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REVIEW OF RADIOISOTOPES PROGRAM, 1964

J. H. GILLETTE

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ISOTOPES DIVISION

REVIEW OF RADIOISOTOPES PROGRAM, 1964

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REVIEW OF RADIOISOTOPES PROGRAM, 1964

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RADIOISOTOPE APPLIED RESEARCH

Reactor-Produced Isotopes

Copper-67

Copper-67 (61 h), useful in medicine and metallurgy, was produced by irradiating ~26 mg of 92.3% ^{67}Zn for seven weeks in a partial fuel element in the ORR. The effective threshold energy for the $^{67}Zn(n,p)^{67}Cu$ reaction is 4.7 Mev.[1] A solution of the zinc target in 5 M HCl was placed on a Bio-Rad 1, X-10, anion-exchange column equilibrated with 5 M HCl; 5.4 mc of carrier-free ^{67}Cu was obtained by elution with 2.5 M HCl. Low-energy gamma radiation ranging from 0.091 to 0.184 Mev associated with the beta decay of ^{67}Cu can be readily shielded and collimated for handling and counting.

Phosphorus-33

Specific uses for ^{33}P include autoradiography, studies of recoil energy, synthesis of complex organophosphorus compounds, and double labeling of phosphorus in complex molecules. In comparison to ^{32}P, ^{33}P has a longer half-life (25 d as compared to 14.3 d) and a lower maximum beta energy (0.25 Mev as compared to 1.7 Mev). Highly enriched ^{33}S or ^{36}Cl must be used to produce millicurie quantities of ^{33}P by the $^{33}S(n,p)^{33}P$ or $^{36}Cl(n,\alpha)^{33}P$ reaction, since ^{32}P from the associated $^{32}S(n,p)^{32}P$ or $^{35}Cl(n,\alpha)^{32}P$ reaction is also present. The effective cross sections of the four reactions in optimum positions of the ORR and the neutron flux are given in Table 1.

Table 1. Effective cross sections for optimum reactor positions in the ORR

Position, ORR	Neutron flux		Reaction	Effective cross section, mb
	Thermal	Fast		
Isotope stringer No. 1	2.2×10^{14}	4.4×10^{13}	$^{32}S(n,p)^{32}P$	9.2
	2.2×10^{14}	4.4×10^{13}	$^{33}S(n,p)^{33}P$	11.3
NF stringer	2.5×10^{14}	1.2×10^{14}	$^{35}Cl(n,\alpha)^{32}P$	13.2
	2.5×10^{14}	1.2×10^{14}	$^{36}Cl(n,\alpha)^{33}P$	10.2

Millicurie quantities of ^{33}P have been produced by the $^{36}Cl(n,\alpha)^{33}P$ reaction using reactor-produced ^{36}Cl.

Rubidium-84

Rubidium-84 (33 d), one of the more widely used isotopes as an indicator of a dynamic system, is important in heart studies. This isotope is usually produced by the $^{84}Kr(p,n)^{84}Rb$ reaction, but contaminating ^{83}Rb (83 d) produced by the $^{84}Kr(p,2n)^{83}Rb$ and $^{83}Kr(p,n)^{83}Rb$ reactions is also present.

The reactor production yields of ^{84}Rb by the $^{84}Sr(n,p)^{84}Rb$ reaction in an ORR partial fuel element using both natural and enriched ^{84}Sr targets are given in Table 2.

Table 2. Reactor production of rubidium-84

Target	Irradiation time, day	Yield, mc
48 mg $^{84}Sr(NO_3)_2$ 83.3% enriched	49	2.05
371 mg natural $SrCO_3$	49	0.103

The effective threshold energy for this reaction is 6.1 Mev.[2] Carrier-free ^{84}Rb was recovered by placing a neutral solution of the target on a Duolite CS-101 cation-exchange column in the ammonium form and eluting with 0.15 M NH_4Ac.[3] No ^{85}Sr could be detected in the ^{84}Rb product (limit of detection, ~0.5 μc of ^{85}Sr).

From the extrapolation of the results in Table 2, yields for the enriched and natural targets are 12 and 8.4 mc, respectively, of ^{84}Rb per 100 mg of enriched ^{84}Sr under identical irradiation conditions.

Manganese-54

The reactor yields of ^{54}Mn, produced by the $^{54}Fe(n,p)^{54}Mn$ threshold reaction, for various positions of the ORR and LITR are given in Table 3.

