MASTER

15 eyf
RECEIVED BY DTIE MAR 12 1970

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

ORNL - TM - 2889

ISOTOPE PROGRAM (5000) PROGRESS REPORT

FOR QUARTER ENDING DECEMBER 31, 1969

J. H. Gillette

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.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of 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. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

- A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or
- B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

Contract No. W-7405-eng-26

ISOTOPES DEVELOPMENT CENTER

ISOTOPE PROGRAM (5000) PROGRESS REPORT
FOR QUARTER ENDING DECEMBER 31, 1969

J. H. Gillette

Isotopes Division

MARCH 1970

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

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

CONTENTS

	<u>Page</u>
TARGET FABRICATION AND DEVELOPMENT	1
Isotopic Metal Preparation by Reduction-Distillation	1
Chemical Vapor Deposition	1
Preparation of Adherent Urania Deposits by Vacuum Evaporation . .	2
Materials Purification and Alloy Preparation	2
Electron Microscopy Studies of Thin Film Nucleation and Growth .	4
Ion-Implanted Targets	5
180° Sector Separator Targets	5
ISOTOPIIC SEPARATIONS	5
Iron	5
Tin	6
Rhenium	6
Tungsten-180 Second-Pass Separation	7
RADIOISOTOPIIC SEPARATIONS	7
Uranium-233	7
Curium-246	8
Uranium-238	9
SECTOR ISOTOPE SEPARATOR	9
Mercury	9
SILVER COIL REPLACEMENT	9
CHEMISTRY PROCESSING	10
Material in Process	10
Material Added to Inventory	10
Decontamination of Neutron-Irradiated Beryllium	11
ISOTOPES PREPARATION AND SALES	13
ADMINISTRATIVE	14

ISOTOPE PROGRAM (5000) PROGRESS REPORT
FOR QUARTER ENDING DECEMBER 31, 1969

J. H. Gillette

TARGET FABRICATION AND DEVELOPMENT

Isotopic Metal Preparation by Reduction-Distillation

We have prepared the first (no known prior accomplishment) target from an enriched potassium isotope (potassium-40) as the metallic element by application of the reduction-distillation technique. This method was originally developed for the conversion of metal isotope oxides to their respective metals.¹ The inventory compound form of the potassium isotope is KCl which was converted to the carbonate, dried in vacuum, and finally reduced with lanthanum metal in vacuum of approximately 10^{-6} torr at 1000°C . At this temperature the product metal is distilled from the pressed pellet of the reductant metal and the carbonate; metal product vapors were condensed onto a 1-cm-dia aluminum disk. All reduction-distillation and handling operations were performed in an argon atmosphere glove box (vacuum type) in which the total concentration of reactive gases (O_2 , N_2 , CO , CO_2 , H_2O , etc.) was maintained at less than 1 ppm (based on appearance of material). The condensed film of potassium metal (10 mg total weight) was encapsulated with a Viton O-ring gasketed flanged cover plate having a 0.010-in.-thick aluminum window to protect the highly reactive metal from oxidation. A 75% efficiency in conversion to metal and collection in target film form was achieved. Other isotopes which have been successfully converted to elemental form of high purity (usually less than 500 ppm total impurity) include the rare earths (except cerium and lanthanum), the alkaline earth metals, lithium, and sodium.

Chemical Vapor Deposition

The application of chemical vapor deposition (CVD) to the formation of thick and thin films and bulk forms of the isotopes is being studied experimentally. This technique promises to be effective in producing films and single crystals of separated isotopes of elements whose susceptibility to being deposited by vacuum evaporation techniques or being formed into thin films by mechanical rolling are poor. For example, silicon readily alloys or reacts chemically with crucible materials below or at its melting point and, therefore, is difficult to evaporate and condense into thin film form. Furthermore, the element is refractory and does not yield to mechanical forming techniques such as rolling or extrusion. However, the compound silane (SiH_4) is easily and quantitatively disproportionated into its constituent elements at temperatures in excess of 900°C . Development of this technique to form thick and thin deposits of silicon has proved effective and efficient. Silane is carried in a stream of hydrogen and jetted

¹E. H. Kobisk and W. B. Grisham, Application of Reduction-Distillation Method for Preparing High Purity Rare-Earth Isotope Metals, Mat. Res. Bull. 4: 651-62 (1969).

through a glass nozzle onto an appropriate substrate, which is heated by radiofrequency induction. Silver and gold metal substrates have proved satisfactory for this process and can be readily dissolved to yield self-supported films of silicon having areal density of 1 mg/cm^2 or greater. Development of this technique has been limited to normal assay silane on 1-cm-dia surfaces; development of quantitative conversion of SiO_2 (inventory form of the separated isotopes of silicon) to SiH_4 is now being studied, and apparatus required for conversion is being designed. Chemical vapor deposition techniques using W, Mo, Ru, Os, Ir, Re, Tc, and other refractory elements are also being studied for adaptation to formation of films and other shapes using separated isotopes.

