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RADIOISOTOPE PRODUCTION AND PROCESS DEVELOPMENT

ANNUAL REPORT FOR 1956

H. E. Seagren

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RADIOISOTOPE PRODUCTION AND PROCESS DEVELOPMENT ANNUAL REPORT FOR 1956

H. E. Seagren

Compiled from data by:
E. E. Beauchamp
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RADIOISOTOPE PRODUCTION AND PROCESS DEVELOPMENT ANNUAL REPORT FOR 1956

H. E. Seagren

RADIOISOTOPE PROGRAM SUMMARY

This report summarizes the main radioisotope production and process development activities during the calendar year 1956. There were 13,585 shipments made in CY-1956, a 6.5% increase over the 12,749 shipments made in CY-1955, which was only a slight increase over the 12,585 shipments made in CY-1954. The total amount of radioactivity shipped, however, has continued to rise rapidly – from 28,879 curies in CY-1954 to 49,383 curies in CY-1955 and to 109,500 curies during the past year. Thus the trend toward larger quantities per package is quite pronounced, due primarily to the increasing sales of large Co^{60} sources. The average curie-per-shipment figure for Co^{60} rose from 52 in CY-1955 to 571 in CY-1956, while the actual number of cobalt shipments decreased from 431 in CY-1955 to 164 in CY-1956. Details are given in Tables 1 through 4. Service irradiations in the reactors increased from 785 in CY-1955 to 963 this year; unprocessed irradiation units decreased from 2010 to 1704. Gross income increased 31%, from $1,690,871 to $2,210,033.

Table 1. Radioisotope Sales Distribution

	Calendar Year	
	1956	1955
Foreign	$ 112,364	$ 87,360
Project	63,065	63,320
Domestic	1,750,761	1,323,504
Cancer Program Subsidy*	238,880	196,246
Technical Co-operation Program	97	777
Local Sales to Carbide	40,841	19,349
Civilian Defense	4,025	315
Total	$2,210,033	$1,690,871

*Income subsidized by the AEC. Does not include cash received.

Table 2. Sales and Income of Major Categories

	Calendar Year 1956		
	Income	Shipments	Amount (mc)
Carbon-14	$ 272,211.80	258	9,286
Cesium-137	64,616.86	127	2,861,824
Cobalt-60	885,798.36	164	93,616,415
Iodine-131	190,898.47	4,620	735,467
Phosphorus-32	198,568.98	2,445	164,950
Others	597,938.53	5,971	12,112,075
Total Radioisotopes	$2,210,033.00	13,585	109,500,017
Boron	$47,389.00	43	12,454 g*
Helium-3	$11,600.00	33	30,785 cc*

*This is the accumulated figure as reported by the Accounting Department; the Division figure was not used because it did not take into consideration credits issued.

Table 3. Radioisotope Sales

HSA – High specific activity
LSA – Low specific activity
CF – Carrier-free
E – Enriched target

	Quantity Sold (mc)	
	1956	1955
Antimony-122	21	
Antimony-124, solution, HSA	151	278
Antimony-Be, neutron sources (9 sources)	52,100	29,000
Antimony-125, solution, CF	6	5
Argon-37, gas (4 ampoules)	9	11
Arsenic-76, solution, HSA	28	
Arsenic-77, solution, CF	19	
Barium-131, solution, HSA	5	
Barium-133, solution, HSA	13	9
Barium-140, solution, CF	982	933
Beryllium-7, solution, CF		7
Bismuth-210, solution, HSA	2	16
Bromine-82, solution, HSA	233	
Cadmium-115, solution, HSA	20	21
Cadmium-115*m*, solution, HSA	38	8
Calcium-45, solution, LSA	685	453
Calcium-45, solution, HSA	1,298	1,643
Calcium-45, solution, E	133	70
Calcium-45, solution, CF	8 (μc)	13 (μc)
Carbon-14, dry $BaCO_3$, HSA	9,286	5,595
Carbon-14, sheets	27	
Carbon-14, organic compounds		187
Cerium-141, solution, CF	99	235
Cerium-144, solution, CF	2,344	1,001
Cesium-134, solution, HSA	1,789	2,366
Cesium-137, solution, CF	78,801	34,608
Cesium-137, pelleted CsCl (46 sources)	2,783,022	1,997,000
Chlorine-36, solution, HSA	8.475	4
Chromium-51, solution, HSA	2,926	7,570
Chromium-51, solution, E	1,105	65
Cobalt-57, solution, CF	4	23
Cobalt-58, solution, CF	55	2
Cobalt-60, solution, HSA	1,175	1,401
Cobalt-60, metal (7196 sources)	93,616,415	42,081,000
Copper-64, solution, HSA	3,834	
Europium-152, -154, solution, HSA	76	
Europium-155, solution	11	22
Fission products, solution	890	1,332
Gallium-72, solution, HSA	10	
Gold-198, solution, HSA	36,091	305
Gold-199, solution, CF	29	
Hafnium-181, solution, HSA	97	
Hydrogen-3, gas (171 ampoules)	5,696,712	512,309

Table 3 (continued)

