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ISOTOPES DIVISION

QUARTERLY REPORT

January - March 1961

By

J. H. Gillette

Prepared from reports by:

P. S. Baker
E. E. Beauchamp
Eugene Lamb
L. O. Love

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

APPLIED ISOTOPES DEVELOPMENT

Routine Processing

Carbon-14

The new large-scale equipment was tested during this period. Several difficulties were found and corrected. For example, the stainless steel lines contained organic matter, such as oil, which would contribute dead carbon and cause a reduction in the specific activity of the product. When these exigencies had been corrected, it was found that the equipment would operate as expected. An eight-curie run giving product with a 70% C^{14} ratio was made. This run demonstrated that the equipment can be expected to produce up to 20 curies of C^{14} in one run.

Cesium-137

Fifty sources were shipped containing a total of 8,174 curies of Cs^{137}. Thirty-nine sources were fabricated during the quarter.

Cobalt-60

Eight shipments were prepared which contained a total of 75,000 curies of Co^{60}. Notable among these shipments was the 50,000 curies prepared in 12 capsules for the National Bureau of Standards for use in an irradiator.

The Co^{60} storage garden and irradiator at the end of the quarter contained ~200,000 curies of Co^{60} which provided a gamma flux of 2.0×10^6 r/hr in the irradiation chamber. It should be noted that this facility is being used almost continuously by personnel of the Laboratory engaged in research, as well as by some outside investigators who need the services of this facility which has no known counterpart in size and scope in the United States.

Iodine-131

A total of 913 curies of I^{131} was produced during this period. A charcoal filter was installed in the cell ventilation system of this facility.

Iridium-192

A total of 2,155 curies of Ir^{192} was contained in the 76 sources which were assayed, decontaminated, and prepared for shipment during this period.

Twenty-two cans containing decayed iridium sources and one can containing inactive iridium sources were prepared for irradiation.

Krypton-85

No purification runs were made during this period. However, there was an adequate amount of Kr^{85} held in intermediate storage to more than satisfy the demands for shipment.

Phosphorus-32

A total of 131 curies was produced during the quarter. The processing of P^{32} continued to be done on a temporary basis in manipulator cells. The preliminary studies for the design of a fixed equipment facility for this processing indicated that the cost would exceed the $20,000 allotted for this equipment. Further studies are being made in an attempt to reduce the cost of the proposed facility.

Technetium-99

A facility for weighing, packaging, and storing Tc^{99} was installed in a semi-hot hood in Building 3032.

Units Prepared for Irradiation and Processed

Two hundred eighty-nine units were prepared for irradiation in the ORR, LITR, and OGR. One hundred fifty-eight units were opened, processed, and product batches prepared.

Radioisotope Packaging and Shipping

Containers Loaded	Number	Percent
Returnable	374	11.4
Non-Returnable	2,834	86.4
Customer	73	2.2
	3,281	100.0

A new system for making shipments was installed in which any radioisotope normally in stock can be shipped on any day of the week. Any order received by the Shipping Department by noon will be shipped on the morning of the succeeding day.

The revision of the shipping procedure entails more work since the pipette for dispensing the products must be changed many times each day. As many as 40 radioisotopes have been shipped in one day. This revision of procedure provides a service that should satisfy the demands of most of our customers.

Two clutch-type decappers were installed which provide a pre-set torque in the capping of bottles. This should provide bottles that the customer can open, yet reduce the possibility of caps not being turned tight enough, thereby allowing "leakers."

Miscellaneous Operations

Twenty-nine irradiation cans were opened and the contents repackaged for shipment.

One Pa^{231} and four Am^{241} samples were weighed, packaged, and prepared for shipment in the alpha handling and dispensing facility.

Short-lived fission products continue to be produced in a manipulator cell rather than in the permanent equipment in Building 3028 which is being refurbished. In spite of this handicap, these products are being produced on a schedule which

satisfies the major demands as well as or better than in the past.

Building 3506 process was finished during the period and the equipment is in stand-by condition.

Non-Routine Processing

Carbon-14

A 100-mc batch of low specific activity C^{14} was diluted by blending with 40 g of $BaCO_3$. The blending was done using a Waring blender. The uniformity of the product was adequate to pursue this technique further when trying to blend several batches of C^{14} of varying specific activities to produce a salable product. Previously the materials were gasified, scrubbed, and reprecipitated. The Waring blender technique, if successful, could reduce the time and resultant cost of this operation.

Sulfite-Free Iodine-131

Nine batches were made containing a total of 45 mc of I^{131}.

Elemental Iodine-131

Fifteen products of elemental iodine tagged with I^{131} were prepared. The products contained a total of 37 mc of I^{131}.

Iodine-125

Two cyclotron targets were processed for I^{125}. Both targets were bombarded by protons; however, one target was normal tellurium and the other was 95.4% enriched Te^{126}. The ratio of I^{125} to I^{126} was increased from ~2 to 1 to ~6 to 1 (at analysis time) using the enriched target.

Iron-55 Sources

Two sources were made; one source contained 50 μc and the other contained 2 mc of Fe^{55}.

Lead-204

A batch of Tl^{204} was milked to provide Pb^{204} for the stable isotope inventory.

Promethium-147

A six-curie batch of $Pm_2^{147}O_3$ powder was weighed, packaged, and prepared for shipment.

Strontium-90 Sources

Fifteen sources were made containing a total of 4.8 mc of Sr^{90}. The sources were made of SrF_2 sealed in stainless steel by silver braze. These sources were made to provide radiation to demonstrate the use of the new pocket dosimeter warning device developed at ORNL.

Tellurium-125m

The need for a sealed point source emitting a 0.03-Mev gamma ray originating in a metallic element resulted from studies of the Doppler effect being conducted by the Laboratory. Tellurium-125m was chosen as the activity since it has a 0.035-Mev gamma, a 58-day half-life, and is readily converted to the metal. It was produced by the nuclear reaction

$$Sn^{124}(n,\gamma)\,Sn^{125}\,(10\ d)\,\overset{\beta-}{}\,Sb^{125}\,(2.7\ y)\,\overset{\beta-}{}\,Te^{125m}.$$

Repeated chemical processing was necessary to obtain Te^{125m} activity having a radiochemical purity > 99.999%.

A source containing ~ 20 mc of this activity was made by reducing the tellurium to the metal and then drying and sealing it under 0.5 atmosphere of helium in a quartz ampoule 1 mm i.d. by 2.5 cm long and with 1 mm wall thickness. The radiation intensity at 3 inches was ~ 800 mr/hr.

Miscellaneous Sources

Seven sources, one each of Zn^{65}, Cs^{137}, Se^{75}, Cr^{51}, Hg^{203}, W^{185}, and Tm^{170}, were made by evaporating the activity to dryness in stainless steel capsules and sealing by silver braze.

Cesium-137 - Special Preparation

A 500-curie batch of Cs^{137} was processed to produce low solubility ceramic pellets. The preparation was made in accordance with a technique specified by the customer.

FISSION PRODUCT DEVELOPMENT LABORATORY

Revisions and Maintenance

The decapper assembly for unloading the HEW filter cask is scheduled for completion on April 15, 1961. The final testing and adjusting will be completed when the HEW cask arrives at the Fission Product Development Laboratory.

The off-gas chilled caustic scrubber system was placed in service and the scrubbing solution temperature was set at 10°C. The initial data indicate the removal of ~25 gallons of moisture per day in the No. 1 scrubber. Activity data are limited but initial results indicate a reduction in the normal discharge activity level by a factor of ten.

The Sr^{90} muffle furnace in Cell 11 failed during the fabrication of test pellets. The cell was decontaminated and the furnace was removed with a minimum of effort on the part of the FPDL personnel. A new furnace is scheduled for installation on April 10, 1961.

Since Cell 11 was decontaminated, the decision was made to repair the leaking lead glass window. The repair work should be completed by April 10, 1961.

The CPFF contractor started construction of the Sr^{90} purification Cell 27 and the estimated completion percentage is 6 as of March 30, 1961.

Cells 2 and 5 were decontaminated in 24 hours to repair "frozen" skimmer arms on the centrifuges.

Shielded Transfer Tanks

STT No.	Date Shipped ORNL	Date Received HEW	Status
RDC-43	-	-	Empty
RDC-44 (Decalso)	2-10-61	2-20-61	Loaded with Cs^{137}
RDC-45 (Decalso)	2-6-61	2-20-61	Loaded with Cs^{137}
RDC-46 (Decalso)	11-18-60	11-25-60	Needs gussets
RDC-47 (Decalso)	2-10-61	2-20-61	Loaded with Cs^{137}
RDC-48 (Decalso)	2-6-61	2-20-61	Loaded with Cs^{137}

Process Status

Processing of Sr^{90} scavenge material was halted due to the repair work in Cell 11 and Cell 2. The scavenge processing will commence on April 10, 1961.

FPDL In-Process Inventory

Radioisotope	In-Process Kilocuries	Powder Kilocuries	Remarks
Cs^{137}	1.4	-	Estimated
Pm^{147}	21	-	Estimated
Ce^{144}	24	18.9	Estimated
Sr^{90}	7	6.2	Estimated
Ru^{106}	13.9	-	Estimated

ADVANCED ISOTOPES DEVELOPMENT

Process Development

Strontium-90

Solvent Extraction - Effect of Sodium on Organic Feed Preparation. - The Sr^{90} concentrate from the FPDL mainstream operation is used as feed for the final purification step which separates strontium from calcium by solvent extraction. It is anticipated that this feed can contain as much as 50 g/l of sodium and 1 g/l of strontium. Since the production capacity of the strontium-calcium separation process is affected equally by the sodium and strontium concentrations, failure to remove the sodium prior to the strontium-calcium separation would result in a proportional reduction in Sr^{90} production capacity.

The effect of sodium concentration on the batch separation of strontium and sodium was investigated for a typical strontium, calcium, and barium feed. The amounts

of strontium and sodium extracted by 1.66 M D2EHPA in Amsco at O/A = 0.4 are shown in Table 1.