Table 3. Manganese-54 reactor yields*

Position	Relative fast neutron flux	Thermal flux, $n/cm^2 \cdot sec$	Yield ^{54}Mn per 100 mg Fe, μc
ORR			
Isotope stringer No. 2	low	4.8×10^{14}	420
NF stringer	intermediate	2.1×10^{14}	780
Hydraulic tube No. 1	intermediate	2.6×10^{14}	960
Partial fuel element	high	3.2×10^{14}	1500
LITR			
Partial fuel element	very low	2.4×10^{13}	80

*Yield data normalized to 539-hr irradiation time.

The effective threshold energy (E_{eff}) for this reaction is reported as 4.3 Mev[4] and 5.1 Mev.[5] According to published data,[6-12] the cross section for this reaction ranges from 40 to 76 mb.

Carrier-free ^{54}Mn was separated from the iron targets by passing a 12 M HCl solution of the target through a Dowex-1, X-10, anion-exchange column and eluting the manganese with 8 M HCl.[13]

Reactor Neutron Cross Sections for (n,γ) Reactions

The reactor neutron cross section for the production of ^{109}Cd by the $^{108}Cd(n,\gamma)^{109}Cd$ reaction is 0.22 ± 0.03 barn (confidence limit, 95%). The cross section was determined from two irradiations of enriched ^{108}Cd and from three irradiations of natural silver. The ^{107}Ag was converted to ^{109}Cd by the $^{107}Ag(n,\gamma)^{108}Ag \xrightarrow{\beta^-} {}^{108}Cd(n,\gamma)^{109}Cd$ reaction. In addition to the reactor neutron cross section for the $^{108}Cd(n,\gamma)^{109}Cd$ reaction, the reactor cross sections for the following reactions, calculated from the data obtained in the silver irradiations, are given in Table 4.

Table 4. Comparison of reactor and literature cross sections

Reaction	Product half-life	Cross section, barns	
		Measured	Literature
$^{107}Ag(n,\gamma)^{108,108m}Ag$	≥5 y, 2.3 m	33.8	30
$^{107}Ag(n,\gamma)^{108}Ag$	≥5 y	0.4	*
$^{107}Ag(n,\gamma)^{108m}Ag$	2.3 m	33.4	30
$^{109}Ag(n,\gamma)^{110,110m}Ag$	24 s, 270 d	99.1	113
$^{109}Ag(n,\gamma)^{110}Ag$	24 s	96.9	110
$^{109}Ag(n,\gamma)^{110m}Ag$	270 d	2.2	2.8
$^{108}Cd(n,\gamma)^{109}Cd$	1.3 y	0.22	2.7 ± 0.9
$^{110}Cd(n,\gamma)^{111}Cd$	49 m	≤1.7	3.1

*No data available.

Cyclotron-Produced Isotopes

Nickel-57

The two readily available nickel radioisotopes (8 x 10^4-y ^{59}Ni and 92-y ^{63}Ni), which decay by electron capture and weak beta emission, present formidable problems in analysis as well as inconvenient half-lives. Nickel-66 (55 h) can be produced in the reactor by the $^{64}Ni(n,\gamma)^{65}Ni(n,\gamma)^{66}Ni$ reaction; however, calculations show that for a 58-mg isotopically enriched ^{64}Ni target only 300 μc may be made from a week's irradiation in the ORR. Of the two remaining useable nickel isotopes, 6.4-d ^{56}Ni is produced by

the $^{54}Fe(\alpha,2n)^{56}Ni$ reaction and 36-h ^{57}Ni by the $^{58}Ni(p,pn)^{57}Ni$ and the $^{58}Ni(p,2n)^{57}Cu \xrightarrow{decay} {}^{57}Ni$ reactions.

Nickel-57 was produced by irradiating 0.007-in. high purity normal nickel foil in the window target facility of the ORNL 86-Inch Cyclotron with an 80-90 μa, 23-Mev proton beam. After irradiation, the nickel foil was dissolved in concentrated HCl and passed through a Bio-Rad AG-1 ion-exchange column which effectively removed manganese, cobalt, and copper. The yield of ^{57}Ni for a 0.306-g nickel target was 9.7 mc/hr with a specific activity of 31.7 mc/g.

Cobalt-57

Cobalt-57 (267 d) was formed in the above target by the $^{58}Ni(p,pn)^{57}Ni \xrightarrow{\beta^+} {}^{57}Co$, $^{58}Ni(p,2n)^{57}Cu \xrightarrow{decay} {}^{57}Ni \xrightarrow{\beta^+} {}^{57}Co$, $^{58}Ni(p,2p)^{57}Co$, and $^{60}Ni(p,\alpha)^{57}Co$ reactions. The amount of ^{57}Co obtained was 148 μc/hr which is in fair agreement with a previous rate of ~500 μc/hr since the target foils were wrapped in 0.002-in. aluminum foils which degraded the beam energy.