	Quantity Sold (mc)	
	1956	1955
Hydrogen-3–zirconium (112 targets)	150,862	80,000
Indium-114, solution, HSA	90	65
Iodine-129, solution	10.4	4 (mg)
Iodine-131, solution, CF	735,467	668,578
Iridium-192, solution, HSA	361	368
Iridium-192, metal (348 sources)	5,720,822	3,559,000
Iron-55-59, solution, HSA	33	60
Iron-55, solution, E	142	104
Iron-55, metal (1 source)	4	485
Iron-59, solution, E	648	489
Krypton-85, gas, CF	338,935	65,501
Lanthanum-140, solution, HSA	411	
Manganese-54, solution, CF	6	22
Mercury-203, solution, HSA	663	877
Molybdenum-99, solution, HSA	248	
Neodymium-147, solution, CF	7	3
Nickel-63, solution, HSA	77	76
Niobium-95, solution, CF	55	47
Osmium-191, solution, HSA	2	
Palladium-109, solution, HSA	120	
Phosphorus-32, solution, HSA	164,519	157,456
Phosphorus-32, solution, CF	430	668
Potassium-42, solution, HSA	2,757	302
Praseodymium-142, solution, HSA	12	
Praseodymium-143, solution, CF	8	1
Promethium-147, solution, CF	14,156	5,697
Rubidium-86, solution, HSA	2,190	628
Ruthenium-103, solution, CF	74	209
Ruthenium-106, solution, CF	330	717
Samarium-153, solution, HSA	114	
Scandium-46, solution, HSA	20,498	167
Selenium-75, solution, HSA	421	420
Silver-110, solution, HSA	368	672
Silver-111, solution, HSA	31	
Sodium-22, solution		59
Sodium-24, solution, HSA	1,474	1,676
Strontium-85, solution, CF		12
Strontium-89, solution, CF	1,250	1,924
Strontium-90, solution, CF	23,097	129,455
Strontium-90, SrF_2 glaze		9,000
Sulfur-35, solution (sulfate), CF	12,689	14,733
Sulfur-35, solution (sulfide), HSA	345	322
Sulfur-35, solution (elemental), HSA	788	980
Tantalum-182, solution, HSA	161	75
Technetium-99, solution	0.37	1
Thallium-204, solution, HSA	251	1,712
Tin-113, solution, HSA	26	118
Tungsten-185, solution, HSA	95	

Table 3 (continued)

	Quantity Sold (mc)	
	1956	1955
Tungsten-187, solution, HSA	35	126
Yttrium-90, solution, CF	40	
Yttrium-91, solution, CF	940	250
Zinc-65, solution, HSA	1,036	1,640
Zinc-65, solution, CF		13
Zirconium-95, solution, CF	961	1,110
Helium-3, gas (49 ampoules)	30,785 (cc)	5,195 (cc)
Deuterium-zirconium (5 targets)	6.5 (cc)	7 (cc)

Table 4. Irradiations in Other Reactors

	Calendar Year	
	1956	1955
At Hanford		
Antimony		2
Antimony metal	3	1
Calcium carbonate	3	
Cesium chloride	4	2
Cobalt	41	
Iron-54	2	
Iron oxide	9	7
Mercury		3
Mercuric oxide	2	
Potassium chloride	16	32
Silver nitrate	3	
Tantalum foil	3	2
Thallium	1	
Tin	2	
Tungsten		3
Tungsten oxide	4	1
Zinc	2	
At MTR		
Barium chloride	1	3
Cesium chloride		3
Cobalt	32	18
Nickel		5
Silver metal		2
Tin		5
Zinc		5
At SRO		
Cobalt	208	

Major development items were the construction and operation of the cobalt storage garden and irradiator; the successful remote-control sealing of source containers by shielded-arc welding; the development of a simple, efficient method for I^{129} recovery; the removal of the Am^{241} contaminant from Pr^{147}; the chloroplatinate method of Cs^{137} purification and recovery; and the testing of flowsheets for the Fission Products Pilot Plant.

IODINE-131

Iodine-131 continued to lead in the number of shipments made. While the number of shipments made was reduced from 4965 in CY-1955 to 4620 in CY-1956, the total I^{131} activity shipped increased by 10%, from 668,578 mc in CY-1955 to 735,467 mc this year.

The production cost per millicurie sold was reduced from $0.257 in CY-1955 to $0.234 in CY-1956. This unit cost includes packaging, shipping, and selling, which contribute about 45% of the total. The decrease in unit cost was due chiefly to the increase in demand with no increase in operating costs.

PHOSPHORUS-32

The number of shipments of P^{32} decreased in number from 2504 in CY-1955 to 2445 in CY-1956, while the total P^{32} activity shipped increased 4%, from 158,124 mc in CY-1955 to 164,950 mc this year.

The unit cost of production, about 25% of which consists of allocated charges for packing, shipping, and selling, increased this year from $0.84 to $0.96 per millicurie as a result of increased reactor charges.

A method was developed in which irradiated sulfur was distilled, leaving P^{32} with sufficient purity to be dissolved as a product. This procedure has been made feasible by the availability of higher flux densities in the LITR. The ORR will provide even greater flux densities. It is anticipated that 5 to 10 g of sulfur irradiated in the ORR will produce the same quantity of P^{32} as will be produced by 10,000 g irradiated in the ORNL Graphite Reactor.

Phosphorus-32 products prepared by the above method contained no detectable radiochemical or ionic impurities. Total solids in a 2-ml sample were below the limits of detection. A decay study was made, and after seven half lives, no long-lived impurities were detected.

Although the recovery of the pure sulfur for re-irradiation in the reactor was demonstrated, it was found more economical to prepare pure sulfur for each irradiation. By discarding the sulfur, the P^{32} recovery equipment was greatly simplified.

Equipment was designed and placed in operation to purify 8-kg batches of sulfur. A three-week run requiring five actual man-days of operator time will purify enough sulfur to provide a target supply for about two years of ORR P^{32} production.

A half-element sulfur irradiation assembly was designed for irradiation of sulfur in the LITR. The sulfur was sealed in quartz ampoules.

After a six-week irradiation, the tubes were removed from the reactor and found to be broken. The breakage is believed to have been caused by an expansion of the sulfur resulting from a radiation-induced phase change. It is anticipated that this problem will be solved by irradiating the sulfur in re-usable platinum tubes.

Approximately 100 mc of P^{32} was recovered for each gram of LITR-irradiated sulfur.

CARBON-14

Ion chambers were added to the C^{14} system to determine the amount of radiocarbon being generated at any time and the amount being discharged from the system. The chambers will also provide a check on the efficiency of the gas scrubbers. A significant amount of silica was found in the $BaC^{14}O_3$. This was probably due to a reaction between the caustic solution used to absorb $C^{14}O_2$ and the gas column. A stainless steel column to replace the existing glass column has been fabricated but has not yet been operated.

COBALT-60

Sales

A total of 151 shipments of Co^{60} sources containing approximately 93,100 curies was made. This represents a curie increase of 77% over that of the previous year.

A total of approximately 242,000 curies of Co^{60} was received, assayed, segregated according to specific activity and size of pellets, and stored as stock material.