Table 1. - Distribution of Sr and Na Between D2EHPA Organic and Aqueous Phases

Initial Sodium Concentration (Molar)	Strontium Extracted (%)	Sodium Extracted (%)	Weight Ratio Sr/Na in the Organic Phase
1	95.2	7.0	0.48
2	88.9	5.0	0.31
3	82.8	3.8	0.25
4	74.7	3.2	0.20

Following the forward extraction, the organic phase was washed with water adjusted to various acidities to back extract Na^+ in preference to Sr^{++}. A pH of 3.5 was found to be optimum. With O/A = 0.4, the strontium loss was ~1.2% and the sodium removal was ~90%.

Table 2 shows the final composition of the organic phase after one forward extraction of the Sr-Na solution followed by one aqueous back extraction with weak (pH 3.5) HNO_3.

Table 2. - Composition of Organic Phase After One Forward and One Back Extraction

Initial Sodium Concentration (Molar)	Sr and Na Remaining in the Organic Phase (%)		Weight Ratio Sr/Na in the Organic Phase
	Sr	Na	
1	94.0	0.63	6.5
2	87.7	0.45	4.2
3	81.7	0.34	3.5
4	73.7	0.29	2.8

Extraction of Iron by D2EHPA. - An investigation was made to determine the conditions under which iron could be removed during the preparation of a feed solution for the separation of Sr^{90} from calcium in the FPDL. The process is to be carried out in the batch extraction equipment of Cell 9. Significant loss of Sr^{90} can be avoided if the iron is extracted from solutions of 0.1 M or stronger acid. It is anticipated that the raw solutions will contain from 0.1 to 1.0 M nitric acid.

The distribution coefficients of iron between 1.66 M D2EHPA in Amsco diluent and aqueous nitric acid solutions were determined using Fe^{59} tracer which was given additional treatment to remove any impurities having low distribution coefficients. Distribution coefficients of iron obtained with aqueous phases in the HNO_3 concentration range of 0.1 to 1.0 M were too high for accurate measurement, but exceeded

10^4. The organic phase loading was one gram of iron per liter.

The effect of Fe^{+3} concentration in the organic phase on its distribution coefficient was investigated by contacting 1.66 M D2EHPA with 0.5 M HNO_3. In the lower range of loading, the agitation was continued until the aqueous phase count became negligible. The tests at higher loading were terminated before equilibrium had been established because of the excessively long times involved.

Table 3. - Effect of Loading on the Distribution Coefficient of Iron Between 1.66 M D2EHPA and Nitric Acid

Loading g/l of organic	Kd	Approximate Agitation Time
1	>10,000	30 minutes
2	>10,000	30 minutes
4	9,500	40 minutes
6	>10,000	40 minutes
8	>10,000	30 minutes
12	>10,000	55 minutes
16	870	165 minutes
20	290	105 minutes
24	220	105 minutes
28	160	105 minutes
32	94	105 minutes

A sample containing 32 g/l iron in 1.66 M D2EHPA and the equilibrated aqueous phase was exposed to 5×10^7 R in the Co^{60} irradiation facility. The sample became amber in color and developed a definite odor but no solid material was formed.

It can be concluded that a very effective separation of iron from fission product solutions is possible when the organic loading is less than 12 g/l of Fe. Based on the observed distribution coefficients for Fe, Sr, and Pm in the HNO_3 concentrations of 0.1 to 1.0 M, the separation factor between Fe and Sr exceeds 10^6 and between Fe and Pm exceeds 5×10^3.

Iron can be removed from D2EHPA by treatment with a strongly basic aqueous solution in which ferric hydroxide is precipitated. Although this reaction produces a clean looking precipitate which settles rapidly to the bottom of the aqueous phase, the formation of this solid necessitates an additional mechanical separation step. Therefore, tests of possible methods of removing iron in a soluble form were made.

The usual method of removing cations from D2EHPA is an extraction by a strong acid. However, tests have shown that useful iron removal cannot be accomplished by nitric acid at concentrations as high as 8.0 M. The extraction of the iron into solutions containing strong complexing agents was chosen for investigation. Tests were performed using Versene Fe-III Specific (a commercial preparation sold in liquid form) and EDTA.

A 66-ml portion of 1.0 M D2EHPA was made by diluting a 1.66 M D2EHPA solution

with TBP. Iron was extracted from 0.5 M nitric acid solution to produce an organic phase iron concentration of 4.0 g/l. The iron was back-extracted into an equal volume of solution containing 90 ml of Versene Fe-III Specific per liter of solution. At an equilibrium pH of 8.4, 98% of the iron was removed. The iron removal was poor at lower pH's. Upon standing at a pH of 8.4, a yellow solid separated from the aqueous phase and a small amount of gas was evolved as a result of decomposition of the complexing agent.

The back extraction of iron from D2EHPA by EDTA was also investigated. The iron was back extracted from a 40 ml sample of the D2EHPA solution described above into an equal volume of aqueous solution containing a 50-g/l concentration of the tetrasodium salt of EDTA. The pH was adjusted by the addition of sodium hydroxide and sodium bicarbonate. The equilibrium pH's and distribution coefficients are given in Table 4. At two points during the test, hydrazine was added because it was believed that this material might reduce the ferric iron to ferrous iron and aid back extraction.

Partial removal of the iron was accomplished. The removal is at a maximum and is approximately constant over the pH range of 6.6 to 8.3. The iron distribution coefficient is not low enough to allow efficient iron removal in a single batch extraction under the conditions of the test. However, efficient removal could be obtained in a multistage countercurrent unit.

The hydrazine is believed to have had little effect. Its addition had no apparent effect on the distribution coefficient and the solutions retained the characteristic red-brown color of trivalent iron throughout the test.

Table 4. - Distribution of Iron Between D2EHPA and Aqueous EDTA Solution

pH	Distribution Coefficient Concentration in Organic Phase ÷ Concentration in Aqueous Phase
5.1	11.7
5.4	2.1*
5.8	0.94
6.6	0.30
8.0	0.36
8.0	0.33
8.3	0.34*

*Hydrazine added before equilibration

Strontium Titanate Pellets. - Work continued on perfecting the mechanical aspects of the pellet fabrication techniques.

Isotope Enrichment by Electromigration

An electromigration tube to measure the enrichment of Cs^{137} from fission product cesium was described in previous reports. The enrichment of Rb^{87} from a natural

mixture of rubidium isotopes was chosen to test the operation of the tube; a 200-hour experiment was carried out. At the end of 102 hours of operation, samples were taken for isotopic analysis. The tube had operated at 66°C for 168 hours by the end of the report period. The average electrode potential was 2,250 volts and the average current was 165 ma.

During the course of the experiment it was necessary to leave the electromigration tube unattended for periods of 4 to 10 hours during which polarization resulting from bubble formation caused a reduced current and a resulting reduction in efficiency of the isotopic separation.

The light, mobile hydroxide ion in this experiment carried more current than the rubidium ion. Since a heavier anion would carry a smaller percent of the total current, an evaluation of rubidium salts with various heavy anions is planned.

Foam Separation

The foam separation process makes use of the tendency of certain substances in solution to concentrate at the liquid-gas interface. A foam (which can be formed by the bubbling of a gas through a liquid) has a very large surface area compared to the bulk liquid. When the foam is removed and allowed to collapse, the resulting liquid contains a higher concentration of surface active material than the original solution.

In the foam separation process for the separation of cations from solution as developed by Radiation Applications, Inc., an anionic foaming agent is added to the solution. The anions of the surface agent concentrate at the surface. Cations of equivalent total charge are held by the surface layer to achieve electrical balance. Since some of the foaming agents are also chelating agents, the cations can be held by chelating action in addition to electrostatic attraction. As a result of this selective action, a single cation can sometimes be preferentially concentrated from a solution containing other cations. Radiation Applications, Inc. has utilized foaming agents which effectively remove strontium and rare earths from solutions containing a high concentration of sodium.

A desirable application of the foam separation process is the removal of Cs^{137} from stored waste solutions which contain a high concentration of sodium nitrate. However, investigators at RAI found no foaming agent which was more than moderately effective for the removal of cesium under these circumstances.

Early in the work at RAI it was discovered incidentally that the enrichment of strontium could be increased by the use of reflux in a foam column. This effect was not pursued, since it did not contribute to the principal object of the work, solution decontamination.

The present work at ORNL is directed primarily toward the multiplication of the comparatively poor cesium enrichment factors by means of foam reflux in a tall column. A foam column 100 mm in diameter and 6-1/2 feet tall is being constructed. It will be possible to measure accurately reflux ratios and all flows into and out of the column.

Exploratory experiments have been performed to determine the foam forming tendency and the distribution coefficients of various substances. Three phenol derivatives - p-octyl phenol, p-nonyl phenol, and p-dodecyl phenol - all exhibited foam forming ability in 0.1 and 1 molar NaOH with the p-nonyl phenol being the best.

To determine the distribution coefficient of cesium in the foaming agent, foam was produced by blowing air through a sintered glass disc into 500 ml of the solution of foaming agent in water contained in a 2-inch diameter glass cylinder. When a foam layer 25 cm high had been produced, the upper portion of the foam was sampled and the ratio of the Cs^{137} activity in the foam (after collapse) to that in the solution was determined. The results are tabulated below.

Table 5

Foaming Agent	Na^+ Concentration (M)	Air Flow (cc/min)	Distribution Coefficient
0.1% p-octyl phenol	0.1	230	1.1
0.02% sodium lauryl sulfate	0.0	76	106
" " " "	0.0	380	72
" " " "	0.1	60	17
" " " "	0.1	190	4
" " " "	0.1	380	1
" " " "	1.0	76	1.1
" " " "	1.0	190	1.2
" " " "	6.0	(no foam formation)	
Derphat 170C	0.01	40	1.1

Iodine-125

Several methods of producing I^{125} are known. The most promising is by the irradiation of Xe^{124}, either in natural or enriched xenon. The reaction is as follows:

$$Xe^{124} \xrightarrow{n,\gamma} Xe^{125} \xrightarrow[18\ h]{E.C.} I^{125} .$$

The data from the experiments in progress will be used to design an in-pile production loop for the continuous production of I^{125}. The use of a loop in which the xenon is circulated provides a means of removing the Xe^{125} from the reactor so that the decay of Xe^{125} to I^{125} takes place in a holdup vessel outside the reactor, thus preventing I^{125} loss by burn-up (σ 1200b) to undesirable I^{126}. If the Xe^{125} is allowed to decay in the reactor, the I^{125} might be adsorbed on the interior of the pipe.