Preparation of Adherent Urania Deposits by Vacuum Evaporation

Evaporation-condensation of UO_2 in a vacuum environment has been used in preparing large fission-chamber plates having a coated area of 185 cm^2 and a coating areal density of $0.5\text{--}4.0 \text{ mg/cm}^2$. Thin copper substrates, 0.0001-in. thick, were used to support these large area UO_2 deposits and were prepared by chemically etching a centered 6-in.-dia section of an 8-in.-dia by 0.010-in.-thick copper disk. Excellent adherence was attained by maintaining the substrate temperature at 300°C during deposition; the coating proved to be swipe-proof under conditions of mild abrasion. Uniformity of each deposit was determined to be less than $\pm 4\%$ of the mean areal density over the entire surface.

To perform this deposition a 4-in.-long, 1/4-in.-dia tungsten rod was machined by electric erosion to form a boat with a depressed section 1/8 in. wide and 3-1/4 in. long. Powdered UO_2 , placed in this depression, was heated to its vaporization temperature (2300°C) by electron bombardment using three filaments (resistively heated) parallel to the horizontal boat. At a potential of 850 V dc, the bombarding electron density was 3.5-4.0 Amp.

Six fission-chamber plates of copper and two of stainless steel having similar diameters were coated. The stainless steel disks had been highly polished and the resultant UO_2 deposit appeared as a black mirror and had excellent adhesion. Flexure of both the copper and stainless steel disks did not cause the deposit to peel or crack until the base metal was ruptured. Storage in the atmosphere and experimental use of these plates over a period of three to four months resulted in no deterioration of adherence or uniformity, indicating that the deposits were of high stability.

Materials Purification and Alloy Preparation

Continued application of the vacuum reduction-distillation technique to prepare small (or fairly large) quantities of high-purity elemental isotopes has successfully produced neptunium-237, americium-241, and curium-244 metal. The properties of these metals differ from those obtained by calcium reduction of the corresponding fluorides. Neptunium, for example, is very much more reactive toward atmospheric gases to the extent of being totally oxidized within one or two minutes after exposure to air. The distilled metal was quite malleable as compared with the brittle nature it exhibits

in bulk form when obtained by calcium reduction. Americium metal was rolled easily into foils and curium exhibited flexure properties characteristic of malleable metals. Rolling of curium metal has not yet been attempted, but will be tried within a short period. Efficiency of the reduction-distillation process was 75-85% for all of these metals when thorium metal was used as the oxide reductant.

Americium metal, produced by the technique noted above, was formed into a 3% alloy with virgin thorium-232 metal by levitation melting and homogenization of the mixture. This technique permitted accurate formulation to $\pm 0.1\%$ of the desired concentration of americium. Pressed disks of this alloy were encapsulated in lucite for use as Mössbauer sources at liquid nitrogen and helium temperatures. Levitation melting, performed by suspending the material between two equal and opposing induction fields, has generally proved a satisfactory method of alloy preparation.² If the alloy components have low vapor pressures (less than 10^{-4} torr) at their melting points, levitation melting is performed in high vacuum. If one or more components exhibit high vapor pressures, an argon, helium, or reductant gas can be used as a blanket to prevent gross loss due to volatilization. An argon atmosphere was employed for the thorium-americium alloy formation to inhibit loss of americium.

It should be noted that the levitation technique is ideally suited for purification of isotope metals, radioactive or stable, that contain gaseous inclusions, thermally unstable compound impurities such as copper oxide in copper, or volatile elemental impurities such as antimony in tin. In each case the impurity may be removed through decomposition or volatilization or both. Thus some metal isotopes that have sufficient impurity to impede cold rolling can be purified by simple vacuum levitation melting; this process inherently does not add any impurities because of the exclusion of a confining crucible during melting. From the viewpoint of removing volatile impurities, levitation melting is akin to zone refining. Zone refining is a versatile technique capable of removing or reducing impurities in a bar of material by moving them to one end of the sample by fractional dissolution in a molten zone which is caused to traverse the bar with time. Recently, zone refining has been used to homogenize and purify crystal bars of silicon-28, titanium isotopes, and zirconium isotopes. Crystal bars of these materials were formed by the Van Arkel-de Boer iodide process. In the cases of titanium and zirconium isotopes, the bars were formed by deposition of the element on a tungsten filament from which the purified metal was cut away. This product was then formed into a filament by extrusion and used in the regrowth of a crystal bar of the same isotope. Zone refining of the resultant bar provided ultra-pure isotope metal having oxygen and nitrogen contents of approximately 50 ppm. Silicon, being too brittle to extrude into wire form, was cracked from the tungsten filament, arc melted, and drop cast into a 1/4-in.-dia rod suitable for zone refining. Isotopes of zirconium purified by these combined techniques have been prepared in massive cylindrical form having dimensions of 2-cm dia by 2-cm height and containing approximately 42 g of each isotope. These samples will be preserved as neutron scattering

²H. L. Adair, Levitation Melting and Americium-Reduction Technique for Preparation of Thorium-Americium Alloys, J. Inorg. Nucl. Chem. (to be published).

standards which can be used by universities and atomic energy research institutions for normalizing experimental data; these represent the first such standards produced in the Isotope Target Laboratory.