Cobalt Storage Facility and Irradiator

The Co^{60} storage garden and irradiation facility was completed and placed in operation. Several modifications were made after operations were started. A cooling-gas temperature indicator was added, and the irradiator shaft was reworked in order to prevent binding during use.

With approximately 157,000 curies of Co^{60} in storage at the end of the year, the gamma flux as determined by chemical dosimetry was 1.68×10^6 r/hr (see Fig. 1).

IRIDIUM-192

A total of 105 shipments of Ir^{192} containing approximately 5670 curies was made. This represents a curie increase of 65% over that of last year.

STRONTIUM-85

Sixty-five-day Sr^{85} with a radiochemical purity of greater than 99% was produced by LITR neutron irradiation of a strontium nitrate target enriched in Sr^{84}. The cost of producing Sr^{85} by neutron bombardment of enriched Sr^{84} appears to be much less than that of the cyclotron-produced material. Therefore this product should be useful where carrier-free material is not required.

IODINE-129

A simple, rapid, and efficient method for the recovery of I^{129} from tower scrubber units was developed. It was found that ultrasonic vibrations are effective in removing AgI from the BerI saddles that are used in the tower scrubbers. Details were reported in the Operations Division Report for June 1956. A total of 350 mg of high-purity I^{129} was recovered for distribution under the Radioisotope Program.

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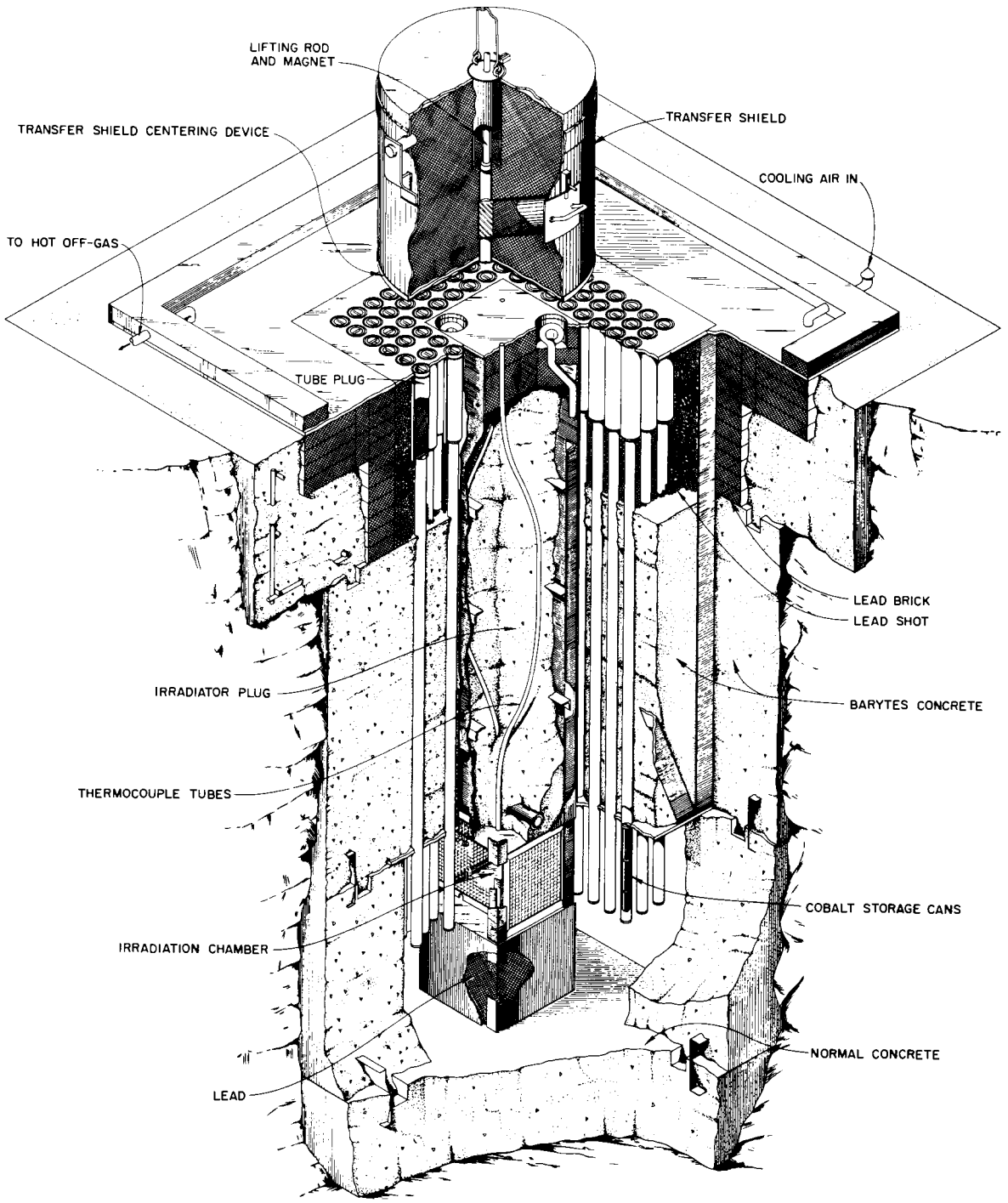

Fig. 1. Cobalt Storage and Irradiation Facility.

PROMETHIUM-147

A process was developed for removing the Am^{241} which contaminates the Pm^{147} that has been separated from the other rare earths by ion exchange methods. The process is based upon the precipitation of promethium directly since there will be macro amounts of promethium in the solutions from the Fission Products Pilot Plant. The precipitation is accomplished by using H_2SiF_6 to precipitate PmF^3, leaving Am^{241} complexed in solution. The H_2SiF_6 decomposes upon heating to form HF and SiF_4. The procedure is cyclic since adequate decontamination is not obtained in one precipitation. Further details are in the Operations Division Report for May 1956.

CESIUM-137

Sales

Twenty-four Cs^{137} sources containing a total of 3816 curies were fabricated. One of these sources was for teletherapy, and contained 2045 curies.

Approximately 12,600 curies of Cs^{137} was collected in the revised Building 3506 equipment as crude product which will be finally purified in the Multi-Kilocurie Manipulator Cell (MKC).