In the initial experiments two tubes containing one gram of 6-14 mesh activated coconut charcoal on which 64 cc of natural xenon had been adsorbed were irradiated for five days in the ORR at a flux of $\sim 2 \times 10^{14}$ n/cm^2/sec. The equilibrium pressure of xenon (T = 25°C) was 10 psia. Each sample yielded ~103 mc of I^{125}.

The first sample was processed by burning the activated carbon. The combustion products were scrubbed to recover the I^{125} and both the residue from the carbon combustion and the aluminum tube in which the samples were irradiated were leached with 0.1 M Na_2SO_3. The I^{125} was distributed as follows:

Scrubber solution	17 mc
Combustion residue	54 mc
Al tube washing	32 mc
Total	103 mc

In the second sample, the activated carbon was leached with 0.1 M Na_2SO_3. The activated carbon was then burned as in the first sample. The following yield was obtained:

Na_2SO_3 solution leach	29 mc
Al tube washing	32 mc
Remaining in carbon	42 mc
Total	103 mc

These results indicate that to recover the iodine in a continuously operating loop, the Xe^{125} must be allowed to decay outside of the neutron flux region.

Rare Earth Separation by Ion Exchange

Using the 0.5% diethylenetriaminepentaacetic acid (DTPA) system described in the last quarterly report for the separation of rare earth elements by ion exchange, 336 curies of Pm^{147} was separated from its daughter Sm^{147} and from fission Nd. The starting material for the separation was 472 curies of Pm^{147} which was 30 - 40% pure.

Results of the analysis to determine the purity of the promethium will be reported next quarter.

The separation was carried out in a manipulator cell illuminated with mercury vapor lamps. An interesting phenomenon was observed when the various fractions from the column were concentrated. Fractions of neodymium, ordinarily lavender pink under normal light, were a blue color under the mercury vapor spectrum. The fractions of promethium, also lavender pink under incandescent light, remained the same color or became slightly more intense. Solutions of neodymium examined under an ultraviolet light showed no color change, indicating another component of the light from the mercury vapor lamps was causing the color change.

About 75 grams of fission 99.99% pure Nd_2O_3 was separated from a solution containing about 50 curies of Pm^{147} and 5 - 10 millicuries of Ce^{144}. This separation was accomplished using a 0.5% DTPA ion-exchange system. Very little Pm^{147} or Ce^{144} remained in the neodymium fractions. The Nd_2O_3 powder measured < 1 mr/hr at contact using a thin window geiger tube instrument.

Fission neodymium is interesting since it has one less isotope and markedly different isotopic abundances than that found in nature. The following compares the mass analyses of both fission and natural neodymium:

Isotope	% Abundance Fission	Natural
142	-	27.7
143	27.7	12.4
144	26.5	23.8
145	19.1	8.3
146	14.9	16.8
148	8.3	5.6
150	3.5	5.4

Electroplating Plutonium on Platinum

Conditions were established for electroplating plutonium on platinum. Plutonium in 2 N nitric acid solution is oxidized to the plutonyl ion with ozone. From 2 N potassium hydroxide solution, plutonium is reduced at the cathode and precipitated as $Pu(OH)_4$.[1]

Upon firing at approximately 300°C the $Pu(OH)_4$ is dehydrated to plutonium dioxide. Oxide films with thicknesses up to 0.3 mg/cm^2 were produced. The current density was adjusted so as to electroplate for one hour.

Electroplating Uranium

Uranium is plated on platinum and stainless steel from 0.16 M ammonium oxalate solution. Uranium is reduced at the cathode and precipitated as $U(OH)_4$. The uranium hydroxide is fired at 300°C to form U_3O_8 oxide. Adherent oxide coatings up to 1 mg/cm^2 have been prepared, and current efforts are being directed to produce coating thicknesses up to 2 mg/cm^2.

Radioactive Source Development

Radioactive sources resistant to leaching can be prepared by adsorbing radioisotope cations on Decalso (an alumino silicate cation exchanger), mixing the Decalso with a flux, and firing to a glass. All cationic radioisotopes tested were found to be strongly adsorbed on Decalso.

Adsorption Properties of Decalso

Cations which were found to be strongly adsorbed on Decalso are cesium, rubidium, calcium, strontium, barium, scandium, titanium, chromium, manganese, iron, cobalt, nickel, copper, zinc, yttrium, zirconium, ruthenium (III), silver, indium, cadmium, the rare earths, gold, mercury, thallium, lead, americium, uranium, and neptunium. Anions did not adsorb on Decalso; however, $P^{32}O_4^{\equiv}$ was adsorbed

(1) Miller, H. W., R. J. Brouns, "Quantitative Electrodeposition of Plutonium," Analytical Chemistry, 24, 536 (1952)

on the zinc form of Decalso at pH 10 with a distribution coefficient of ~30. Other anions have not been tested.

The Decalso used as a source material is first ground to ~325 mesh and the desired amount of material is contacted with the radioisotope solution for from one to two hours. The exchange is complete for tracer concentrations in 30 minutes, but greater concentrations of cations require up to two hours for 90 - 95% adsorption and 24 hours for ~99% adsorption.

Glass Preparation

Decalso, which is 7.5% Na_2O, 15.9% Al_2O_3, 49.5% SiO_2 and 26.1% water, melts at 1450 - 1500°C to a glass. Several compounds were tested as fluxes to lower the melting range and viscosity of the glass. Fluxes tested were B_2O_3, Na_2O, CaF_2 and CaO; however, only CaO gave a glass resistant to leaching. The melting points of Decalso-CaO mixtures are given in Table 6.

Table 6. Melting Points of Decalso-CaO Mixtures

Composition (Percent by Weight)		Melting Point
Decalso	CaO	(Centigrade)
100	0	1400
96.5	3.5	1240
92.5	7.5	1180
90.0	10.0	1160
88.5	11.5	1180
85.0	15.0	1210
80.0	20.0	1230
75.0	25.0	1250
70.0	30.0	1280
67.0	33.0	90
56.5	43.5	1320
53.0	47.0	1400

Since the range 8 - 12% CaO gave the lowest melting glass, 10% CaO was used in the glass prepared for leaching tests. The composition of this glass after firing is 8.8% Na_2O, 18.7% Al_2O_3, 58.1% SiO_2, and 13.1% CaO. The physical properties of this glass have not yet been quantitatively tested. The glass is harder than ordinary plate glass, and more resistant to thermal and mechanical shock.

Leaching Tests

Leaching tests were run at room temperature on 0.1-mc sources of Cs^{137}, Sr^{89}, Tl^{204}, and Ce^{144}. Decalso loadings equivalent to one curie per gram were used in these tests. A Cs^{137} source equivalent to 15 curies per gram was also tested. The percent of the activity leached by various solutions after 68 hours is shown in Table 7.

Table 7. - Leachability of Decalso Glass Sources

Leaching Solution	Percent of Isotope Leached from 1 Cm^2 Source in 68 Hours at 25°C			
	Cs^{137}	Tl^{204}	Sr^{89}	Ce^{144}
Distilled water	$<1 \times 10^{-5}$	$<1 \times 10^{-5}$	$<1 \times 10^{-5}$	$<1 \times 10^{-5}$
Process water	$<1 \times 10^{-5}$	4×10^{-5}	1×10^{-5}	1×10^{-5}
8 M HNO_3	1×10^{-2}	3×10^{-2}	2×10^{-2}	1×10^{-2}
6 M HCl	3×10^{-2}	3×10^{-3}	3×10^{-3}	3×10^{-3}
12 M H_2SO_4	1×10^{-2}	6×10^{-2}	2×10^{-2}	1×10^{-3}
25% HF	7.0	10.0	3.0	4.0
6 M NaOH	2×10^{-2}	1×10^{-3}	3×10^{-2}	1×10^{-2}
4 M NH_4OH	1×10^{-5}	2×10^{-4}	2×10^{-3}	1×10^{-4}
4 M $NaNO_3$	1×10^{-3}	1×10^{-2}	4×10^{-2}	1×10^{-4}
4 M NH_4NO_3	1×10^{-3}	1×10^{-2}	4×10^{-2}	5×10^{-5}
1 M NH_4F	5×10^{-3}	5×10^{-3}	-	-
Simulated sea water	1×10^{-4}	1×10^{-3}	1×10^{-3}	1×10^{-4}

A 400-hour leach of the glass at room temperature removed from 2 - 5 times as much activity as a 68-hour leach. The glass was resistant to all salts, acids, and bases tested except HF. The highly loaded cesium sources (15 c/g) were less resistant to leaching than sources containing small amounts of cesium (1 c/g).

Strontium-89 Source Preparation

A 7.4-mc Sr^{89} source was prepared and 99.9% of the activity was adsorbed on 0.5 gram of Decalso after 24 hours. The Decalso was mixed with 10% CaO and fused at 1300°C in an alundum cup. The thickness of the glass layer was 0.01". There was no detectable loss of Sr^{89} on ignition. The surface of the source smeared 10^3 counts/min. After equilibrating two days with 2 M HNO_3, the surface smeared 8×10^3 counts/min., therefore etching does not reduce the smear count. The smear count can be reduced by fusing a thin layer of glass over the source surface. The observed radiation output was 3.1 r/hr at 20 cm as compared to a calculated value of 3.5 r/hr assuming no self-absorption in the source.

Alpha Sources

Heavy elements such as Am^{241}, $U^{233}O_2$, or $Np^{237}O_2$ ions can be adsorbed on Decalso and used as alpha sources. Thin films of glass of the order of 0.3 - 5.0 micron thickness can be fused to ceramic or stainless steel. A flux of 10% CaO was used with ceramic materials; however, 20% B_2O_3 was used in fusing Decalso to stainless steel because of lower temperature requirements. The 20% B_2O_3-Decalso system melts at 975°C.