Beryllium-7

Lithium metal was irradiated for 3.25 hr with a 210-μa beam of 17.7-Mev protons to produce a ^{7}Be yield of ~36 mc/hr by the following procedure. After the short-lived isotopes had decayed, the target was dissolved in 0.5 M HCl and ~5 mg of iron carrier and NH_4OH were added to coprecipitate ^{7}Be along with $Fe(OH)_3$. After three such precipitations, the $Fe(OH)_3$ precipitate was extracted three times with bis-(β-chloroethyl)ether to remove the iron carrier. After evaporation of the aqueous phase to incipient dryness and treatment with 15 M HNO_3 to destroy organic matter, the product was converted to the chloride with 12 M HCl.

Yttrium-86

A 100-mg sample of 97.6% isotopically enriched ^{86}Sr as $SrCO_3$ was irradiated for 6 hr by a 79-μa beam of 12-Mev protons to produce ^{86}Y by the $^{86}Sr(p,n)^{86}Y$ reaction. After dissolution of the target and nickel container in concentrated HCl, radiocolloidal separation (with the iron content of the nickel tube acting as scavenger) was followed by elution of yttrium while the iron was retained on the ion-exchange column. The following yttrium radioisotopes were observed in the product: 63.0 mc of ^{86}Y, 0.96% ^{88}Y, and a fraction of a percent of ^{87}Y.

A cooperative study of the decay of ^{86}Y was initiated with Prof. J. H. Hamilton and his group at Vanderbilt University and N. R. Johnson, Chemistry Division, ORNL, to clarify the proposed ^{86}Y decay scheme[14] with regard to intensities and multipolarities of the transition. Internal and external conversion measurements were made in an iron-free double focusing spectrometer at Vanderbilt and a lithium-drifted germanium solid state spectrometer at ORNL. Gamma spectroscopy measurements with NaI(Tl) crystals were made at both laboratories.

Yttrium-87

Carrier-free ^{87}Y (80 h) was produced by the $^{87}Sr(p,n)^{87}Y$ reaction from 24 g of >95% isotopically enriched ^{87}Sr using the 80-90 μa internal beam and irradiating for 4-6 hr. To optimize the (p,n) reaction and to reduce the (p,2n) reaction (which would contribute ^{86}Y), the proton energy was degraded to 12 Mev by the use of aluminum absorbers. After irradiation, the nickel tube and target were dissolved in hot concentrated HCl. A few milligrams of iron and NH_4OH were added to coprecipitate the yttrium with $Fe(OH)_3$. After filtering and washing, radiocolloidal precipitation gave >99% yield.

Yttrium and iron were separated on a Dowex-1, X-8, ion-exchange column; the iron was retained and yttrium passed through. After evaporation and purification with HNO_3, HCl was added to form the chloride. The yield was 8.4 mc/hr.

Yttrium-88

Yttrium-88 (108 d) was produced by a 2-hr irradiation of 2.4 g of $SrCO_3$ in a nickel capsule by the $^{88}Sr(p,n)^{88}Y$ reaction, using 23-Mev protons degraded with aluminum absorbers. Small quantities of short-lived ^{86}Y and ^{87}Y were also produced from the ^{86}Sr and ^{87}Sr present in the natural strontium target (^{88}Sr = 82.56%). The chemical separation, isolation, and purification procedures for ^{86}Y were essentially identical to those used for ^{86}Y and ^{87}Y. Analysis of the ^{88}Y product was based on the observation of the 1.845-Mev photo peak, using a 3 by 3 in. NaI(Tl) crystal gamma spectrometer. The yield was 2.3 mc/hr with no radiocontaminants in excess of $\sim 10^{-3}$%.

Promethium-144

Carrier-free ^{144}Pm (380 d) was produced by the $^{144}Nd(p,n)^{144}Pm$ reaction using 100 mg of 97% enriched ^{144}Nd as the oxide. A beam current of 84 μa was used in the 10-hr irradiation. Promethium-144 was separated and purified by ion exchange using a Bio-Rad 50W column and eluting with 0.4 M α-hydroxyisobutyric acid. A yield of ~10 mc/hr was obtained.

Optimum Target Thicknesses

In the ORNL 86-Inch Cyclotron, where protons of 17-23 Mev energy are available at beam currents up to ~3 ma, the thick target yield of a particular isotope will increase with increasing proton energy. This increase is due to the fact that the reaction cross section does not drop to zero after peaking (but decreases to ~10% of the peak value with an increase of 3-4 Mev in proton energy). For (p,2n) reactions, the yield per kilowatt-hour or per milliampere hour rises with increased energy — at least to 23 Mev. For (p,n) reactions, however, there is an optimum energy at which the yield per kilowatt-hour starts to decrease, even though the yield per milliampere hour is still rising. Since the ability of the target to withstand irradiations at high beam currents, the target material costs, the specific activity, and the general operation of the cyclotron are

dependent on optimizing the target thickness for both (p,n) and (p,2n) reactions, a study was undertaken to determine the penetrability of the beam into flat-plate targets.