Gamma-Ray Source

The largest $Cs^{137}Cl$ gamma-ray source fabricated to date was completed in February of this year for the University of Michigan. The source was made from three stacked pellets of compressed $Cs^{137}Cl$ powder which were sealed in a double container of type 316 stainless steel. Data for the source are as follows:

Total activity, curies	2025
Weight of CsCl, g	91
Specific activity	
By weight, curies/g	22.4
By volume, curies/cc	67.45
Dimensions	
Diameter, cm	3.183
Height, cm	3.75
Volume, cc	29.65
Density, g/cc	3.07

Preparation of $Cs^{137}Cl$ Powder

The methods considered to be the most promising were investigated for obtaining dry $Cs^{137}Cl$ powder suitable for the fabrication of gamma-ray sources. In the first method the salt was precipitated from an aqueous solution by the addition of hydrochloric acid and ethanol. The salt crystals were separated by filtration, air-dried, and then heated in an agitator-drier for final moisture removal. A satisfactory product was obtained, but this method has the disadvantages of requiring several transfers of material and of introducing an organic substance into the process.

The second method investigated involved the simple procedure of evaporating a solution of CsCl to crystals and continuing heating to dry the crystals in the same container. The equipment required was a heated cylindrical vessel fitted with a motor-driven agitator-scraper. Test runs made with inactive CsCl resulted in a fine crystalline powder of low moisture content (less than 0.004% by weight).

A 2000-curie batch of $Cs^{137}Cl$ was dried by the latter method in a manipulator cell with excellent results. Approximately 99% of the salt was recovered from the drier in a form satisfactory for forming compressed pellets. For more details see Operations Division Report for May 1956.

Purification of Cs^{137} by Chloroplatinate Method

Chloroplatinic acid was investigated as a reagent for the final purification and recovery of fission-product Cs^{137}. A new approach to the problem was indicated because of excessive radiation damage to anion-exchange resins used to remove sulfate ions from cesium alum. The use of H_2PtCl_6 allows a complete separation of cesium from aluminum and sulfate ions in one step and subsequent recovery of cesium as the chloride and platinum as chloroplatinic acid for re-use according to the following reactions:

$$Cs_2Al_2(SO_4)_4 + H_2PtCl_6 = Cs_2PtCl_6 + Al_2(SO_4)_3 + H_2SO_4$$

$$Cs_2PtCl_6 + H_2NNH_2 = 2CsCl + Pt + 4HCl + N_2$$

$$2H_2NNH_2 \xrightarrow{(Pt)} 2NH_3 + N_2 + H_2 \text{ (side reaction)}$$

$$Pt + \text{aqua regia} \longrightarrow H_2PtCl_6 \text{ (re-used)}$$

A Cs^{137} product from the alum purification sequence, containing 1919 curies of Cs^{137}, was processed according to the method outlined above. A calculated excess of 4% H_2PtCl_6 was added to

an aqueous solution of the cesium salt to precipitate Cs_2PtCl_6. The filtrate from this precipitation contained 303 mc of Cs^{137}, which indicated a precipitation of 99.9+% of the activity.

The Cs_2PtCl_6 was washed with 3 liters of dilute hydrochloric acid, which resulted in a solubility loss of approximately 800 mc of Cs^{137}. The total loss of Cs^{137} was approximately 1.1 curies or approximately 0.06% at this point. The slurry of Cs_2PtCl_6 was reduced with hydrazine and the platinum metal recovered as H_2PtCl_6 which was contaminated with 370 mc of Cs^{137}. The $Cs^{137}Cl$ product was reported to contain approximately 0.1 ppm aluminum and no detectable platinum or sulfate. These results would indicate that process losses in this method should not exceed 0.1% and that the method is suitable for large-scale recovery of Cs^{137}. A high platinum inventory is required but nearly complete recovery and re-use may be expected. For more details see Operations Division Report for August 1956.

Accumulation of Cs^{137} by Chloroplatinate Method

The possibility of accumulating Cs^{137} from a fission-product waste solution of the Redox type by the use of chloroplatinic acid was investigated. It was concluded that this method was not applicable to solutions high in ammonium ion concentration (such as Arco type of waste) because of the competition for the chloroplatinate ions between the high concentration of ammonium ions and the low concentration of cesium ions. The highest recovery of Cs^{137} was 80% from such a solution.

Carrying of Cs^{137} with Nickel Ferrocyanide

Experiments indicated that cesium was quantitatively carried from simulated Arco wastes on 0.01 *M* nickel ferrocyanide. Large amounts of ammonium did not interfere with the carrying. Strontium contamination of the ferrocyanide precipitate was 5%. The volume of the 0.01 *M* nickel ferrocyanide was 200 ml when well settled and 70 ml when centrifuged. Only 10% of the rubidium carried on the nickel ferrocyanide precipitate in the presence of excess ammonium ion.

Experiments indicate that the processing of the nickel ferrocyanide precipitate must proceed on a straight-line batch basis. Cesium was not adsorbed quantitatively by a preformed nickel ferrocyanide precipitate. For example, on a cyclic basis, cesium losses would amount to 60% on the fifth batch, while, by comparison, the corresponding cesium loss with ammonium alum carrier would be much less than 1%.

It was found that cesium could be removed from nickel ferrocyanide with excess silver nitrate; $AgNO_3$ that is 0.03 *M* or 0.015 *M* removed 99% or more of the cesium, while 0.001 *M* $AgNO_3$ removed less than 1% of the cesium from nickel ferrocyanide precipitates.

FISSION PRODUCTS PILOT PLANT (F3P)

Plant Construction

Phase I. – Construction of the building and cell block of the Fission Products Pilot Plant met with many delays during the year. Accidental contamination of the area while excavation was in progress, displacement of forms during high-density concrete pours, and leakage into the cell ventilation duct were among the causes of delay. At the end of the year, construction was estimated to be 93.8% complete.

Phase II. – Purchase requisitions were issued for all equipment to be installed in Phase II of the project. Detailed drawings were prepared for prefabricated piping to be installed by the cost-plus-fixed-fee contractor, H. K. Ferguson Company.

Because of the delay in completion of Phase I, installation of equipment and piping was not begun.