The efficiency of the alpha source as a function of the thickness of the glass is show in Table 8.

Table 8. - Efficiency of Am^{241} Alpha Source for Various Thicknesses

1 mg Decalso = 6885 dpm α

Mg Decalso in glass/20.25 Cm^2	Thickness (Calc.) μ	Measured Alpha dpm	Measured Alpha dpm/mg	Efficiency
1.20	0.3	8,088	6,735	.978
2.20	0.6	14,773	6,714	.975
4.60	1.3	30,200	6,564	.953
4.90	1.5	34,280	6,996	1.016

The technique of preparing 0.3 micron thick glass consists of spreading 0.1 mg/cm^2 of finely ground glass mix (80% Decalso, 20% B_2O_3) on the backing material with benzene containing ~1 - 2% Canada Balsam, drying, and fusing at 975°C.

The thin glass surface read 30,000 dpm and smeared 18 dpm with filter paper and 100 dpm with 4/0 Carborundum paper. The glass was resistant to leaching in water and salt solution; however, acids etched the glass.

Technetium

Separation of Technetium

A total of 5,500 grams of technetium concentrate was received in 22 shipments from the Paducah operations. During this period, 2176 grams of 98.5% NH_4TcO_4 and 85.3 grams of Tc metal were added to inventory. The remaining material is in process.

Technetium Chemistry

In the purification of technetium by extraction into lutidine, some lutidine and other unidentified organic residue remain with the aqueous solution of pertechnetate anions following the removal of most of the lutidine by steam distillation. It was found that the presence of the organic contaminants does not interfere with the adsorption of pertechnetate ions on Dowex-1. After washing out the organic contaminants, the TcO_4^- can be eluted with 12 - 16 N HNO_3. This procedure was incorporated in the purification process to precede the final purification by repeated NH_4TcO_4 crystallizations.

Burn-up of Tc^{99} to Ru^{100} by Neutron Irradiation

It was calculated that 0.314 grams of Ru^{100} would be formed by irradiating 9.9 grams of Tc^{99} metal for six months at a flux of 1 x 10^{14} n/cm^2/sec. Two samples of technetium metal (2.1755 g and 2.1956 g) were sealed in quartz and placed in welded aluminum cans for irradiation in the ORR at a flux of 3 x 10^{14}. The first sample will be processed July 30, 1961 (almost four months' irradiation time) and should contain

about 100 milligrams of Ru^{100}. The second sample will be removed at a later date.

Engineering Development

Fission Products Development Laboratory Equipment Modifications

Hot Off-gas Scrubbers. - Two coolers for the scrubber solution recirculation lines were designed, fabricated, and installed for the purpose of reducing the moisture content of the hot off-gas stream. A drain was installed in each of the hot off-gas filter plenums between the roughing filters and the absolute filters.

Closed Loop Process Water System. - A preliminary design proposal for the closed loop system contained provisions for a 500-gallon surge tank, a 100-gallon sample tank plus a recirculating pump located in a pit on the west side of Building 3517, and a cooling unit and cooling tower located above ground. However, measurements indicated flow rates as high as 500 gph in the condensate headers. Although some of this flow is probably the result of leaking service valves between the cooling water and condensate systems, it was apparent that the condensate flow would be too great for the proposed system.

A revised proposal was prepared, which included a new 75-ton chilled water unit connected to operate either in parallel or in series with the existing chilled water unit. The existing chilled water system will be extended so as to service all of the plant process water requirements. The process water return lines will be disconnected from the process waste header and connected to a new header in the service tunnel which will return the water to the chilled water system. The existing process waste header will be used only for steam condensate, which will flow through a radiation monitor to a new 2500-gallon steel tank. The accumulated condensate will normally be pumped into the ORNL process waste system but will be diverted to the hot waste system if radioactive contamination is detected.

Solvent Extraction Equipment, Cells 9 and 10. - Piping changes were made to permit an organic feed to be prepared in Cell 9 and fed to the mixer-settlers in Cell 10. These included the installation of a new air lift from the bottom of batch extractor N-29 to the Cell 10 feed tank, R-510.

Changes in the Cell 10 equipment were made to correct equipment malfunctions observed during the first hot run. Cells 9 and 10 were operated satisfactorily with cold solutions after the revisions had been completed.

Cell 27 - Strontium-90 Handling Cell. - The design concept was changed in order to expedite the completion of the Sr^{90} cell as follows:

1. The stainless steel cell liner was eliminated except for the stainless steel floor pan and tray. The cell walls will be coated with phenol- or vinyl-base paint. Samples of paint will be tested in the cobalt irradiation facility.

2. The armor-plate cell top was eliminated and in its place a 3/4" thick steel plate with lead bricks stacked on top will be used.

Construction of the cell was begun on March 13, 1961.

Preliminary calculations on the heat transfer problems involved in shipping $SrTiO_3$ powder indicate that cans up to 3" inside diameter can be used. The length of the can will depend upon handling problems and probably should not exceed 12". If 2 inches of free space is left in the can, about 15,000 curies of $Sr^{90}TiO_3$ with a powder bulk density of 0.5 g/cc can be shipped in a 3" diameter x 12" long container.

Shielded Filter Casks for the Hot Off-gas and Cell Ventilation Systems. - Design of the Hot Off-gas portion of the project is complete. Preliminary modifications were made to the filter plenum during a recent shutdown. The complete assembly will be installed as soon as schedules will permit.

The preliminary sketches and cost estimate for the Cell Ventilation Filter Cask have been delayed because of high priority work in the Engineering Department.

Manipulator Booting. - The standard boot now being used on the Model 8 manipulators is not entirely satisfactory. The use of a laminate over nylon mesh prevents tearing but the operation of the manipulator is often hampered by the boot, which prevents upward travel. A bellows-type section has been designed and experimental boots will be fabricated and evaluated.

Demineralized Water Unit. - A demineralized water unit was installed for all water which is added to the process. Effluent water from the columns contains 21 - 40 ppm impurities.

Cask Transfer Station - Cell 1. - Assembly of the fluid transfer device is ~95% complete. Piping modifications were made to allow the elution of the HAPO-1A (Decalso insert) cask and the STT in Cell 15. Other piping changes will be required to cool the HAPO-2 (Sr^{90} carbonate) cask with steam and air.

Tank Farm Instrument Cubicle Enclosure. - The estimated cost of an enclosure over the tank farm instrument cubicles and the pipe tunnel valves on the outside north wall of the Fission Products Development Laboratory (FPDL) is $7800.

Shielded Transfer Tanks Modifications

A heated, thermocouple-type liquid level probe was installed in each of the Shielded Transfer Tanks (STT's). A test is being run to determine the life expectancy of the probes which are operated at a higher current (~1.5 amp) than the design current (1 amp) in order to obtain a faster response.

Gondola Car Modifications for Shipping Shielded Transfer Tanks

Two Southern Railway gondola cars were modified to protect the STT's during cross-country shipments. A rigid framework consisting of 3/8" steel bulkheads supported by three gussets and connected with I-beams was installed in the two gondolas used to transport the STT's. The attachment of the STT to the car was strengthened to resist an anticipated maximum acceleration of 51.5 G by replacing the mild steel bolts in the tie-down with high tensile strength steel bolts.

Slurry Filter Casks and Bumpers

The two protective bumpers for the Slurry Filter Casks were completed by the Chicago Bridge and Iron Company and have been shipped to Hanford on two deep-well railway cars.

The first Slurry Filter Cask was completed and has been delivered to Hanford. This cask was equipped with the annular Decalso insert rather than a filter in order to permit its use for strontium shipments.

The cask did not meet specifications with respect to lead bonding, lead density in the cover, and certain external dimensions. The ultrasonic inspection for lead bonding indicated that only about 10% of the outer surface was bonded and 50% of the inner surface. A heat transfer test was conducted in order to determine whether the lack of bond was sufficient to make the cask unsuitable for transporting Ce^{144}. The design allowance for temperature drop across the carrier walls was 37°F with a 5 Kw heat input. An actual temperature drop of 70°F across the cask wall was determined by extrapolating data obtained while heating the interior of the cask with a 5 Kw electric heater. This temperature drop is within the design tolerance and will cause the cake temperature under normal conditions to be 509°F instead of 476°F as originally calculated. The cask was accepted on the basis of its demonstrated heat transfer capability.

The cask walls were radiographed using a 750-curie Co^{60} source and no imperfections were found. Substantial streaming of scattered radiation from the cooling coil lower outlets was observed, but lead plugs can be used to cover these flanges during transit. The cask cover was scanned using a scintillation counter and a 20-curie Co^{60} source. One area around the periphery of the cover and located in the stepped portion of the cover was found to be lacking ~1" of the required 8-3/4" of lead. This cover was accepted on the basis that it is to be used for Sr^{90} shipments requiring only 6-1/2" of lead shielding. Before this cask and cover are used for Ce^{144} shipments, however, the cover will have to be replaced or re-poured.

The cover and shell for the second cask have been rolled and welded but the lead has not been poured. The vendor has prepared additional samples in an attempt to improve his technique for achieving a bond. One sample which was electroplated with a first coat of copper and a second coat of tin was found to be ~95% metallurgically bonded.

Radioisotope Development Laboratory - Building 3047

All of the exterior concrete block has been installed except for a central section left open to permit pouring of the cell block. All of the underground piping has been installed and tested and the cooling tower and associated equipment have been installed. Sealing of the roof has been essentially completed, permitting much of the interior work to proceed independently of the weather.

Some of the stainless steel materials being received from sub-contractors failed the Huey Corrosion tests as performed by the Inspection Engineering group. The most serious failures were in material scheduled for use in cell ventilation and

chemical fume hood ducts. The contractor has submitted a request for a design deviation on this and other materials that have not met specifications. However, ORNL has not concurred with this deviation.

Building 3026-C Modifications

The modifications to Cells 3 and 4 of Bank I have been completed and the cells are now in use. Cell 3 is a chemical processing cell and Cell 4 is equipped with Model 8 Master-slave manipulators.

An additional office will be renovated for use by the Isotopes Division. Studies by the Engineering and Mechanical Division are in progress for renovating and repairing the building and for converting additional cells in the two cell banks into manipulator cells.