Stacked-foil targets were placed at an incident angle of 6° to the deflected beam at a probe position in the deflected-beam pipe. A 0.375-in. water-cooled, graphite collimator (Fig. 1) was used to center the beam on the stack of ten 0.001 x 0.5 x 3.75 in. foils. The center 3.0 in. of the foils was exposed to the beam (~0.2 μa-hr) and then set aside until the short-lived radioisotopes decayed to negligible levels. To eliminate scattering effects, an additional 0.250 in. was cut from each end of the 3.0 in. sections.

Analyses were performed with gamma spectrometers using (1) a 3.0 by 3.0 in. NaI(Tl) crystal with a 0.732 g/cm^2 Lucite absorber and (2) a 1.5 by 0.080 in. NaI(Tl) or 3.0 by 0.080 in. NaI(Tl) crystal with beryllium windows. Data were collected with a 512 channel analyzer.

Results of the penetration studies are shown in Table 5. For (p,n) reactions studies, the optimum thickness (i.e. minimum thickness required to produce >95% of the desired activity which would be produced in an infinitely thick target) appears to be ~0.006 in. For (p,pn), (p,2n), or (p,3n) reactions, the optimum thickness for the reactions studies appears to be ≤0.003 in.

Table 5. Results of 23-Mev proton penetration studies

Reaction	Product half-life	Detected photon, Mev	Min thickness mils	Min thickness mg/cm^2
$^{56}Fe(p,n)^{56}Co$ $^{57}Fe(p,2n)^{56}Co$ $^{58}Fe(p,3n)^{56}Co$	77.3 d	0.845	3.0	60
$^{58}Ni(p,pn)^{57}Ni$ $^{58}Ni(p,2n)^{57}Cu \rightarrow {}^{57}Ni$	36.0 h	1.38	<2.0	45
$^{58}Ni(p,\alpha)^{55}Co$	18.0 h	0.930	3.2	72
$^{58}Ni(p,2p)^{57}Co$ $^{58}Ni(p,pn)^{57}Ni \xrightarrow{\beta^+} {}^{57}Co$ $^{58}Ni(p,2n)^{57}Cu \rightarrow {}^{57}Ni \xrightarrow{\beta^+} {}^{57}Co$	267.0 d	0.122	2.5	57
$^{55}Mn(p,pn)^{54}Mn$	314 d	0.840	2.0	37
$^{65}Cu(p,n)^{65}Zn$	245 d	1.119	6.0	135
$^{103}Rh(p,pn)^{102}Rh$	206 d	0.475 1.08	2.5	77
$^{103}Rh(p,n)^{103}Pd$	17 d	20.2 kev*	6.0	190
$^{109}Ag(p,n)^{109}Cd$	1.3 y	88.0 kev	~5.0	133
$^{196}Pt(p,n)^{196}Au$	6.2 d	0.356 0.331	2.5	136
$^{207}Pb(p,n)^{207}Bi$ $^{208}Pb(p,2n)^{207}Bi$	30 y	1.77	3.5	100

*22.5 kev reported by independent analysis group.