Electron Microscopy Studies of Thin Film Nucleation and Growth

Use of the Hitachi HU-11B electron microscope (100 kV) to observe in situ condensation of metal and compound vapors into thin film form has provided some basic information on growth phenomena which can be directly related to isotope target preparation. Sequence photography of the growth of a tin film on a 20- $\mu\text{g}/\text{cm}^2$ amorphous carbon film has indicated that at thicknesses up to 350 Å, the film is three-dimensionally discontinuous with little crystalline structure. The carbon substrate was not heated during vapor condensation and therefore mobility of "crystallites" on the surface was practically nonexistent. Similar results were observed with lead deposits. Presently single crystal substrates of MgO, NaCl, and other materials are being studied together with increased substrate temperature to enhance mobility of the condensate and formation of continuous films. Studies of the structure of evaporated-condensed parting agents on metal substrates are also in process to determine the most effective techniques for obtaining uniform, flat deposits. This type of structure will reduce sharp discontinuities in condensed film structures so that internal defects and film strain can be minimized.

All of these observations are being performed using an ORNL-designed evaporation stage integral with the specimen chamber of the microscope and compatible with the ultrahigh vacuum environment of approximately 10^{-8} torr.³ This stage is capable of evaporating materials at temperatures up to 3000°C using electrical resistance or electron bombardment heating.

The unique vacuum capability and the clean internal environment of the electron microscope make this instrument ideal for studying high-temperature annealing of metals without interference by oxidation of the specimen and/or contamination by hydrocarbon deposits during long-term viewing in the electron beam. Studies of the annealing of neutron radiation-damaged aluminum alloys and pure nickel have been made⁴ at specimen temperatures up to 750°C. At 500°C continuous viewing of the specimen for up to 8 hr has shown the samples to be unoxidized or contaminated, and the void structures caused by radiation damage could be studied continuously. At 750°C, significant oxidation to nickel samples was observed after 30 min of continuous viewing.

³D. N. Braski, A High-Vacuum Evaporation Stage for the Electron Microscope, J. Vac. Sci. Tech. (to be published).

⁴N. H. Packan and D. N. Braski, Annealing of Irradiation-Produced Voids in X8001 Aluminum Alloy, ORNL-TM-2645, Oak Ridge National Laboratory (August 1969).

Ion-Implanted Targets

Accelerator targets for the Washington University were formed by implanting 37-keV ruthenium-100 ions into 50- $\mu\text{g}/\text{cm}^2$ carbon substrate material with an ordinary electron bombardment ion source. The source performed well to yield an isotopic purity of >>99% ruthenium-99 (97% purity was the highest previously achieved).

An ion source, designed primarily for separating radioactive materials, has been modified for use in the ion implantation program. Improvements over the basic ion source are: (1) the water coolant is introduced through the main high-voltage bushing and a second bushing is not necessary, (2) a sliding vacuum air-lock valve is easier to operate than the original ball valve, and (3) the source tube has been reduced in diameter and can be more easily inserted and removed against atmospheric pressure.

180° Sector Separator Targets

Methods for preparing targets in the 180° sector separator (ORSIS) are being perfected and studies show that the sector has several inherent advantages over the calutron in making isotopic targets. The mass dispersion is several times that of the calutron and will yield improved isotopic purities, especially in the high-mass range. Ion deceleration problems are considerably less than those experienced with the calutron, and a simpler, more efficient deceleration receiver has been fabricated for sector implantation work. The receiver permits deposits of highly enriched 40-keV isotopic ion beams directly on any substrate at energies from near 0 to 80 keV. The 0- to 80-keV energy range can be extended to greater than 320 keV at reduced intensities by using multiple-charge ions. The ORSIS beam quality remains excellent from submicroampere beam currents to the present maximum current of 7 mA. A 20- μA sulfur-34 beam has been collected at approximately 300 eV to a density of 10 $\mu\text{g}/\text{cm}^2$ on 50- $\mu\text{g}/\text{cm}^2$ carbon substrates. Future direct-beam deposited targets will be prepared with the 180° ORSIS, rather than the calutron, since it has much more flexibility for target-making activities.

ISOTOPIC SEPARATIONS

Stable isotopic separations of iron, mercury-198, mercury-201, neodymium, and tin are currently underway in the electromagnetic isotope separators. The rhenium isotopic separation in the beta calutrons and the developmental efforts related to the 540° modification in the alpha tanks were terminated in November to prepare for the coil-replacement program. There was a short second-pass separation of tungsten-180 of a developmental nature using a previously used feed material.