Technetium-99 Equipment Modifications

New equipment for the production of Tc^{99} is being designed and installed. The equipment will include new tanks for neutralization of the feed material and chemical make-up, larger extractors (~30 liters), an evaporator for volume reduction and solvent recovery, and a new steam distilling unit.

A hydrogen reduction furnace for reducing ammonium pertechnetate salt to technetium metal has been installed in Cell 4, Cell Block I in Building 3026-C. A mixture of hydrogen and argon flows through a quartz tube containing the compound, then through two water scrubbers, a hydrogen burner, and a CWS filter before it is exhausted to the hot off-gas system.

Replacement of Shielding Glass Damaged by Electrical Discharge

The four-inch-thick pane of non-browning glass and the one-inch-thick cover plate that were damaged by an electrical discharge in November 1960 were removed from the manipulator cell window in Building 3029. Precautions taken to protect personnel from possible electrical shock during the removal operation included protective covers over the glass surfaces and grounding straps on the framing around the glass and on all tools.

The damage to the cover plate and the first four-inch-thick glass piece is described in CF-60-12-13. A close examination revealed that one small discharge had also occurred in the second glass piece from the hot side. The damaged section of glass has been shipped to Corning Glass Works for examination and non-destructive testing.

Containment - Buildings 3027, 3028, and 3029

Containment design of Buildings 3027, 3028, and 3029 has been completed by the architect engineer and minor changes are being made by ORNL design groups.

Charcoal Trap for Iodine-131 Cell Filter Pit

A charcoal trap was designed and installed in the I^{131} cell filter pit. Data are not yet available to determine the efficiency of the trap. As soon as the production schedule permits, an inspection will be made of the filter pit to check on the possible dusting of the charcoal and to determine the activity on the charcoal trap.

Thermal Diffusion Development

Column Operation

The two columns for the enrichment of Ar^{36} have performed as designed. The 36/40 isotopic ratio for column No. 15 is 1.7 and for column No. 17 is 2.0. The concentration of N_2 in each column has continued to build up slowly. This, plus a small leak that developed in the control apparatus during a temporary power failure, brought the concentration of N_2 up to about 40% in each column.

The Ar^{36} production from chemical clean-up of previously separated product includes 3790 mg (weight of elemental Ar) at the 40 - 50% Ar^{36} level and 1976 mg at the 56 - 58% Ar^{36} level.

Wires have been replaced in the Kr^{86} plant and the system is in operation.

Development

Neon-21 Enrichment. - Minor difficulties with the Ne^{21} facility have decreased the production from these columns to values less than that anticipated. A leaky valve at the product sample point on column No. 16 caused a small loss in product and a faulty control on the breathing apparatus delayed the build-up of product in all three columns for several weeks. At present about 2.8 liters (STP) of neon at the 1% Ni^{21} level has been accumulated. This intermediate product represents about 30% of that needed for the second cycle, which will be carried out with the three columns connected in series.

Carbon-13 Enrichment. - Operation and testing continued in the experimental columns for the enrichment of $C^{13}H_4$. Adjustment of the system feed rate and strip-off rate of impurities at the bottom of the cascade at one time produced a C^{13}/C^{12} isotopic ratio of 0.012. However, with no further change in the system, the C^{13}/C^{12} ratio later varied above and below this value. Since this behavior indicated that carbon deposits from the cracking of CH_4 were interfering with the gas transport, the operation was discontinued and the columns were disassembled for examination. About 100 grams of carbon was found deposited at various points in the system.

An experimental test column has been set up with thermocouples attached to the calrod heater to check the sheath temperature at various electrical power levels and to determine CH_4 decomposition rates under operating conditions at various temperatures. A quick preliminary test run with CH_4 in the column indicates considerable decomposition with the heater in the temperature range of 450°C. The decomposition rate increases rapidly with increasing temperature.

Calculations for the C^{13} separation system show that methane in a system using heaters at 400°C will be competitive with carbon monoxide at 600°C, and both of these molecules are greatly superior to carbon dioxide. If the decomposition rate of methane at 400°C is small enough to be tolerated, methane should be used as a feed gas in a low temperature (400°C) system.

Preliminary calculations also indicate that a more efficient plant can be constructed by using smaller annular spacings and higher pressures in the columns. If an annulus

0.125" wide and a pressure of 4.5 atmospheres are used, 0.5 gram per day of 90% C^{13} can be produced in a plant consisting of sixty 7' long columns. This system compares with the preliminary design requiring 62 columns with 0.290" annulus and operating at one atmosphere to produce 60% C^{13} at a rate of 0.45 gram per day. More refined calculations are currently being made by the cascade evaluation group at ORGDP.

Krypton-85 Enrichment. - Preliminary tests were made on the prototype heater for the Kr^{85} separation system. The surface reached an average temperature of 675°C with a variation of ± 5-1/2%. This temperature distribution should be adequate for good operation. A pinhole leak was found in one of the welds on the lead-filled annulus.

The heater was returned to the machine shop for repair of the leaky weld and for assembly with the gas annuli. When completely assembled, the column will be given an operational check with non-radioactive krypton gas and its actual performance will be evaluated.

Column Development. - Coiled wire spacers are now used to hold the heater in the center of the water-cooled tube of the thermal diffusion columns. The cost of fabricating the large number of these spacers required is a considerable portion of the total column cost, and the spot welding operation attaching the spacers to the heaters occasionally causes a leak through the heater sheath. To eliminate these two objections to wire spacers, a machine was constructed to put dimples in the cold tube which accurately position the heater in the center of the tube. A short column will be constructed using these dimple spacers and will be given an operational test. If the cooling effect of these dimples resting on the heater is not too great, a substantial savings in both money and time should result from using dimpled tubes instead of coiled wire spacers.

Xenon-124 Enrichment. - Columns for the Xe^{124} plant are about 50% complete. They will be completed as soon as tubular heaters of the proper wattage are acquired.

Enlarged Facility

The engineering for expansion of the thermal diffusion facilities in Building 9736 has been completed and is now awaiting an estimate by the construction contractor.

ISOTOPE SEPARATIONS

Calutron Operations

Calutron time during this period was allocated to the separation of the isotopes of calcium, nickel, dysprosium, potassium, ytterbium, zirconium, chlorine, magnesium, and tin. The separations of calcium, potassium, zirconium, magnesium, and tin are still in progress.

The distribution of calutron time is presented in Table 9. In Table 10 production data are tabulated for each separation. The values shown in this tabulation are the summation of the entire separation series to date.

Table 9. - Tank Time Allocation

	Hours	Percent
Production (Innage - 76.5% of production)	50,942	95.3
Development	782	1.5
Inter-run	968	1.8
Holding	756	1.4
Tank Time Available	53,448	100.0

Table 10. - Summary of Isotope Separation - Production Data

Element	Series	Process Efficiency (%)	Weight equivalent to monitored ion currents (g)	
Ca*	JW	17.5	Ca-48	7.8040
			Ca-46	0.1328
			Ca-44	79.6529
			Ca-43	5.4802
			Ca-42	23.6005
			Ca-40	5710.5538
				5827.2242
Ni	KE	8.3	Ni-64	16.0299
			Ni-62	48.9899
			Ni-61	16.4728
			Ni-60	338.8138
			Ni-58	1134.4348
				1554.7412
Dy	KF	3.1	Dy-164	147.1653
			Dy-163	29.0327
			Dy-162	131.6869
			Dy-161	21.6820
			Dy-160	11.6859
			Dy-158	0.5556
			Dy-156	0.7999
				342.6083
K*	KI	9.8	K-41	3.3766
			K-40	0.0736
			K-39	60.0460
				63.4962

(continued)

Table 10 - continued

Element	Series	Process Efficiency (%)	Weight equivalent to monitored ion currents	
Yb	KJ	8.4	Yb-176	16.5551
			Yb-174	40.7322
			Yb-173	21.0410
			Yb-172	28.2562
			Yb-171	18.2224
			Yb-170	4.1602
			Yb-168	0.1714
				129.1385
Zr*	KK	4.0	Zr-96	14.1838
			Zr-94	86.2901
			Zr-92	83.0352
			Zr-91	53.9284
			Zr-90	257.9396
				495.3771
Cl	KL	1.0	Cl-37	15.2862
			Cl-35	43.5399
				58.8261
Mg*	KM	11.8	Mg-26	4.7012
			Mg-25	3.9537
			Mg-24	29.4682
				38.1231
Sn*	KN	--	Sn-124	0.7947
			Sn-122	0.6158
			Sn-120	4.2425
			Sn-119	0.9181
			Sn-118	0.4894
			Sn-117	0.7963
			Sn-116	0.2853
			Sn-115	0.0352
			Sn-114	0.0795
			Sn-112	0.0932
				8.3500

*Series incomplete

Stable Isotopes Separation Schedule for Next Quarter

	Building 9731		Building 9204-3		
	XAX	XBX	Track 1	Track 2	Track 3
April	Sm	Mg	Zr	Ca	Sn
May	Sm	Ti	Zr	Ca	Sn
June	Sm	Ti	W	Ca	Sn

Special Services

Special service projects conducted by chemical refinement personnel included conversion of regular inventory forms of enriched isotopes to elements or particular compounds requested by the users, and sealing of inventory forms in ampoules for shipment. Table 11 summarizes these special services.

Ductile, malleable zirconium metal suitable for pressing and rolling into foils for cyclotron studies has been prepared from 4 of the 5 isotopes. The inventory form, zirconium oxide, was first reduced to finely divided metal using calcium chloride and sodium metal for reduction at 850°C. The finely divided metal was reacted with elemental iodine, under vacuum, at a temperature of 225°C in the presence of a hot 7-mil tungsten wire, and the zirconium was allowed to deposit in a pure form by thermal dissociation on the filament. Approximately 75% of the available metal in the dissociation cell can be deposited. The coated wire was pressed into a thin sheet and the tungsten filament removed. By remilling, foils of pure zirconium metal can be made available for cyclotron target work.