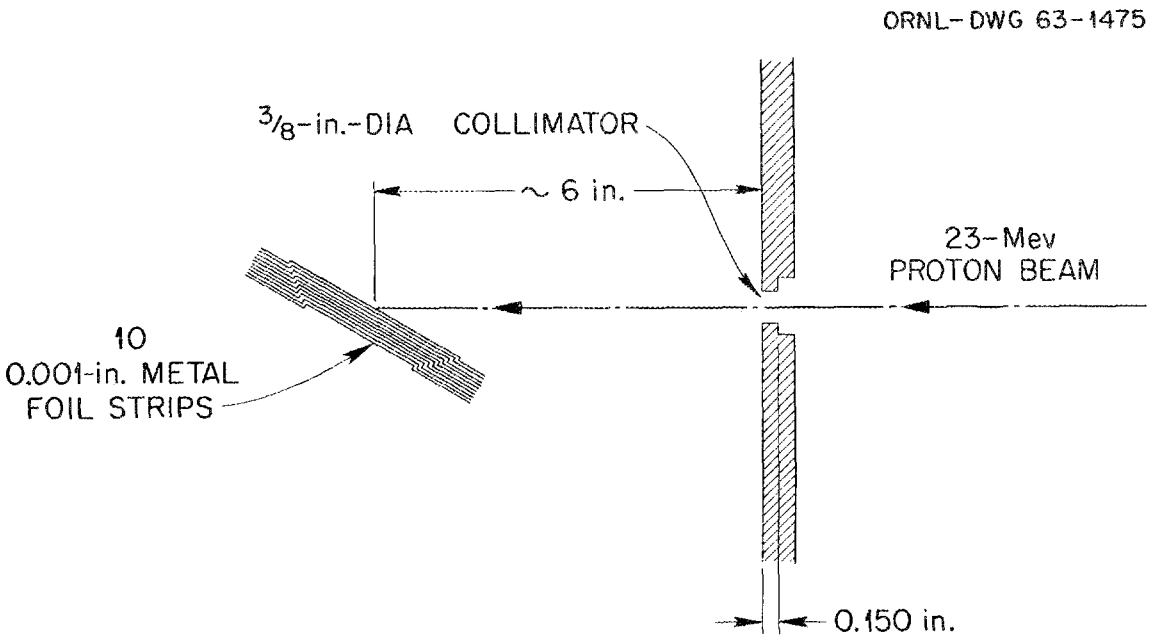


Fig. 1. Schematic Diagram of Ten 0.001-in. Foil Strips in the 86-Inch ORNL Cyclotron

Radioisotope Generators

Short-lived isotopes can be conveniently obtained by separation from a long-lived parent which is fixed in a matrix. Usually, the parent radioisotope is isolated and adsorbed on an ion-exchange resin; the carrier-free decay product (daughter) is then eluted from the ion exchanger or "generator" for convenient use on location.

Preparation of 2.8-h Strontium-87m Generator

High specific activity ^{87}Y was produced by the $^{87}Sr(p,n)^{87}Y$ reaction from 130 mg of 95.4% enriched ^{87}Sr as $SrCO_3$, which minimized ^{88}Y production (only 0.96 mc ^{88}Y as compared to ~19 mc using normal strontium). After chemical separation of the parent ^{87}Y, the solution (of ^{87}Y) was evaporated to incipient dryness and taken up in 10-20 ml of 0.01 M $(NH_4)_2CO_3$ which was then transferred to a Bio-Rad AG-1, X-10, 100 to 200 mesh, ion-exchange column (Fig. 2) previously equilibrated with 0.01 M $(NH_4)_2CO_3$. The column was rinsed with 25 ml of 0.01 M $(NH_4)_2CO_3$; then the 2.8-h ^{87m}Sr daughter was eluted, as needed, by passing more 0.01 M $(NH_4)_2CO_3$ solution through the column. The elution curve is shown in Fig. 3. No radiocontaminants were detected in the ^{87m}Sr prepared by evaporating the $(NH_4)_2CO_3$ (limit of detection, 5×10^{-5} μc, corresponding to 5×10^{-7}% with a 10-mc generator).

Preparation of 6.0-h Technetium-99m Generator (Model I)

An ammonical solution of reactor-produced ^{99}Mo was evaporated to dryness and 0.01 M HCl was added. The solution was passed through an HCl-acidified ion-exchange column of Alcoa alumina, F-20, which adsorbed >99% ^{99}Mo. After the column had been rinsed with 30 ml of methyl ethyl ketone, ^{99m}Tc was eluted with 20 ml of methyl ethyl ketone (5 vol % 0.01 M HCl) and both solutions were discarded. After sufficient ^{99m}Tc had accumulated, the column was eluted with 20 ml of methyl ethyl ketone (5 vol % 0.01 M HCl) which removed >75% of the ^{99m}Tc in ~5 min. Generators charged with ≤300 mc ^{99}Mo (~200 mg molybdenum) had $<3 \times 10^{-4}$% molybdenum as the only radiocontaminant in the ^{99m}Tc product and no alumina or other solids.

Preparation of 6.0-h Technetium-99m Generator (Model II)

A 0.01 M HCl solution of reactor-produced ^{99}Mo was adsorbed onto a Bio-Rad HZO-1, 50 to 100 mesh, inorganic exchanger and >98% ^{99m}Tc was eluted with 20 ml of methyl ethyl ketone containing 5 vol % 0.01 M HCl. A dual experiment using ~60 mc of ^{99}Mo for each column demonstrated that the product from the zirconia column was ~10 times purer than that using the alumina column; some ^{99}Mo leaked through the alumina column.