Phase III. – Delivery of all Model 8 manipulators and the General Mills manipulator was made during the year. Shielding windows for the manipulator cells are being fabricated.

FISSION-PRODUCT-SOLUTION TRANSFER

One shipment of fission-product waste solution from Arco was made during the year. Shipments were temporarily suspended to allow time for development of shipping procedures for maximum safety in transit.

The pressure in the vapor space of the shielded transfer tank, containing 6060 gross beta curies in solution, was observed over a period of several months. The maximum pressure reached was 4.7 psig.

Determination of the radiation characteristics of fission-product waste from Arco by irradiating film packs in the special shielding plug in the shielded transfer tank revealed that a shield thickness of 2 in. of lead plus $1\frac{3}{4}$ in. of steel in the tank shells will reduce radiation on the exterior of the carrier to 200 mr/hr.

A preliminary investigation was made of the possibility of obtaining waste solution from Savannah River to be used as feed for the Fission Products Pilot Plant. Neutralized waste solution, containing approximately 4.25 curies of Cs^{137} per gallon, can be obtained from underground storage tanks. This waste cannot be evaporated prior to shipment because of the high solids concentration. There is a possibility of obtaining highly concentrated acid waste by making changes in the Savannah River plant process piping.

The number of 500-gal shielded transfer tanks necessary to supply waste containing 200,000 curies of Cs^{137} per year to F3P was calculated. The requirements are as follows: Arco waste, nine containers; Savannah River neutralized supernate, three containers; Savannah River concentrated acid 1-AW, one container.

Allowing a five-year amortization of container cost, combined shipping and container costs per curie of Cs^{137} will be $1.18 in the case of Arco waste, $0.36 in the case of Savannah River neutralized supernate, and $0.13 in the case of Savannah River concentrated acid 1-AW.

Design is in progress on six shielded transfer tanks capable of transporting 500 gal of either Arco waste or Savannah River supernate. It is planned to transfer waste from Arco, using the existing transfer station, until an installation at Savannah River for this purpose can be approved, designed, and installed.

TANK FARM EVAPORATOR REVISION

The equipment in the fission-products laboratory used in the primary production of Cs^{137} became inoperable because of the spread of radioactivity under the floor of the operating gallery and cells, making repairs to equipment economically infeasible. To maintain the production of Cs^{137} in the interim period prior to operation of the Fission Products Pilot Plant, the tank farm evaporator was converted to use as a crystallizer by the addition of an agitator and by piping changes.

A decant filter, similar in design to the filter to be used in the F3P crystallizers, was installed in the tank and was proved to be efficient in operation. The use of the crystallization temperature, indicated by a discontinuity in the recorded temperature curve on the cooling cycle, proved feasible as an indicator of the concentration of crystals of ammonium alum formed.

FISSION PRODUCTS PILOT PLANT PROCESS STUDIES

F3P Chemical Process – Purex Waste

The proposed chemical flowsheet for Purex type of waste to be used in the Fission Products Pilot Plant was tested for the first time on a carrier-free acidic fission-product solution obtained from metal recovery operations.

The nitric acid waste was concentrated by a factor of 10, and the behavior of Ru^{106} was noted at various concentrations of nitric acid. The Ru^{106} loss by volatilization as RuO_4 was not excessive from nitric acid solutions under 10 *N*.

Gross Ru^{106} removal was effected by carrying on an $Fe(OH)_3$ precipitate formed by NH_3 gas addition to the partially neutralized concentrate.

The rare earths and alkaline earths were precipitated as a group by the addition of sodium hydroxide and carbonate. Approximately 95% of the gross activity, other than Cs^{137}, was recovered in the precipitate. An attempt to separate the rare earths from the alkaline earths by a CO_2-free hydroxide precipitation was not successful in a single pass since half the Sr^{90} accompanied the rare-earth hydroxide. A number of such passes would be required for a satisfactory separation. The problem is being investigated, and an alternative process may be desirable.

Cerium-144 was separated in good yield from the other rare earths by precipitation as CeP_2O_7 following oxidation with $KMnO_4$.

Cesium was recovered in nearly 100% yield by carrying with ammonium alum.

Reduction of Mercury in Redox Wastes

Mercury was found to be selectively precipitated as a basic mercurous salt from acidic wastes high in aluminum nitrate content. Cesium and technetium did not carry. Less than 6% of the ruthenium carried. Hydrazine hydrate currently appears to be the most suitable reducing agent to use. Over 99% of the mercury was precipitated consistently.

Redox Waste Flowsheet

The tracer chemistry of the various fission-product groups in aluminum nitrate wastes was investigated. A flowsheet was developed for obtaining carrier-free ruthenium, rare earths, strontium and cesium, as well as the removal of gross amounts of mercury.

Ion Exchangers for Final Decontamination

IRC-50 Resin. – Highly salted solutions with ruthenium, rare-earth, and strontium and cesium tracers were contacted with IRC-50 resin, a carboxylic type of resin. Ruthenium and rare earths at pH of 9 to 10 were adsorbed very well on IRC-50 resin. Strontium was adsorbed satisfactorily only from diluted solutions, and cesium was relatively unadsorbed. Thus further decontamination of alkaline wastes may be obtained by running them through an IRC-50 resin column prior to final disposal.

Adsorption of Cesium on Fuller's Earth and IR-100 Resin. – Cesium was preferentially adsorbed on fuller's earth and IR-100. Ammonium ion interfered with the adsorption of cesium.

SHORT-LIVED FISSION PRODUCTS

Six runs were made during the year in existing facilities to recover short-lived fission products. Adequate amounts of material products were produced to provide an adequate schedule of delivery.

Equipment for separating short-lived fission products and recovering uranium, by use of a solvent-extraction process, was installed in Building 3028. At the end of the year the installation of equipment was about 95% complete and testing of vessels and instruments was in progress. The feed material will be dissolved uranium solution from which the I^{131} has been removed in the Iodine-131 Processing Plant.

The following radioisotopes will be produced by using this new equipment in conjunction with the resin columns which are in existence: Sr^{90}, Ba^{140}, Ru^{103}, Ce^{141}, Pr^{143}, Zr^{95}-Nb^{95}, Y^{91}, La^{140}, and unseparated fission products. The procedure for separating these products is given below.