Iron

An iron separation, with the primary purpose of collecting iron-57, was started in June 1969 to fill the AEC Division of Research request for 100-g quantities of 85% iron-57. Because of the relatively low isotopic

purity requirement, two-arc units were used; however, the units did not function well and overfeeding was begun to compensate for the unstable operation. Consequently, the total ion output was only approximately 75% of the output established in 1967 and the first control mass analyses averaged only 80% iron-57, approximately 5% below specification. The ion exit slits were reduced from 6 in. to 5 in. in length and the total ion output gradually increased to 150% of the best previous level with mass assays averaging approximately 90% iron-57.

Tin

A separation to collect the tin isotopes was started October 1, 1969, in eight beta-type calutrons of Segment I. Tin (II) sulfide-hydrogen sulfide charge was used because it had successfully replaced the usual tin tetrachloride charge in the previous separation series. Two improvements using the sulfide charge are: (1) preliminary tin-112 mass assays are considerably higher and continue to improve and (2) tin depositions on the ceramic bushings, inherent with the tetrachloride charge material, are eliminated. Higher tin-112 retention is expected at the receiver with the sulfide charge.

The tin tetrachloride (SnCl_4) charge had been used extensively in previous separations because it was a liquid at room temperature and could be easily introduced into the ion source. However, significant problems with corroded leak valves, tin-coated bushings which ultimately led to breakage, low retentions, etc. prompted experiments with a different charge material. When the sulfide (SnS) charge was used in the 1965 separation (Series MQ), retention was 71.7% compared with 26.4% in 1961 (Series KN), which used SnCl_4 charge. The hydrogen sulfide gas was added to the tin sulfide charge because process efficiency was too low; the efficiency ultimately increased from an average of 2.2% for SnS alone to 4.3% for the combination.

Rhenium

The rhenium separation (Series OS) was started September 1969 in the two 24-in. beta calutrons. Since the war-time silver coils were to be exchanged for copper-wound coils, the separation was terminated in November 1969. Although the separation was incomplete (originally scheduled for four additional months), an estimated 92.753 g of rhenium-185 and 159.178 g of rhenium-187 were collected in 2735 innage hours.

The preferred charge material in each of the rhenium isotopic separations to date (CP, 1948; HX, 1956; and LD, 1961) has been rhenium oxide, whether prepared externally as Re_2O_7 or internally by passing oxygen over powdered rhenium located in the charge container. The volatility of Re_2O_7 was too great for satisfactory vapor flow, and troublesome metal deposits in the arc chamber resulted; unfortunately, no better charge form was known. While the technique of oxidizing by passing oxygen over the metal in the source did not eliminate metallic deposits, it did improve vapor control. For this reason, this method was chosen to start the most recent separation.

In an effort to eliminate persistent charge decomposition with subsequent deposits of rhenium metal, internal chlorination was tried using carbon tetrachloride, which has proved to be compatible with other charges. During operation carbon collected on the surface of the metallic charge within a very short time reducing the chlorination to impractical levels. However, this charge system does have some attractive points, such as higher ion output and a slightly higher operating temperature.

The best charge system found to date consists of combining the internal oxidation and internal chlorination systems by passing a mixture of oxygen and carbon tetrachloride over the element while operating the oven and arc regions at relatively low power levels (200 to 300 W). Some advantages of both systems were realized: problems of metallic deposition were essentially eliminated and operational parameters were stabilized and improved in several areas as noted by a comparison of present results with results obtained in earlier separations (Table 1).

Table 1. Rhenium Separations

	Series		
	HX 1956	LD 1961	OS 1969
Feed System	Re ₂ O ₇	Re + O ₂	Re + O ₂ + CCl ₄
Ion Reception, hr	339	1496	2735
Average Total Ion Current, mA	3.3	9.5	13.3
Charge Consumption Rate, g Re per innage hr	2.0	1.3	1.5
Process Efficiency, %	1.2	5.2	6.1

Tungsten-180 Second-Pass Separation

A special 8-hr second pass separation using 2945 mg of tungsten as tungstic oxide enriched to 5.25% in tungsten-180 was made. In an effort to obtain high-purity tungsten-180 with high retention, the tungsten-180 run incorporated a number of innovations (i.e., calcium vapor pumping and a two-part, water-cooled, copper pocket). The recovered sample, 3.37 mg of tungsten-180, had an isotopic purity of 97.0%, which is the highest enrichment we have experienced. Highly enriched tungsten-180 is used to make high specific activity tungsten-181 for Mössbauer sources.

RADIOISOTOPIC SEPARATIONS

The heavy-element isotopic separations program was primarily involved in completing the separation and recovery of greater than 50 g of uranium-233 containing less than 15 ppm of uranium-234. A short curium isotope separation was completed; approximately 15 mg of greater than 90% curium-246 was collected. A separation of uranium-238 is underway with the goal of obtaining about 1 kg of uranium-238 containing approximately 2 ppm of total uranium isotopic impurities.