Preparation of 6.0-h Technetium-99m Generator (Model III)

Fission product ^{99}Mo was used in the Bio-Rad HZO-1/methyl ethyl ketone system. To prevent large losses of ^{99m}Tc during evaporation, the acid solution was initially made ammonical. After the column was charged and eluted, ^{103}Ru and ^{131}I were detected in the eluate. An inert diluent

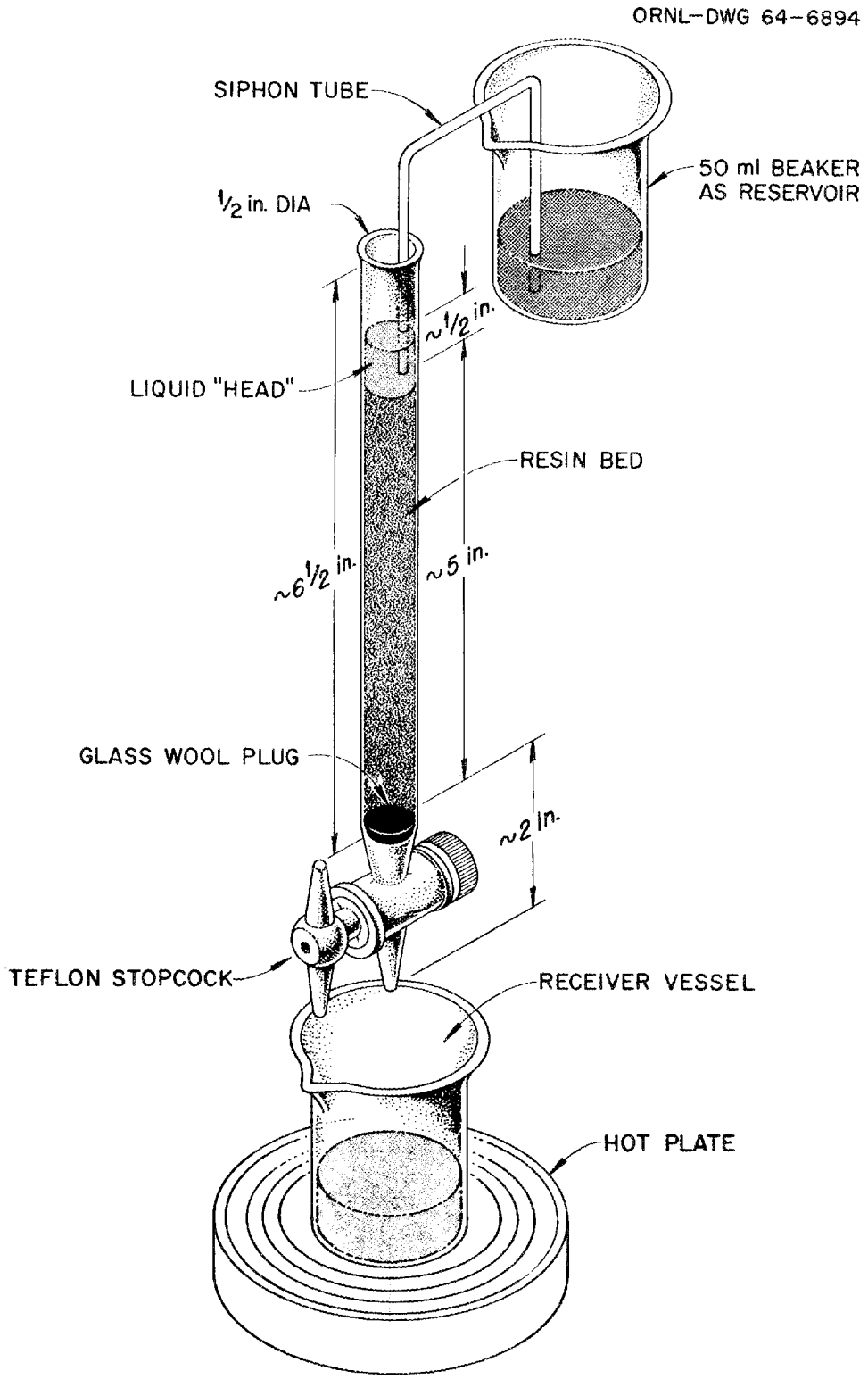


Fig. 2. Diagram of Strontium-87m Generator Assembly.