Table 11

Element and Isotope	Lot Number	Quantity (g) of Element Desired	Type of Conversion
B^{10}	SS 1 (a) -54	82.5	Boron → H_3BO_3
B^{10}	SS 1 (b)	11.0 total (2 samples)	Boron → H_3BO_3
B^{10}	SS 1(b)	5.0	Boron → B_2O_3
Br^{79}	HG 896 (a)	0.[illegible]00	NH_4Br → $PbBr_2$
Br^{81}	EJ 553 (a)	0.014	AgBr → $PbBr_2$
Br^{81}	HG 897 (a)	0.045	NH_4Br → $PbBr_2$
Cl^{35}	HY 967 (a)	3.0	NaCl → RbCl
Cl^{37}	FW 737 (b)	0.060	NaCl → AgCl
Cr^{50}	IP 1033 (a)	0.750 total (2 samples)	Cr_2O_3 → CrO_3
Cr^{52}	HA 872 (a)	0.750	Cr_2O_3 → CrO_3
Cr^{53}	IP 1035 (a)	0.250	Cr_2O_3 → CrO_3
Li^{6}	SS 5 (h)	124.18 total (5 samples)	Li metal → LiF
Li^{6}	SS 5 (h)	4.0	Li metal → Li_2SO_4
Li^{6}	SS 5 (h)	236.8 total (4 samples)	Li metal → Li_2CO_3
Sn^{119}	LJ 1005 (a)	0.025	Sn metal → SnO_2
Sr^{88}	RS 69 (a)	0.040	$Sr(NO_3)$ → SrO
Cd^{111}	HQ 946(a)	0.008	CdO → Cd
Cr^{50}	GR 836 (a)	0.025	Cr_2O_3 → Cr
Cr^{52}	HA 872 (a)	0.050	Cr_2O_3 → Cr
Cr^{54}	IP 1036 (a)	0.025	Cr_2O_3 → Cr

(Continued)

Table 11 - continued

Element and Isotope	Lot Number	Quantity (g) of Element Desired	Type of Conversion
Fe^{54}	RS 115 (b)	0.500	$Fe_2O_3 \rightarrow Fe$
Fe^{56}	GU 852 (a)	0.200	$Fe_2O_3 \rightarrow Fe$
Fe^{57}	DL 467 (z)	0.120 total (2 samples)	$Fe_2O_3 \rightarrow Fe$
Fe^{57}	RS 117 (a)	0.070	$Fe_2O_3 \rightarrow Fe$
Ge^{70}	JL 1135 (a)	1.0	$GeO_2 \rightarrow Ge$
Hg^{202}	JG 1113 (b)	0.005	$Hg(NO_3)_2 \rightarrow Hg$, encapsulate in Pyrex
Ni^{64}	JJ 1130 (z)	0.293	Homogenize JJ 1130(a), JJ 1130 (brs) and GT 848(a) into one sample of NiO, $NiO \rightarrow Ni$
Pb^{207}	RD-G 1182(a)	0.150	$Pb(NO_3)_2 \rightarrow PbCO_3 \rightarrow PbO \rightarrow Pb$
Pb^{208}	RS 124 (a)	0.070	$PbO \rightarrow Pb$
Sn^{118}	RS 186 (b)	0.600 total (2 samples)	$SnO_2 \rightarrow Sn$
Sn^{118}	RS 186 (b)	0.200	$SnO_2 \rightarrow Sn$, encapsulate in quartz
Sn^{119}	LJ 1005 (a)	0.250	$SnO_2 \rightarrow Sn$
Sn^{120}	LJ 1006 (a)	0.700	$SnO_2 \rightarrow Sn$
Sn^{122}	EC 535 (a)	0.050	$SnO_2 \rightarrow Sn$
Sn^{124}	LJ 1008 (a)	0.050	$SnO_2 \rightarrow Sn$
Ti^{48}	RS 193 (a)	0.100	$TiO_2 \rightarrow Ti$
Zn^{64}	JN 1149 (a)	0.460	$ZnO \rightarrow Zn$
Zn^{68}	HR 954 (c)	0.500	$ZnO \rightarrow Zn$
Pb^{204}	JQ 1167 (a)	27.382	$PbO \rightarrow Pb$, cast in cylinder 0.625" in diameter
Pb^{206}	JV 1186(z)	140.0	$PbO \rightarrow Pb$, cast in cylinder 1-1/8" diameter x 3/4" high
Pb^{208}	JU 1135(a)	141.585	Remelted and cast in cylinder 1-1/8" diameter x 3/4" high
Zr^{90}	RS 198(d)	0.926	$ZrO_2 \rightarrow Zr \rightarrow ZrI_4 \rightarrow Zr$, crystal bar of Zr prepared on 7-mil tungsten wire
Zr^{91}	RS 199(d)	1.130	
Zr^{92}	RS 200 (d)	1.021	
Zr^{94}	KK 1252 (a)	1.163	
Hg^{198}	JG 1109 (a)	0.010	Encapsulated $Hg(NO_3)$ in Pyrex for shipment
Hg^{198}	SS 8 (a)	0.010	Encapsulated $Hg(NO_3)$ in Pyrex for shipment
Hg^{199}	JG 1110 (a)	0.015	Encapsulated $Hg(NO_3)$ in Pyrex for shipment

Table 11 - continued

Element and Isotope	Lot Number	Quantity (g) of Element Desired	Type of Conversion
Hg^{199}	JG 1110(b)	0.020 total (2 samples)	Encapsulated $Hg(NO_3)$ in Pyrex for shipment
Hg^{200}	JG 1111 (a)	0.050 total (2 samples)	Encapsulated $Hg(NO_3)$ in Pyrex for shipment
Hg^{201}	JG 1112 (a)	0.035 total (2 samples)	Encapsulated $Hg(NO_3)$ in Pyrex for shipment
Hg^{201}	JG 1112 (b)	0.010	Encapsulated $Hg(NO_3)$ in Pyrex for shipment
Hg^{202}	JG 1113 (b)	0.010	Encapsulated $Hg(NO_3)$ in Pyrex for shipment
Hg^{204}	JG 1114(a)	0.030 total (2 samples)	Encapsulated $Hg(NO_3)$ in Pyrex for shipment
Hg^{204}	DR 491 (b)	0.020	Encapsulated $Hg(NO_3)$ in Pyrex for shipment

Charge Preparation

Charge materials were supplied for the separation of the isotopes of calcium, dysprosium, lithium, nickel, potassium, tin, zirconium, europium, magnesium, samarium, and ytterbium. Charge materials for calcium, tin, and tungsten were supplied in smaller amounts for exploratory calutron studies.

The chemical recovery of osmium, dysprosium, europium, enriched potassium, and ytterbium from calutron components used in the recent isotopic separations of these elements is continuing. To date, 341 g of salvaged osmium powder has been purified for fabrication into a target for particle accelerators.

The following quantities of charge material have been prepared for use in the calutron program.

Calcium metal	11,385 g
Calcium metal (exploratory)	173 g
$DyCl_3$ (fused and pressed)	36,425 g
KCl (enriched in K^{40})	1,297 g
LiCl (fused)	7,247 g
$NiCl_2$ (fused)	14,324 g
$SnCl_4$	9,649 g
$SnCl_4$ (exploratory)	418 g
WO_3 (exploratory)	105 g
$EuCl_3$ (fused)	697 g
Magnesium metal (massive)	1,116 g
$SmCl_3$ (pressed)	434 g
$YbCl_3$ (fused)	4,711 g

Refinement Chemistry

Chemical recovery and refinement were completed on new inventory lots of Ca^{46}, Ca^{48}, Cd^{110} (2 samples), Cd^{112} (2 samples), Cl^{35}, Cl^{37}, Ir^{191}, K^{39}, K^{40}, K^{41}, Pb^{208}, and Zr^{94}. Refinement also was completed on "mixed" returns of Ti^{47}, Zr^{90}, and Zr^{91}; on current returns of Pb^{110}, Te^{128}, and Ti^{46}; and on target recovery samples of Pb^{204}, Pb^{206}, Pb^{207}, Pb^{208}, Si^{28}, and Ti^{48} (2 samples). Nine new lots of calcium, cadmium, chlorine, iridium, lead, and zirconium, eight current return samples, and four "old loan agreement" samples were prepared for inventory subsequent to refinement and to spectrochemical and mass analyses. In addition, ten samples of lead, silicon, titanium, and zirconium were returned to inventory after being reclaimed and purified from target preparation salvage.

The inventory lots of Cl^{35} and Cl^{37} were both enriched to greater than 99% isotopic purity through a single-pass separation, whereas the Pb^{208} was enriched to greater than 99.9% in a second pass through the electromagnetic separator. Table 12 summarizes these data.

As part of the refinement and development program, 131 individual samples were recovered on a preliminary basis from isotope collectors of graphite and/or copper. The samples were homogenized and partially refined in order to obtain data concerning retention, charge contamination, or mass assay.

Table 12

Isotope	Lot Number	Chemical Form	Quantity of Element(g)	Isotopic Purity Atom %	Chemical Purity*
Cl^{35}	KL 1254(a)	NaCl	2.002	* 35 - 99.58 37 - 0.42	Ag < 0.02 FT Ca 0.01 K 0.02 Li 0.001
Cl^{37}	KL 1255 (a)	NaCl	2.494	* 35 - 0.72 37 - 99.28	Ag < 0.02 FT Al 0.05 Ca 0.05 K 0.01 Li 0.001 Mg < 0.02 T Si 0.05
Pb^{208}	JZ 1198 (b)	$Pb(NO_3)_2$	0.190	**204 - 2 ± 1 ppm 206 - 49.6 ± 1.0 ppm 207 - 58.2 ± 0.6 ppm 208 - < 99.989%	No chemical impurities detected

* Analysis by ORNL Analytical Chemistry Division

** Analysis by ORNL Referee Analyses Laboratory

Special Separations

Stripping and decontamination operations in preparation for the new plutonium chemical processing area were completed. The concrete flooring inside the calutron liner wash area was chipped out to a depth of two inches and the enclosing wooden structure was dismantled and buried. After removing the top layer of concrete, the highest surface measurement indicated less than 30 alpha counts/minute/cm^2. A new two-inch concrete topping was poured over the area.