Uranium-233

A uranium-233 first- and second-pass isotopic separations program, with the objective of obtaining greater than 50 g of high-purity uranium-233 containing less than 15 ppm uranium-234 for use in an AEC program at Knolls

Atomic Power Laboratory, was completed during the last quarter of 1969. Samples that were collected and recovered are given in Table 2.

Table 2. Calutron-Separated Uranium-233

Batch Number	Weight of U, g	Mass Analysis, at. %				
		²³³ U	²³⁴ U	²³⁵ U	²³⁶ U	²³⁸ U
A1 652-2 ^a	2.00 ^b	99.991	0.0057	0.0008	0.0002	0.0022
FU-233-3 ^a	3.72	99.975	0.0107	0.0021	0.0011	0.0110
	96.76	99.99	0.0071	0.0009	0.0002	0.0019
A1 240A ^c	12.961	99.9986	3.1 ^d	0.4 ^d	9.4 ^d	0.8 ^d
A1 240B ^c	50.000	99.996	10 ^d	2.5 ^d	4.7 ^d	20 ^d

^aFirst-pass products.

^bTransferred to SRL.

^cSecond-pass products.

^dParts per million.

The most time-consuming problem in completing a separation of this kind, where there is a need to reduce specific impurities to very low levels, is in providing a sufficient feed bank that will permit the recovery of the required amount of material with a minimum number of recycles and yet give the stated isotopic specifications. In order to complete this separation, it was necessary to process all (greater than 641 g) of the highest quality uranium-233 feed available (i.e., 0.8 ppm uranium-232 and 0.184% uranium-234) in order to add approximately 100 g of enriched uranium to the existing feed bank (approximately 114 g). These two batches, both resulting from first-pass separations, were rerun through the separators, with appropriate recycle operations, to obtain approximately 63 g of uranium-233 of required quality.

A 50-g sample of this uranium-233 is scheduled to be blended with similar material (Knolls) from a previous collection; the combined samples will be converted to UO₂ and subsequently used in neutron cross-section measurements where the initial uranium-234 content is critical.

Curium-246

A curium isotope separation was made to obtain 15 mg of curium-246 for a possible second-pass separation. The 1274 mg of curium feed, in the form of curium oxide containing 16.13% curium-246, was obtained by irradiating plutonium-242 (made at Savannah River Laboratory) in the HFIR for the equivalent of 600 days at full power. The curium was separated in the ORNL Transuranium Processing Plant. Table 3 summarizes the enriched products obtained from the separation.

Table 3. Curium Series am Separation

Batch Number	Isotope Enriched	Curium Collected, mg	Sample Composition, at. %				
			²⁴⁴ Cm	²⁴⁵ Cm	²⁴⁶ Cm	²⁴⁷ Cm	²⁴⁸ Cm
am-2	Curium-245	2.93	59.84	20.62	19.12	0.187	0.235
am-2-A	Curium-246	11.91	4.36	0.107	95.37	0.083	0.081
am-2-B		2.08	11.41	0.238	87.72	0.260	0.372
am-2	Curium-247	1.15	34.77	0.530	31.09	30.01	3.60
am-2	Curium-248	1.31	29.00	0.357	14.68	1.23	54.73

Uranium-238

Four calutrons are being used to separate 1000 g of uranium-238 containing approximately 2 ppm uranium-235 for inventory replenishment. The feed for this separation consists of a few kilograms (from the early 1940 era) of calutron-separated uranium-238. The volatile charge compound for ion production is obtained by passing carbon tetrachloride from an external source over uranium dioxide loaded into a charge bottle. Uranium dioxide is preferred for chlorination and is prepared in the laboratory by heating a mixture of U₃O₈ and carbon powder at 1100°C in a stream of nitrogen.

SECTOR ISOTOPE SEPARATOR

Mercury

Experimental runs to develop techniques and equipment for collecting highly enriched isotopes continue in the 180° ORSIS. A few short mercury runs yielded a total of 41.2 mg of 97.6% mercury-198 and 8.5 mg of 99.0% mercury-198. Collections made on an Octoil-S coated, water-cooled, rotating cylinder show increased retention of the mercury isotopes. Since the ORSIS' dispersion between masses for mercury isotopes is 7.5 times greater than the calutrons, it should be possible to use several oil-coated rotating cylinders in combination with oil-coated regular pockets to collect all mercury isotopes simultaneously. It is thought that isotopic contamination for all mercury isotopes will be reduced by as much as 30 times if this procedure is successful. On this basis, mercury-198 would assay ≥98.5% and per-milligram costs would be comparable to calutron costs for mercury separations.

SILVER COIL REPLACEMENT

The last of the silver which was loaned to the Manhattan District Project in the early 1940's for use as current-carrying conductors in electromagnetic separator coils was recalled for return to the Treasury Department. Of the 14,700 tons originally loaned, all but the 67 tons involved in stable isotopes separations have been returned. This remaining silver,

contained in three beta coils and three alpha coils, provided the magnetic field for the four electromagnetic isotope separators which constituted the original electromagnetic facility and, more recently, the beginning of the stable isotopes program.