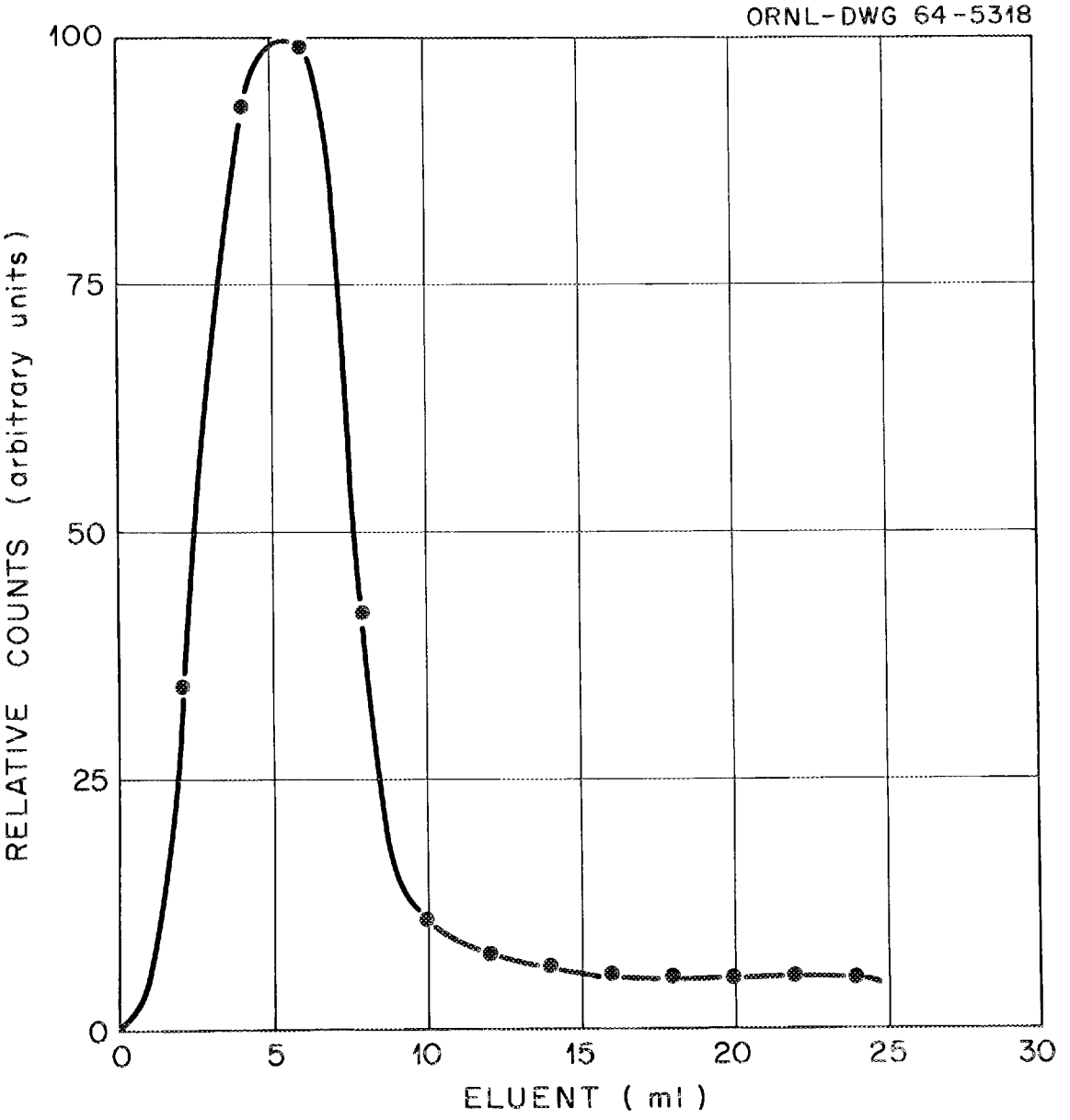


Fig. 3. Elution Curve of Strontium-87m from a Bio-Rad AG-1, X-10, Column with 0.01 M $(NH_4)_2CO_3$.

(hexane) was added to the organic solvent and the ^{99m}Tc was extracted into the aqueous phase, achieving a ^{99m}Tc-^{131}I separation factor of $\sim 10^5$. Comparisons using 60 mc each of fission-product and (n,γ) reactor-product ^{99}Mo showed no radiocontaminants in the reactor-product generator and in the fission-product generator a progressive decrease in the amount of ^{131}I and ^{103}Ru (150 μc ^{131}I and 40 μc ^{103}Ru in the first milking) in the first three milkings. After the third milking, there were no detectable radiocontaminants (limit of detection, $\sim 10^{-5}$% of ^{99m}Tc).

Preparation of 1.7-h Indium-113m Generator

The separation of the ^{113m}In daughter from 118-d ^{113}Sn was accomplished using Bio-Rad HZO-1, 50 to 100 mesh, inorganic exchanger to retain the parent and eluting with methyl ethyl ketone containing 5 vol % 0.01 M HCl. The first 5 ml of the eluate contained >90% of ^{113m}In. Twenty milkings over a period of one month did not reveal the presence of any radiocontaminants.