Chemical purification of depleted uranium, Batch Q1B, was completed. Approximately 9.7 kilograms of purified UO_3 from this batch was converted into green salt and reduced to metal in preparation for rolling into foil.

A special request involving the preparation of 450 alpha sources was completed. This work consisted of adsorbing U^{233} on an inorganic ion exchange media and fusing the complex onto 7" x 3" stainless steel plates. The fused film appeared quite uniform and adherent with a thickness of about one micron. The alpha sources were prepared at specified activity levels ranging from 2.0×10^5 c/m to 1.9×10^6 c/m.

Purification is essentially complete on approximately 35 g of Np^{237} to be used in the fabrication of 300 detector foils for a customer.

The following shipments were made:

Uranium	18
Neptunium	3
Plutonium	1
Thorium	2

Construction Progress

With the exception of the Radioactive Containment Improvements Title II Engineering most time schedules have been met for the renovation and construction projects associated with the Stable Isotope Separations Department.

The Title II Engineering drawings received from Catalytic Construction Company in January have undergone necessary corrections by the Y-12 Engineering Department and completed drawings and specifications are scheduled for submission to the Purchasing Division April 10, 1961.

The containment schedule for renovations and installations, including completion dates, is outlined below.

Job Phase			Starting Date	Completion Date
I.	a.	Coil installation		Complete
	b.	Tank front renovation		Complete
	c.	Yoke steel installation		Complete
	d.	Site preparation		5-1-61
II.	a.	Title II Engineering		4-10-61
	b.	Glove Box Procurement	1-21-61	5-15-61
	c.	LSSC Construction	6-1-61	10-1-61

Job Phase	Starting Date	Completion Date
d. Title III engineering and final testing	6-1-61	11-1-61
e. Work Order completion		11-1-61

Other construction and renovation schedule:

Job Phase	Completion Date
Neutralization System, Bldg. 9204-3	4-8-61
Medium Level Radioactive Laboratory renovation, Bldg. 9204-3	6-1-61
Chemical Refinement Exhaust Replacement, Bldg. 9731	8-1-61

Process Improvement

Isotope Separator Studies - General

Necessary preparations and preliminary calculations required for the construction and operation of the new double-focusing isotope separator are being continued. Calculations are being made to find the optimum shape for the pole tips. Considering the surfaces of the shims as magnetostatic equipotentials, an assignment of potential is made to an array of net points covering the area of interest, subject to the condition that $\nabla^2 F = 0$. The field resulting from a trial pole surface is compared with the desired field shape and the surface then altered to produce the correct field. Since the pole surfaces are limited by the tank volume of the calutron, this type of calculation will allow correction for disturbances produced by physical restrictions imposed on the pole pieces.

Effects due to composition irregularities, saturation effect, distortion of the main field, and the unsymmetric influence of the calutron tank cannot be dealt with in theory. The magnitudes of these irregularities are considered sufficient to produce a harmful effect on the quality of the focused image. In order to be able to detect and correct these faults when they occur, a magnetic field-measuring system is being purchased and assembled. The basis of measurement of this system will be the Hall effect - the voltage that develops perpendicular to field and current when a current is made to flow perpendicular to a magnetic field. The voltage (0 - 100 mv in this case) generated in this manner is proportional to the field strength and current. This signal will be amplified and read by a digital voltmeter. The readings will be punched in paper tape so that an electronic computer can be used to evaluate the data and calculate any necessary corrections to the pole pieces. To reduce the amount of time spent in making the thousands of measurements that will be necessary, the system will operate in a semi-automatic manner. The reading of the Hall voltage, reference voltages, and co-ordinate information will be controlled automatically by the system. This unit will also trigger a relay to advance the measuring mechanism to the next position for a reading.

In order to permit accurate radial and angular positioning, a large wheel is being built on which the Hall probe will be mounted. Construction of a modified source and receiver, necessary because of the 255° focal angle, is also in progress.

Development Chemistry

A study is under way to determine whether zinc isotopes collected in copper receivers can be separated from large quantities of copper by an ion exchange technique similar to the one described for cadmium isotopes in the last quarterly report. The method depends on the adsorption of a zinc chloride complex on an anion exchange resin, the copper ions passing through the column. The zinc is then eluted from the column. Previous refinement systems have depended on the time-consuming removal of copper by electrolysis from strong nitric and sulfuric acid solution.

Tellurium isotopes are normally recovered from collectors in the elemental form and as such are suitable for most nuclear uses. However, this form is not suitable for mass analysis due to polymerization in the vapor state and the consequent formation of multiple charges. It has been found that silver tellurate yields a TeO_2^+ ion from a filament source in sufficient quantities for isotopic measurement. Since the TeO_2^+ is obtained by thermal decomposition of the silver salt at a temperature higher than is used for the oxide, discrimination from background peaks becomes possible.

Silver tellurate is prepared by converting the element to TeO_2, oxidizing the TeO_2 to the tellurate ion with NH_4OH and H_2O_2, and then precipitating the tellurate ion with silver acetate. After centrifuging, the silver tellurate is washed with alcohol and dried.

Isotope Separations Development

Increasing demands for target material of high isotopic purity to be used in nuclear studies have prompted an investigation of the feasibility of preparing such targets directly in the calutron. The advantages are twofold:

1. targets could be prepared in a one-step process (present preparation takes at least two steps),
2. preparation in this manner would result in fewer contaminants.

There are certain effects associated with standard collection techniques, however, which would be undesirable in preparation of good target material. The standard calutron receiver is maintained at ground potential; thus the mono-energetic ion beam strikes the collector with the energy imparted by the accelerating electrodes at the source unit. This energy falls in the range of 35 - 40 Kev and is dissipated principally at the receiver in the form of electromagnetic radiation and kinetic energy which is imparted by the impinging ion to atoms struck in the slowing-down process. Since target specifications are for very uniform depositions of high isotopic purity, these high energy releases are very detrimental to target preparation. Sputtering, as well as thermal evaporation, results in an erosion of the target backing and either no deposition in the area of impact, or the collection of a mixture

of backing material and the isotope. These effects, being functions of ion energy, can be minimized by applying a high potential to the receiver itself and decelerating the ion beam entering the receiver to the desired energy. If an ion can be decelerated to an energy where it has a self-sputtering ratio of less than one, a build-up will occur in the area of impact, thus offering a means for making targets.

TARGET PREPARATION

Development

A considerable amount of effort has been expended in developing procedures and improving equipment for doing target work.

A decelerating receiver was constructed which will satisfactorily reduce ion energy by varying the potential from ground to source. (Fig. 1) A vertical grounded plate with a defining slit for beam entrance is established at the focal point of the incoming beam and the electric field which decelerates the ion beam is maintained between the collector and the plate. The minimum distance between the high voltage collector and grounded plate is one-fourth inch and can be varied. Targets may be inserted in the receiver pocket so that direct deposition occurs, or can be placed in such a way that the material is sputtered onto the target backing. Radial defocusing and cycloidation of the ion beam have not been as great a problem as was originally feared. With separations made so far, the loss of material when varying the receiver potential from ground to source potential has not been more than 10%. The defocusing effect, produced by a protrusion of the electric field through the defining slit, limits the slit width. The cycloidating effect is produced when the particle is slowed down causing a change in radius and a loss of energy. This effect only limits the location of the targets themselves and is not considered to be a great disadvantage.

Separations have been made of Sn^{120}, Ca^{40}, Mg^{24}, Eu^{151}, and B^{10} in this receiver with emphasis on target preparation and sputtering effects at different energies. Target preparation by this method has shown various degrees of success, depending on the element. In general these results have been encouraging and future exploration of this device is being planned as a means for preparing suitable targets.

Figure 2 shows the opened receiver with bottom and top plates.

A method for the preparation of adherent films of suitable uranium compounds on magnesium backing is being studied. The electron beam furnace has been assembled and used, with questionable success, to evaporate iron and chromium. Further studies are being made to develop this method.

Preliminary investigations of methods for measuring the thickness of thin self-supporting films of carbon have been disappointing, but the search is continuing.

The examination of electroplating methods for the preparation of thin self-supporting films of chromium have been made, but only negative results were obtained. Methods for nickel and copper are being investigated concurrently.

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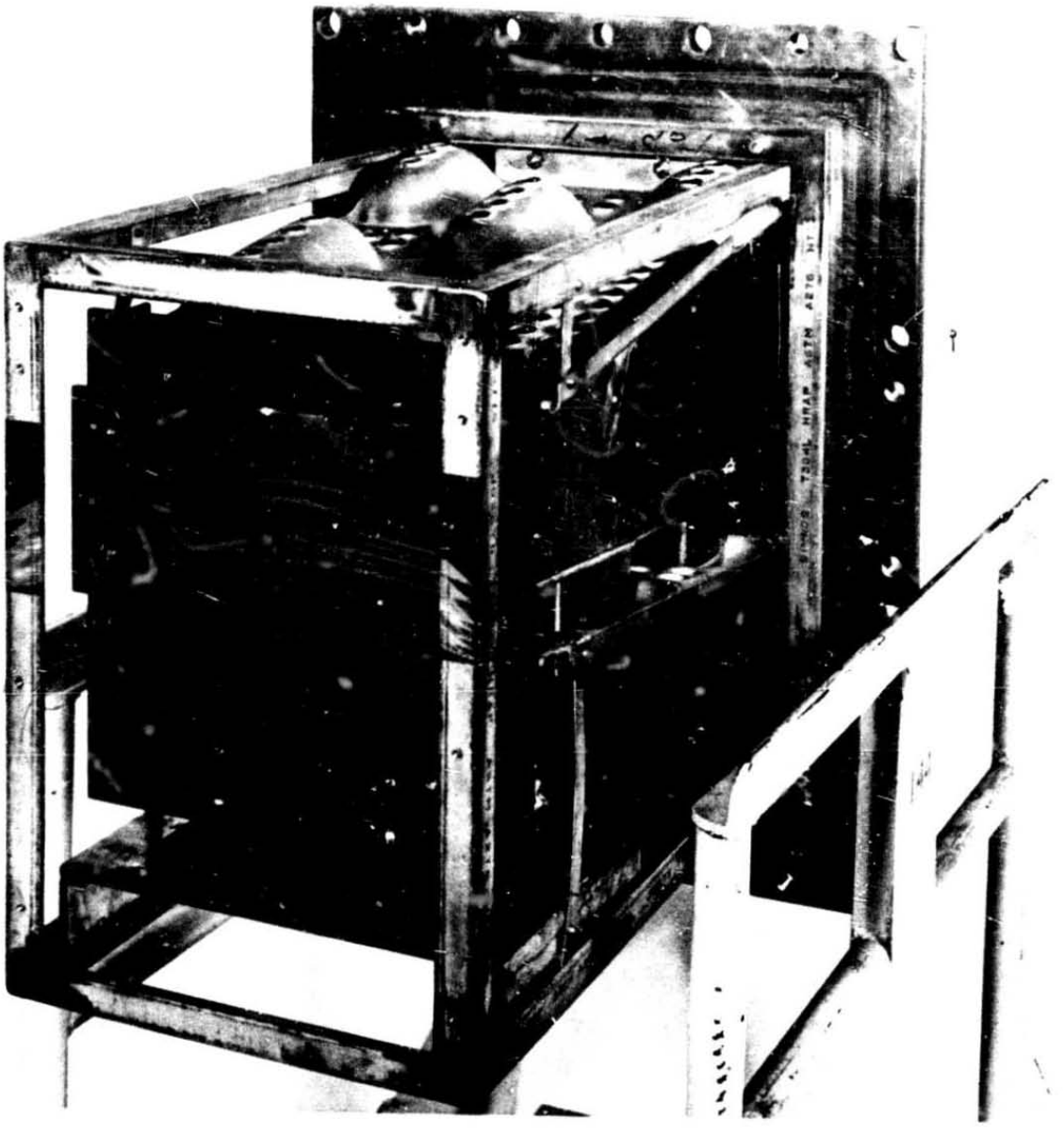