CHEMISTRY PROCESSING

Material in Process

Elements in chemical processing during the quarter ending December 31, 1969, are: bromine, cadmium, cerium, chromium, iron, mercury, neodymium, nickel, rhenium, antimony, selenium, silicon, tin, and tellurium.

Material Added to Inventory

Isotope material released to the Sales Department during the last quarter of calendar year 1969 is given in Table 4.

Table 4. Material Added to Inventory

Isotope	Element Weight, g	Assay %	Compound Form
Bromine-79	175.940	98.61	NaBr
Bromine-81	156.677	97.81	NaBr
Cadmium-113	38.556	96.30	Cd
Cadmium-114	100.390	99.18	Cd
Cadmium-116	24.745	98.07	Cd
Erbium-164	6.707	73.60	Er ₂ O ₃ ^a
	11.475	67.19	Er ₂ O ₃ ^a
Lead-204	15.000	99.73	Pb(NO ₃) ₂
Nickel-61	15.968	92.92	Ni
	14.334	90.40	Ni
Nickel-62	96.107	98.83	Ni
Nickel-64	21.000	97.93	Ni
Selenium-78	102.140	86.80	Se
Selenium-80	245.653	94.46	Se
Selenium-82	56.274	56.48	Se
Strontium-84	0.0094	98.91	Sr(NO ₃) ₂
Sulfur-34	0.166	98.50	S
	17.576	90.00	S

^aThis material has not been added to inventory pending price approval from AEC.

Decontamination of Neutron-Irradiated Beryllium

The enrichment of beryllium-10 in neutron-irradiated beryllium material by means of a calutron is being planned. One of the requirements for the beryllium feed to the calutron is that the radioactivity of the material be reduced to a level such that the beryllium toxicity is a greater hazard than the radioactivity. This radioactivity comes from the nuclides produced through reactions of neutrons with impurities present in the original beryllium material. The major radioactivity is attributable to cobalt-60. The conversion of beryllium to BeCl_2 and its separation from cobalt-60 by a carrier-gas distillation has been selected as the method of decontamination.

Three different methods have been considered for conversion of beryllium to BeCl_2 : (1) SnCl_2 melt-Be reaction, (2) HCl (or Cl_2) gas-Be reaction, and (3) dissolution of beryllium in hydrochloric acid. The first method was chosen in the initial study because the process appeared to be the simplest one.

The purpose of this project is to develop a simple method for decontamination of neutron-irradiated beryllium material with respect to the radioactive impurity cobalt-60. The project objectives are: (1) to develop a method for conversion of beryllium to BeCl_2 with the subsequent separation of BeCl_2 from cobalt and (2) to apply this method to actually decontaminate the neutron-irradiated beryllium material for use as a calutron feed.

Decontamination Process Using SnCl_2 -Be Reaction (Nonradioactive Materials)

Several runs of a simultaneous SnCl_2 -Be reaction and BeCl_2 distillation have been made with the reactant mole ratio Be/SnCl_2 ranging from 0.42 to 1.06. The distillation temperatures were 450 and 500°C. As expected, the purity of the BeCl_2 product improved with an increasing Be/SnCl_2 ratio. The products with high purity obtained in one simple distillation had the following composition:

BeCl_2	94.0-97.4 wt %
SnCl_2	0.56-2.68 wt %
Nickel	0.03-0.07 wt %

The decontamination factor with respect to cobalt was approximately 1000.

Earlier observations of the SnCl_2 -Be reaction (with low Be/SnCl_2 ratios) in a quartz reactor-still indicated that the reaction mixture appeared to consist of three distinct phases: one solid phase (beryllium) and two liquid phases (BeCl_2 plus SnCl_2 in the upper liquid phase and tin in the lower liquid phase). To evaluate kinetics of the SnCl_2 -Be reaction, the upper liquid phase samples were taken at various times and selected samples analyzed for beryllium and tin. The results of the calculations based on the beryllium content of the melt are summarized in Table 5.

Table 5. Composition of SnCl_2 -Be Reaction Mixture and Beryllium Conversion as Functions of Time

Elapsed Time (t), ^a min	Temperature, °C	Be Content of Melt, wt %	Amounts of Reactants Remaining, moles		Cumulative Amount of BeCl ₂ Formed, ^b moles	Conversion of Be, ^b %
			Be	SnCl ₂		
<u>Be/SnCl₂ Mole Ratio = 0.42</u>						
16	245	1.11	0.112	0.417	0.110	49.3
48	300	1.43	0.081	0.386	0.141	63.5
90	290	1.76	0.048	0.353	0.174	78.2
<u>Be/SnCl₂ Mole Ratio = 0.21</u>						
5	265	0.68	0.088	0.920	0.134	60.4
16	305	0.76	0.072	0.904	0.150	67.6
32	385	0.84	0.056	0.888	0.166	74.6
43	415	0.88	0.048	0.880	0.174	78.2
74	415	0.91	0.042	0.874	0.180	80.9

^at = 0 when the reaction begins near the melting point of SnCl_2 .