Zr^{95}-Nb^{95}. – The uranyl nitrate solution, as received from the Iodine-131 Processing Plant, is added to a 1-liter silica gel column. Zirconium and niobium and a small portion of the plutonium adsorb on the silica gel. Washing the silica gel with distilled water removes the uranyl nitrate solution, and further washing with 0.5% nitric acid removes the last portion of plutonium. Zirconium and niobium are then removed from the silica gel with 5% oxalic acid.

Mixed Fission-Product Solution. – The acid concentration of the uranyl nitrate solution is adjusted to 6 *N*, and the uranyl nitrate is removed by extracting with tributyl phosphate solution (30 vol % tributyl phosphate in carbon tetrachloride solution) with the mixer-settler equipment. The tributyl phosphate–carbon tetrachloride solution must be previously equilibrated with an equal volume of 6 *N* HNO_3.

Five extractions with tributyl phosphate solution are necessary to remove the uranium and plutonium to below 200 alpha counts per millicurie of mixed fission products. A portion of this solution is removed as gross fission-product solution.

Rare Earths. – The acid solution, containing the short-lived rare earths and alkaline earths, is reduced in volume by boiling to approximately 5 liters. The nitric acid concentration is adjusted to above 12 *N* HNO_3. The rare earths are extracted from the nitric acid solution into tributyl phosphate solution (30 vol % in carbon tetrachloride) and stripped from the tributyl phosphate with distilled water.

The solution of rare earths is boiled to a low volume to reduce the acid concentration and is diluted to 0.1 *N* HNO_3 to prepare for an ion-exchange separation. The rare earths are adsorbed from 0.1 *N* HNO_3 solution by a Nalcite HCR column (1.0 by 40 cm).

From the ion-exchange resin bed, the rare earths are removed selectively by elution with 0.2 *M* citrate solution adjusted to a pH of 3.2. The temperature during elution is 95°C. Yttrium-91, Nd^{147}, Pr^{143}, $Ce^{141,144}$, and La^{140} are removed in that order from the heated column.

Each product as removed is analyzed, concentrated, and prepared as a purified product solution.

Alkaline-Earth Products. – The 12 *N* HNO_3 solution from which the rare earths have been extracted contains $Sr^{89,90}$ and Ba^{140}. This solution is boiled to a low volume and diluted to 0.1 *N* HNO_3.

The barium and strontium are adsorbed on a Nalcite HCR (high-capacity cation resin) column, and the two activities are separated by elution with 0.2 *N* citrate solution at pH 7.5.

$Ru^{103,106}$. – From the uranyl nitrate solution as received, the $Ru^{103,106}$ can be recovered by oxidation of the Ru^{3+} or Ru^{4+} to the RuO_4 and distillation. The oxidation is accomplished by addition of saturated potassium permanganate solution. The final product is ruthenium chloride ($RuCl_3$ in 6 *N* HCl).

The installation of this equipment will provide for a severalfold increase in capacity to produce

the afore-mentioned radioisotopes, as well as provide an integrated operation which was not possible in the past.

RADIOACTIVE GASES

Krypton-85

A total of 6300 curies of Kr^{85} was purified this year. A high-capacity charcoal-packed column, to be installed as additional equipment in the Kr^{85} production plant, was designed. The addition of this column will provide a severalfold increase in capacity of the present equipment.

Xenon-133

Equipment for the production of Xe^{133} was designed and constructed but has not yet been installed. The installation of this equipment will make available another short-lived fission product which is practical to produce as a part of the Radioisotope Distribution program.

Tritium

A total of 2906 curies of tritium was packaged in 171 ampoules, representing a curie increase of 370% over that of CY-1955; however, the number of ampoules increased by only 63%, which indicates an increasingly large amount of tritium in each ampoule prepared for shipment.

A new tritium system was constructed and installed. This system is discussed in another report.[1] A miniature ionization chamber was added to the tritium system after the report was written. This chamber is used for determining the purity of tritium going into each shipping container. The chamber has been calibrated and is now in regular use.

Considerable effort was expended in advising users, both at ORNL and at other sites, on methods of handling tritium.

TARGETS FOR PARTICLE ACCELERATORS

A total of 114 tritium-zirconium targets was fabricated, 108 of which were made by evaporation and 6 by fusion. This production represents an increase of 39% over that of CY-1955. The amount of tritium held by these targets was 115.49 curies.

Thirty-one targets of stable isotopes were fabricated, representing an increase of 420% over those fabricated during the previous year.

The method of preparing tritium-zirconium targets is given in another report.[2] As mentioned in the report, the only backing metal that gives consistently satisfactory results is platinum. It may be possible to use other metals by impregnation at room temperature. A stainless steel impregnation chamber was designed for study of this problem. It is to be equipped with a miniature ionization chamber for measuring changes in tritium pressure.

The experience gained in the preparation of tritium-zirconium targets has been of considerable value in the preparation of evaporated targets of other materials. Targets, for project use, were made of the following elements: Li^6, B^{10}, Be, and V.

MANIPULATOR CELL OPERATIONS

Special Tools for Manipulator Cell Application

Several special tools were fabricated for use in manipulator cells in order to assist or relieve the load upon the manipulators. Among these tools were a motorized remote-control unit can opener, a modified electric impact wrench for opening and closing Co^{60} storage cans and sealing Co^{60} teletherapy sources, and a Carver hydraulic press modified so that the hydraulic pump could be operated from the face of the cell.

Multi-Kilocurie Manipulator Cell (MKC)

The following data are recorded because of the current interest in manipulator cell operation:

1. For 91 calendar days of the past year the cell was assigned to the final purification of Cs^{137} and to the fabrication of high-level Cs^{137} sources. The low-level radiographic sources of Cs^{137} were fabricated in the Argonne type of cell.
2. The total down time for the cell amounted to approximately two weeks. This was for the purpose of routine maintenance, repairs to manipulators, and removal and cleaning of the viewing window.
3. For the remainder of the year, approximately 37 weeks, the cell was used for Co^{60} work, including the assaying and storing of bulk material for stock, the fabricating of sources, and the loading and preparation of sources for shipment.

