, Laboratory and University Division, AEC, ORO
89. S. J. Marsalis, Mississippi State College for Women, Columbus, Mississippi
90. P. W. McDaniel, Division of Research, AEC, Washington, D. C.
91. J. W. Meadows, Lawrence Radiation Laboratory, Livermore, California
92. M. C. Michel, Lawrence Radiation Laboratory, Berkeley, California
93. D. R. Miller, Division of Research, AEC, Washington, D. C.
94. R. A. Neumann, Princeton University, Princeton, New Jersey
95. H. W. Newson, Physics Department, Duke University, Durham, N. C.
96. A. O. C. Nier, University of Minnesota, Minneapolis, Minnesota
97. G. C. Phillips, Rice University, Houston, Texas
98. D. M. Richmond, Div. of Research, AEC, Washington, D. C.
99. R. B. Roberts, Carnegie Institute of Washington, Washington, D. C.
100. G. L. Rogosa, Division of Research, AEC, Washington, D. C.
101. H. M. Roth, Laboratory and University Division, AEC, ORO
102. B. C. Rusche, SRL, Aiken, South Carolina
103. W. D. Sandberg, AEC, Aiken, South Carolina
104-105. J. Spaepen, CBNM-CEA, Geel, Belgium
106. L. G. Stang, BNL, Upton, New York
107. J. A. Swartout, Union Carbide Corporation, New York
108. W. L. Talbert, Jr., Iowa State University, Ames, Iowa
109. G. Taplin, Dept. of Biophysics & Nuclear Medicine, University of California, Los Angeles, California
110. G. R. Tilton, University of California at Santa Barbara, Santa Barbara, California
111-116. D. H. Turno, SRL, Aiken, South Carolina
117. A. R. Van Dyken, Division of Research, AEC, Washington, D. C.
118. L. E. Wahlin, University of Colorado, Boulder, Colorado
119. M. Williams, Lawrence Radiation Laboratory, Livermore, California
120-121. Division of Technical Information Extension