^bCalculated data.

According to Table 5, the reaction is 50-60% complete in 5-16 min at temperatures near the melting point of SnCl_2 (247°C). Initially, a low Be/ SnCl_2 mole ratio and a high temperature favor the reaction rate. However, these effects seem to become less pronounced toward the final stage of the reaction. A mathematical analysis of the data in Table 5 has indicated that the reaction is probably of the pseudo first-order with respect to the unreacted beryllium.

Although no chemical rate measurements have been made at Be/ SnCl_2 ratios higher than 0.42, the reaction may be reversible as the Be/ SnCl_2 ratio approaches the stoichiometric value. This assumption is based on the amount of tin recovered at the conclusion of BeCl_2 distillation as well as the BeCl_2 product analysis. However, the rate of the forward reaction producing BeCl_2 is so fast that a high conversion to BeCl_2 does not appear to be a problem (see Table 5).

Separation of BeCl_2 from the reaction mixture by distillation is the controlling factor in the entire process. Occasional difficulties encountered in the BeCl_2 distillation have been caused by the failure of the reaction-distillation temperature controller and of the condenser heater which would result in plugging the condenser.

Decontamination Process Based on HCl Gas-Be Reaction (Nonradioactive Materials)

The modification of the glove box to accommodate the HCl gas-Be reaction has been completed. The study on the HCl gas-Be process has started to investigate feasibility of the process in obtaining a BeCl_2 product of greater than 99% purity with a high decontamination factor (greater than 1000) with respect to cobalt.

The initial run has been carried out at 405-475°C using beryllium chips mixed with approximately 3 wt % of cobalt chips. The reactant gas was

4-8 mole % HCl with argon gas as the diluent. The preliminary result seems to indicate that the rate of BeCl_2 production is very low (approximately 1 g per 2-3 hr) in the ranges of temperature and gas composition studied. The reaction-distillation temperature and the flow rate of the mixed gas appear to be the controlling factors in the production of BeCl_2 .

Treatment of Neutron-Irradiated Beryllium

The HFIR beryllium pieces (approximately 470 ppm beryllium-10) from the inner reflector have been cut into 2-6 in. sizes (irregular shapes). The oxide films on these pieces were removed by treatment with a solution containing phosphoric acid and Cr_2O_3 at approximately 80°C for 1 hr. The actual total weight of the defilmed pieces amounts to approximately 1200 g. Further reduction in sizes will be carried out with either a drill press or a hydraulic press.

ISOTOPES PREPARATION AND SALES

The income from 157 stable isotope preparations costing \$28,348 was \$41,066 and the income from 64 radioisotope preparations costing \$48,362 was \$31,539. The radioisotope and stable isotope preparations for the quarter ending December 31, 1969, are given in Tables 6 and 7, respectively.

Table 6. Stable Isotope Preparations

Isotope	Number of Preparations	Isotope	Number of Preparations	Isotope	Number of Preparations
Arsenic-75	1	Lead-206	1	Silicon-28	3
Barium-137	1	Lead-207	1	Silver-109	1
Beryllium-4	4	Lead-208	1	Strontium-88	1
Calcium-44	1	Lithium-7	3	Thorium	2
Calcium-48	4	Potassium-40	1	Tin-116	4
Carbon	5	Magnesium-25	2	Tin-120	1
Cobalt	3	Molybdenum-92	4	Tin-122	1
Cadmium-110	2	Molybdenum-94	1	Tin-124	1
Cadmium-114	1	Molybdenum-96	2	Terbium	1
Chromium	4	Molybdenum-97	1	Titanium-46	9
Chromium-50	1	Molybdenum-98	1	Titanium-47	1
Chromium-52	2	Molybdenum-100	2	Titanium-48	7
Chromium-53	1	Neodymium-142	4	Titanium-49	1
Chromium-54	1	Neodymium-148	1	Titanium-50	6
Dysprosium-161	1	Neodymium-150	2	Titanium	3
Gadolinium-156	1	Nickel-58	17	Vanadium	1
Gadolinium-158	2	Nickel-60	3	Zinc	6
Gadolinium-160	3	Nickel-62	2	Zinc-66	1
Hafnium-180	1	Nickel-64	2	Zinc-68	2
Iron	2	Niobium	2	Zirconium-90	3
Iron-54	1	Platinum-198	1	Zirconium-91	1
Iron-57	2	Scandium	1	Zirconium-92	1
				Zirconium-94	1