[1] B. J. Massey, *Tritium Handling System*, ORNL-2238 (Feb. 8, 1957).

[2] B. J. Massey, *Preparation of Thin Tritium-Zirconium Targets*, ORNL-2237 (Feb. 8, 1957).

REMOTE WELDING OF STAINLESS STEEL CONTAINERS

The sealing of stainless steel containers of highly radioactive materials such as Co^{60}, Cs^{137}, and Ir^{192} has been demonstrated successfully by remotely operated shielded-arc welding and has become a routine procedure (see Figs. 2 and 3).

OTHER ITEMS OF INTEREST

Irradiated Units

A total of 2721 units was prepared for irradiation in the ORNL Graphite Reactor, the LITR, the MTR, and at Hanford. A total of 453 irradiated units was chemically processed.

Radioisotope Product Data

The manual *Radioisotope Specifications for Preparations* was revised and brought up to date. The radioisotope sales catalog was revised and prepared for reissue.

Radioisotope Distribution Data

Effective February 10, 1956, AEC licensing procedures were changed to require all requests to be placed on approved AEC purchase order forms and to eliminate license requirements for shipments out of the United States.

On April 11, 1956, the priority list for Co^{60} orders was discontinued.

Scandium-46 is now priced at $150 per curie for amounts in excess of 10 curies.

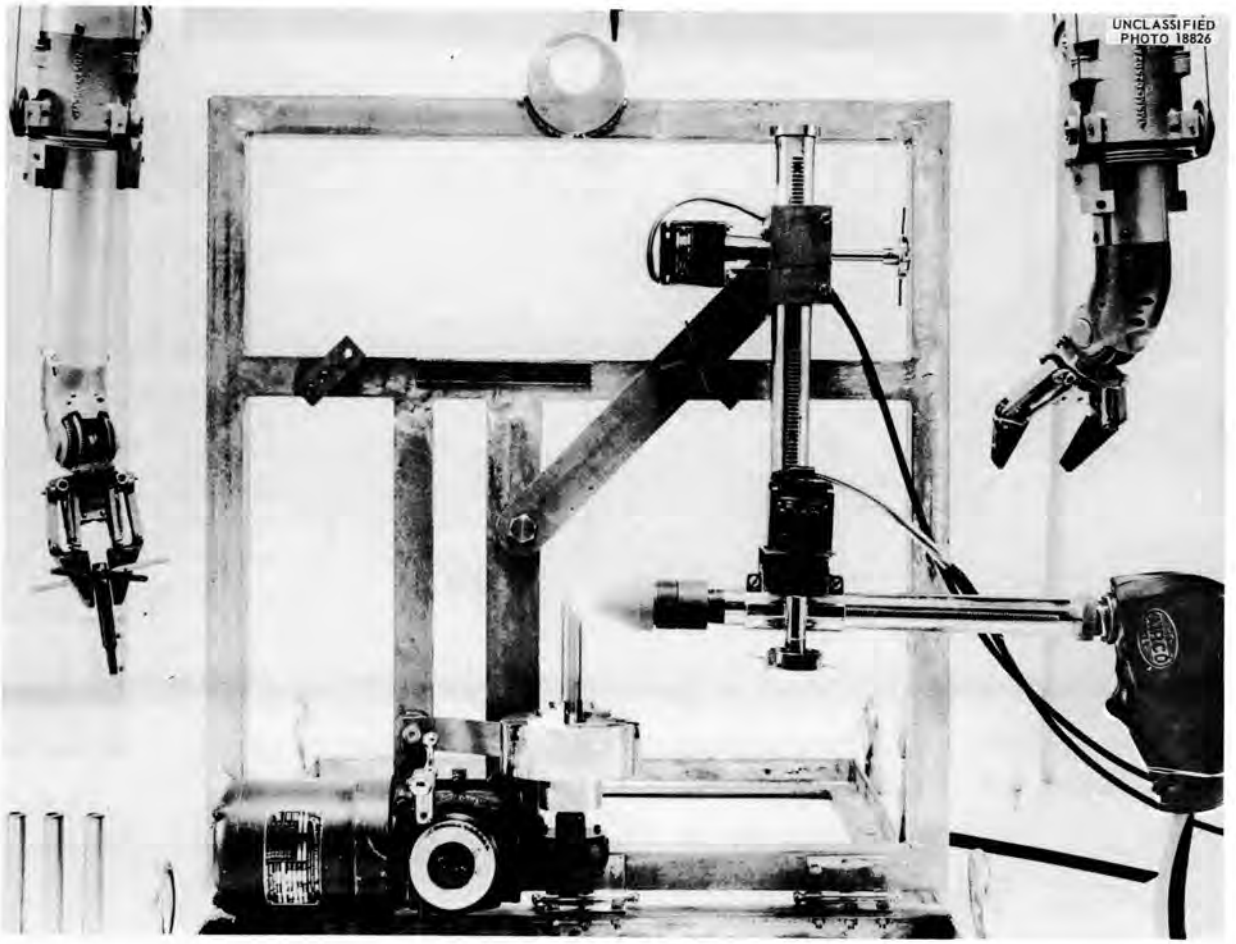

Fig. 2. Remote-Control Sealing of Radioactive Source Containers by Shielded-Arc Welding.