Figure 1 Target Receiver

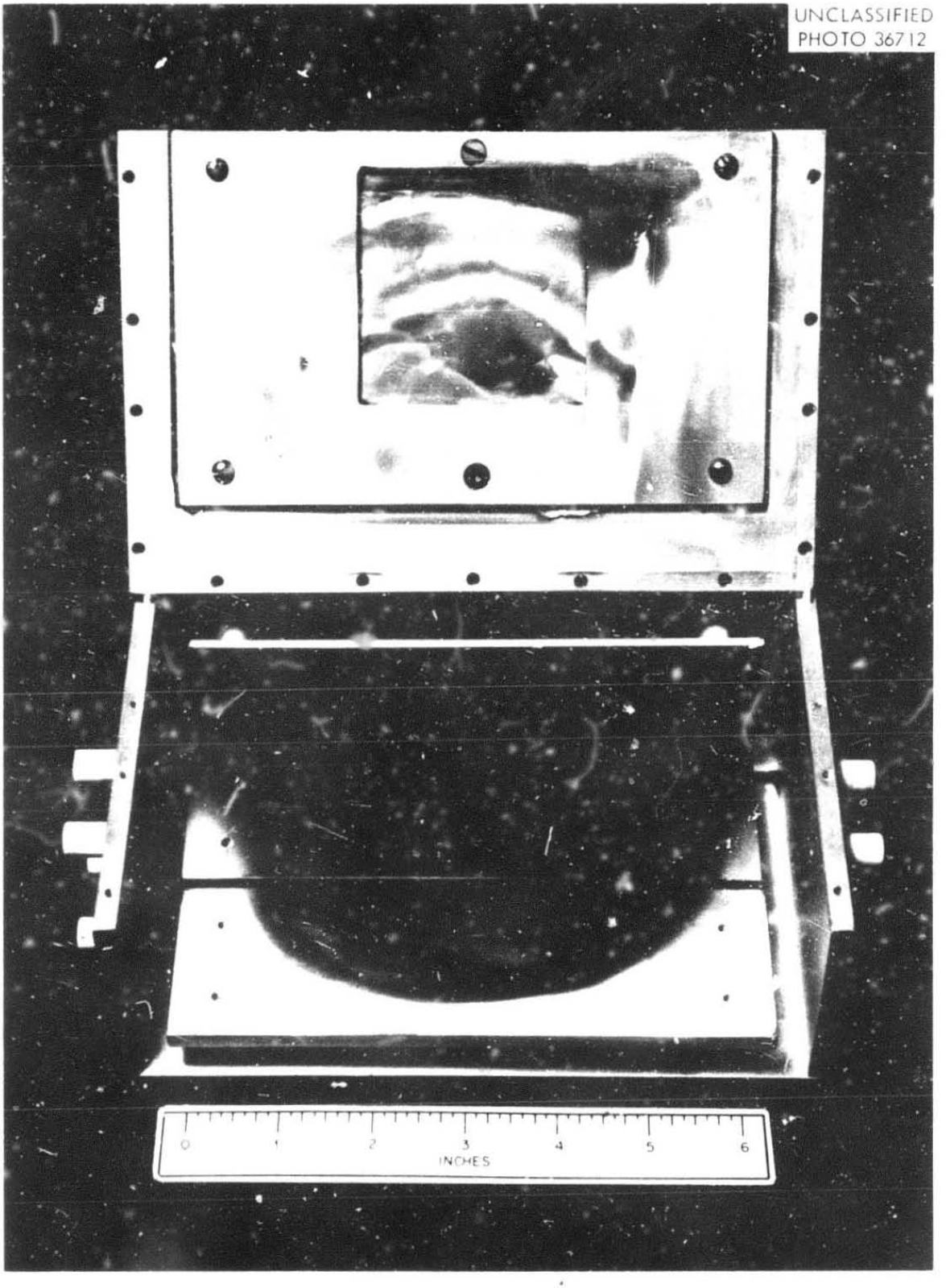

Figure 2 Target Receiver-Open

Fabrication

The preparation of self-supporting lead foils is now a routine procedure. A modification of this method has been used for the preparation of self-supporting copper foils.

Preparation of zirconium foils by rolling is now routine. The technique for rolling zinc and calcium has been investigated and proven successful. Ruthenium was found to be too brittle for rolling.

A die for casting a truncated cone of lithium is being fabricated.

Table 13. - Targets Prepared

Material	No. of Targets	Thickness	Method
Zr^{91}	2	6 mg/cm^2	Rolled
C normal	10	10 μg/cm^2	Evaporated and stripped
C normal	10	25 μg/cm^2	Evaporated and stripped
C normal	10	100 μg/cm^2	Evaporated and stripped
Zn^{64}	2	5 mg/cm^2	Rolled
Zn^{68}	2	5 mg/cm^2	Rolled
Cu^{65}	2	0.00005 in.	Evaporated and stripped
Zr^{92}	2	6 mg/cm^2	Rolled
Pb^{208}	3	4.2 mg/cm^2	Evaporated and stripped
Pb^{208}	2	3.5 mg/cm^2	Evaporated and stripped
Pb^{206}	3	5 mg/cm^2	Evaporated and stripped
Pb^{207}	3	4.5 mg/cm^2	Evaporated and stripped
Mg^{25}	6	30 μg/cm^2	Evaporated and stripped
Pd^{102}	2	10 mg/cm^2	Rolled
Sn^{112}	2	10 mg/cm^2	Rolled
Cr^{50}	4	5 mg/cm^2	Electroplated on gold
Cr^{50}	4	20 mg/cm^2	Electroplated on gold

Miscellaneous Orders

Aluminum was evaporated on plexiglas. Seven uranium oxide films on carbon substrate were prepared.

ISOTOPES SALES AND DISTRIBUTION

Radioisotopes

Table 14. - Radioisotope Sales

	This Quarter	CY to Date
Processed Material	9,268 c	9,268 c
Service Irradiations	188	188
Targets	26	26
Cesium Sources	2,282 c	2,282 c
Cobalt Sources	79,855 c	79,855 c
Iridium Sources	1,976 c	1,976 c

Table 15. - Radioisotope Shipments

	1st Qtr. CY-61	4th Qtr. CY-60	1st Qtr. CY-61	Since August 1946
Processed Material	3,038	2,798	3,099	122,582
Irradiation Units	183	198	291	25,690
Total	3,221	2,996	3,390	148,272
Domestic	1,723	1,611	1,924	
Foreign	222	178	184	
AEC	1,276	1,207	1,282	
Total	3,221	2,996	3,390	

Stable Isotopes

Table 16. Stable Isotope Shipments

	AEC		Domestic		Foreign		Total	
	1st Qtr.		1st Qtr.		1st Qtr.		1st Qtr.	
	1961	1960	1961	1960	1961	1960	1961	1960
SALES								
EM Shipments*	93	75	140	104	166	117	399	296
Lithium	7	9	7	14	10	7	24	30
Total	100	84	147	118	176	124	423	326
Special Orders**	13	19	23	15	32	8	68	42
CAP Uranium			2		3		5	0

(continued)

Table 16 (continued)

	AEC		Domestic		Foreign		Total	
	1st Qtr.		1st Qtr.		1st Qtr.		1st Qtr.	
	1961	1960	1961	1960	1961	1960	1961	1960
LOANS								
EM Shipments*	15	27	8	7	0	3	23	37
Special Orders**	8	3	4	1	0	0	12	4

	AEC	Domestic	Foreign	Total
Total loans under new policy	266	100	36	402
Total returns of new loans	115	75	25	215

*Electromagnetically separated isotopes.
**Included in total shipments.

There are 6 unfilled orders of Cd, Fe, Ir, and Dy.

MISCELLANEOUS

Table 17. - Activation Analyses - SS Accounting

	This Qtr.	Last Qtr.	CY to date	Total Last CY
Activation Analyses completed	32	77	32	292
SS and Special Material:				
Receipts, number of lots	118	104	118	506
Shipments, number of lots	88	83	88	353
Material requests issued	63	53	63	197
Reports issued	41	43	41	172
Internal audits and surveys	19	10	19	55

Table 18. - Cobalt Inventory

Grade c/g	Storage Inventory Curies
0 - 25	62,576
26 - 40	72,542
41 - 55	106,794
>55	12,706
Total	254,618