Table 7. Radioisotope Preparations

Isotope	Number of Preparations	Isotope	Number of Preparations
Americium-241	1	Plutonium-240	1
Americium-242	2	Plutonium-241	1
Americium-243	2	Plutonium-242	1
Californium-252	15	Promethium-147	2
Curium-244	5	Uranium-233	10
Neptunium-237	3	Uranium-235	12
Plutonium-238	1	Uranium-235/Al alloy	1 ^a
Plutonium-239	2	Tritium-titanium	2
		Tritium-zirconium	3

^a10,000 cm².Table 8. Stable Isotope Sales and Services
October 1 through December 31, 1969

	Shipments	Revenue
EM Isotope Sales	975	\$231,695
Non-EM Isotope Sales	108	50,845
Services:		
Target Preparations		41,066
Special Services		10,673
Reprocessing Returned Loans		18,558
Total Services		70,297
Total Stable Isotopes	1083	\$352,837

The stable isotopes sales and services for the previous quarter are given in Table 8.

ADMINISTRATIVE

Visitors for the quarter ending December 31, 1969, are given in Table 9.

Table 9. Visitors

Visitor	Purpose of Visit
School of Science and Engineering Waseda University, Tokyo, Japan	Discuss ion implantation and silicon epitaxial growth
Institute for Anorganische Chemie der Universitat, Munchen, Germany	Discuss calutron laboratories and electromagnetic separation plant
CENTO, Tehran, Iran	Discuss electromagnetic separations
High Voltage Engineering Corp. Burlington, Massachusetts	Discuss design criteria for ion sources and methods of preparation
	Discuss high-voltage components line, accelerators, magnets, sources, and handling devices
Nuclear Physics & Radioactivity Division, National Physical Research Laboratory, Pretoria South Africa	Discuss isotopes produced with the ORNL 86-Inch Cyclotron, separated isotopes, and new heavy ion isotopes

INTERNAL DISTRIBUTION

1. A. L. Allen	27. R. E. Leuze
2. M. A. Baker	28. R. S. Livingston
3. P. S. Baker	29. L. O. Love
4. E. E. Beauchamp	30. W. S. Lyon
5. W. A. Bell	31. H. G. MacPherson
6. D. S. Billington	32. F. C. Maienschein
7. G. E. Boyd	33. F. M. O'Hara
8. T. A. Butler	34. W. K. Prater
9. F. N. Case	35. J. J. Pinajian
10. J. W. Cleland	36. M. E. Ramsey
11. T. F. Connolly	37. S. A. Reynolds
12. W. C. Davis	38-39. R. A. Robinson
13. J. S. Drury	40. A. F. Rupp
14. J. L. Fowler	41. R. W. Schaich
15. J. H. Frye, Jr.	42. M. R. Skidmore
16. J. H. Gibbons	43. A. H. Snell
17. J. H. Gillette	44. H. F. Stringfield
18. H. R. Gwinn	45. D. A. Sundberg
19. J. A. Harvey	46. A. M. Weinberg
20. R. F. Hibbs	47. J. C. White
21. J. E. Keeton	48. A. Zucker
22. O. L. Keller	49-50. Central Research Library
23. Lynda Kern	51. Document Reference Section
24. E. H. Kobisk	52-57. Laboratory Records Department
25. R. H. Lafferty, Jr.	58. Laboratory Records - RC
26. E. Lamb	

EXTERNAL DISTRIBUTION

59. D. Axelrod, National Institute of Health, Bethesda, Maryland
60. N. F. Barr, Office of General Manager, AEC, Washington, D. C.
61. G. W. Barton, Jr., Lawrence Radiation Laboratory, Livermore, California
62. K. E. Chellis, Florida State University, Tallahassee, Florida
63. J. T. Christy, AEC-RLOO, 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. N. 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.
92. A. O. C. Nier, University of Minnesota, Minneapolis, Minnesota
93. G. C. Phillips, Rice University, Houston, Texas
94. R. B. Roberts, Carnegie Institute of Washington, Washington, D. C.
95. G. L. Rogosa, Division of Research, AEC, Washington, D. C.
96. H. M. Roth, Laboratory and University Division, AEC, ORO
97. B. C. Rusche, SRL, Aiken, South Carolina
98. W. D. Sandberg, AEC, Aiken, South Carolina
- 99-100. J. Spaepen, CBNM-CEA, Geel, Belgium
101. L. G. Stang, BNL, Upton, New York
102. J. A. Swartout, Union Carbide Corporation, New York
103. W. L. Talbert, Jr., Iowa State University, Ames, Iowa
104. G. Taplin, Dept. of Biophysics & Nuclear Medicine, University of California, Los Angeles, California
105. G. R. Tilton, University of California at Santa Barbara, Santa Barbara, California
- 106-111. D. H. Turno, SRL, Aiken, South Carolina
112. A. R. Van Dyken, Division of Research, AEC, Washington, D. C.
113. L. E. Wahlin, University of Colorado, Boulder, Colorado
114. M. Williams, Lawrence Radiation Laboratory, Livermore, California
- 115-129. Division of Technical Information Extension