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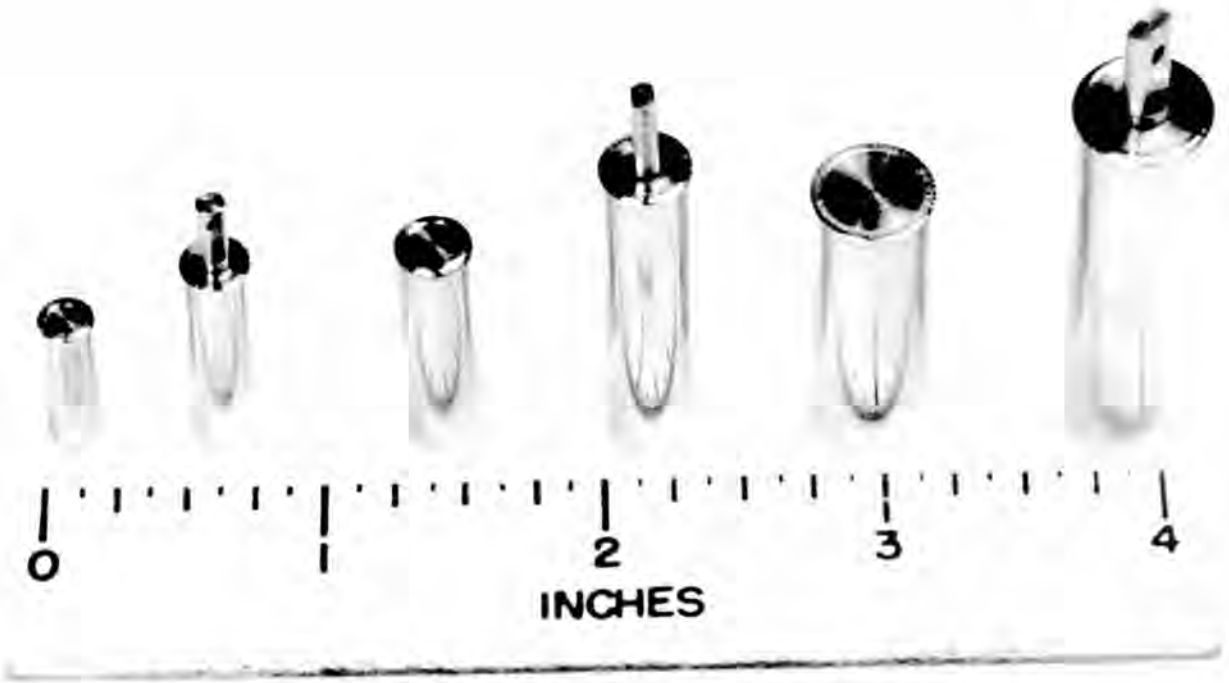

Fig. 3. Typical Radioactive Source Containers Sealed by Remotely Controlled Welding.

Effective July 1, 1956, the prices of I^{131} and C^{14} were reduced as follows:

I-131-P	–	$0.50 per millicurie for shipments through 499 mc
		$0.40 per millicurie for shipments in excess of 499 mc
C-14-P	–	$28.00 per millicurie for shipments through 199 mc
		$22.00 per millicurie for shipments in excess of 199 mc

Boron compounds are now distributed as part of the Stable Isotope Distribution program.

RADIOISOTOPE SHIPPING

Container Changes

A substantial reduction in transportation charges to the customers and a small reduction in the Laboratory's container costs were accomplished on 25 to 30% of the shipments by adding a smaller (4 × 4 × 8 in.) fiberboard box to the other two sizes previously in use. One factor that influences the minimum air transportation rates is the outside dimensions of a package. Reducing the minimum size of the boxes from 8 × 8 × 8 in. to 4 × 4 × 8 in. reduced customer shipping charges based on a minimum weight of 3 to 1 lb, even though the actual weight reduction was insignificant.

In last year's report mention was made of the effort to eliminate wooden boxes on returnable containers, used for about 10% of all shipments, in order to reduce maintenance costs and shipping weights. This project was essentially completed on all containers used for shipping solid materials; the metal shields are now mounted on reinforced wooden pallets and wooden blocks.

The use of wooden outer boxes for returnable liquid containers continues unchanged in about 2% of the shipments. The aluminum outer box, mentioned in last year's report as being approved by the Bureau of Explosives, was tested and found too expensive to fabricate and maintain.

No depleted uranium containers were fabricated besides the four made in 1955. The AEC's restriction of their use to infrequent project shipments has made the additional cost of fabrication over

that of lead containers unjustifiable even though shipping weights were reduced by more than 50% in every case.

An effort was made to improve the appearance of the radioisotope packages. Labels formerly glued to the outside of the fiberboard boxes are now printed on the box as part of the box manufacturer's process. The boxes, previously glued, are now stapled not only to improve the appearance but also to save operators' time. Arrangements have also been made, and an order placed, for painting labels on the tin cans; delivery is expected early in 1957.

Operating Accuracy

Maintaining the necessary accuracy in radioisotope shipping requires careful choice of personnel suited for this type of work and constant follow-up. The shipping operation involves about 100 different products and more than 13,500 packages per year, each "tailor-made" to suit the customer's individual requirements. To improve the follow-up portion of the work and to obtain a record for determining individual responsibility in the event of an error, a new system was established early in 1956 which provides for a written record being made of every step of the packaging operation.

It is believed that the new system is at least partially responsible for the exceptional accuracy accomplished this year. There is no record of a customer's complaint of serious error and no more than three complaints of minor difficulties. Such complaints as leaking bottle caps are normally due to equipment problems.

Personnel Exposure

A complete radiation study of the packing and shipping area was made in an effort to determine the sources of radiation causing a background, in order to reduce the exposure to personnel. This study was prompted by anticipated changes in measurement of exposure; it appeared that it would be impossible to maintain the same people in the shipping area over a long period of time without exceeding the tolerance levels.

The study revealed that a large portion of the exposure was caused by "skyshine" from beta-emitting products stored in an uncovered barricade. Other factors contributing to the exposures were an inadequately shielded product carrier and improperly designed product bottles which force operators to reach behind the barricade in order to empty them completely with a remotely controlled pipette. It was also learned that some exposure could be reduced by changes in operating techniques.

Except for the product storage bottles which are now being redesigned by the Owens-Illinois Glass Company, all the corrective measures have been accomplished. The extent of radiation exposure decrease will not be known until Health Physics exposure reports for the last quarter of 1956 are published.

Operating Costs

For the first full calendar year, complete cost records were kept on the radioisotope packaging and shipping operations. The basic costs (labor and material) were $72,300, or $5.32 per package, while the total costs, including allocated charges (mainly Health Physics and overhead), were $129,820, or $9.56 per package. The overhead accounted for $35,000 or 61% of the allocated charges, and Health Physics accounted for $18,000 or 32% of the same charges. The costs shown include the maintenance and fabrication of all containers except those used exclusively for shipment of special materials such as cyclotron targets and Co^{60} sources.