Half-life of Cesium-137[15]

An accurate half-life value is necessary to determine the decrease in radiation or power output of ^{137}Cs radiation or heat sources with respect to time. The half-life of ^{137}Cs was determined from the following methods using a 9-1/2-yr-old 1500-curie CsCl source: (1) change in the $^{137}Cs/^{135}Cs$ and $^{137}Cs/^{133}Cs$ ratios determined by mass spectrometry; (2) determination of ^{137}Cs and ^{137}Ba content by weight and mass spectrometry; and (3) determination of specific activity of ^{137}Cs by weight, curie content, and mass spectrometry. The half-life measurements as found by the three methods are summarized in Table 6 with the limits of precision given.

Table 6. Half-life determination of cesium-137

Method number	Number of measurements	Half-life, years	Standard deviation of the mean, years	Limit of error at 95% confidence limit, years
1	6	30.55	± 1.55	± 4.06
2	3	29.78	± 0.14	± 0.64
3	20	30.72	± 0.12	± 0.25

Distribution Coefficients

Distribution coefficients involve the extraction of a desired element and the non-extraction of undesired elements and enable the prediction of separation, purification, and decontamination coefficients. In the extraction of radioactive nuclides, the distribution coefficient, K_d, is defined as:

$$K_d = \frac{\text{Radioisotope in organic phase}}{\text{Radioisotope in aqueous phase}} .$$

Extraction of Iron from HCl Solutions with Bis-(β-chloroethyl)ether

To separate iron from ^{7}Be which is coprecipitated with $Fe(OH)_3$, bis-(β-chloroethyl)ether was used since its high density (lower phase) enables multiple extractions using the same separatory funnel. Ferric chloride solutions of 500, 100, 10, 1.0, 0.10, and 0.010 mM in varying concentrations of HCl were extracted at 22°C with equal volumes of bis-(β-chloroethyl)ether (previously equilibrated with acid of the same concentration). The analysis was made by counting the 1.097-Mev and 1.289-Mev photo peaks using a 3 by 3 in. NaI(Tl) gamma spectrometer. The results are shown in Fig. 4, where the distribution coefficient of iron in bis-(β-chloroethyl)-ether is plotted as a function of the concentration of iron and the concentration of HCl. It is evident from these results that the distribution coefficient (i.e., the extraction of iron by the organic phase) increases with an increase in HCl concentration and a decrease in iron concentration.

Extraction of Indium from HCl Solutions with n-amyl and n-octyl Alcohol

Approximately 1.0 μc of ^{113m}In in 100 λ of methyl ethyl ketone containing $<10^{-4}$% tin was used to obtain the distribution coefficients. To simulate a cyclotron target, 34 mg of carrier indium was added. Distribution data were obtained by shaking 10 ml of the organic extractant (n-amyl or n-octyl alcohol) with 10 ml of the aqueous solution (2-10 M HCl) containing the ^{113m}In activity. After the solutions were shaken and the phases allowed to separate at 22°C, 200 λ aliquots of each phase were taken for counting. Results from the 0.393 ^{113m}In photo peak counted on a 3 by 3 in. NaI(Tl) gamma spectrometer are shown in Fig. 5. Data with n-amyl alcohol and 8 M and 10 M HCl could not be obtained since the phases did not separate.

Effect of Hexane on Distribution Coefficients of Technetium-99m and Iodine-131

From 200 to 1000 μc of generator-produced ^{99m}Tc in methyl ethyl ketone (5 vol % 0.01 M HCl) was used to obtain distribution coefficients (Fig. 6) by shaking equal volumes of hexane-diluted methyl ethyl ketone with H_2O or 2 M NH_4OH. After 20 min of contacting time and phase separation, samples were analyzed by counting of the 0.14-Mev photon with a 3 by 3 in. NaI(Tl) gamma spectrometer. Distribution coefficients (shown in Fig. 7) were obtained from 25-125 μc of ^{131}I also in methyl ethyl ketone (5 vol % 0.01 M HCl) by the same extraction procedures and analyses using the 0.364-Mev photo peak.

Extraction of Manganese-54 from HCl Solution with Bis-(β-chloroethyl)ether

Cyclotron-produced carrier-free ^{54}Mn, in varying concentrations of HCl, was extracted with bis-(β-chloroethyl)ether by shaking 20 ml of the aqueous solution containing 80 μc of ^{54}Mn with an equal volume of the organic solution for 5 min. After a settling time of 30 min, the solutions were centrifuged and 1.00 ml aliquots of each phase taken for counting; distribution coefficients obtained are shown in Fig. 8. A 3 by 3 in. well-type NaI(Tl) and a 3 by 3 in. solid NaI(Tl) gamma spectrometer were used to detect the 0.840-Mev photo peak.

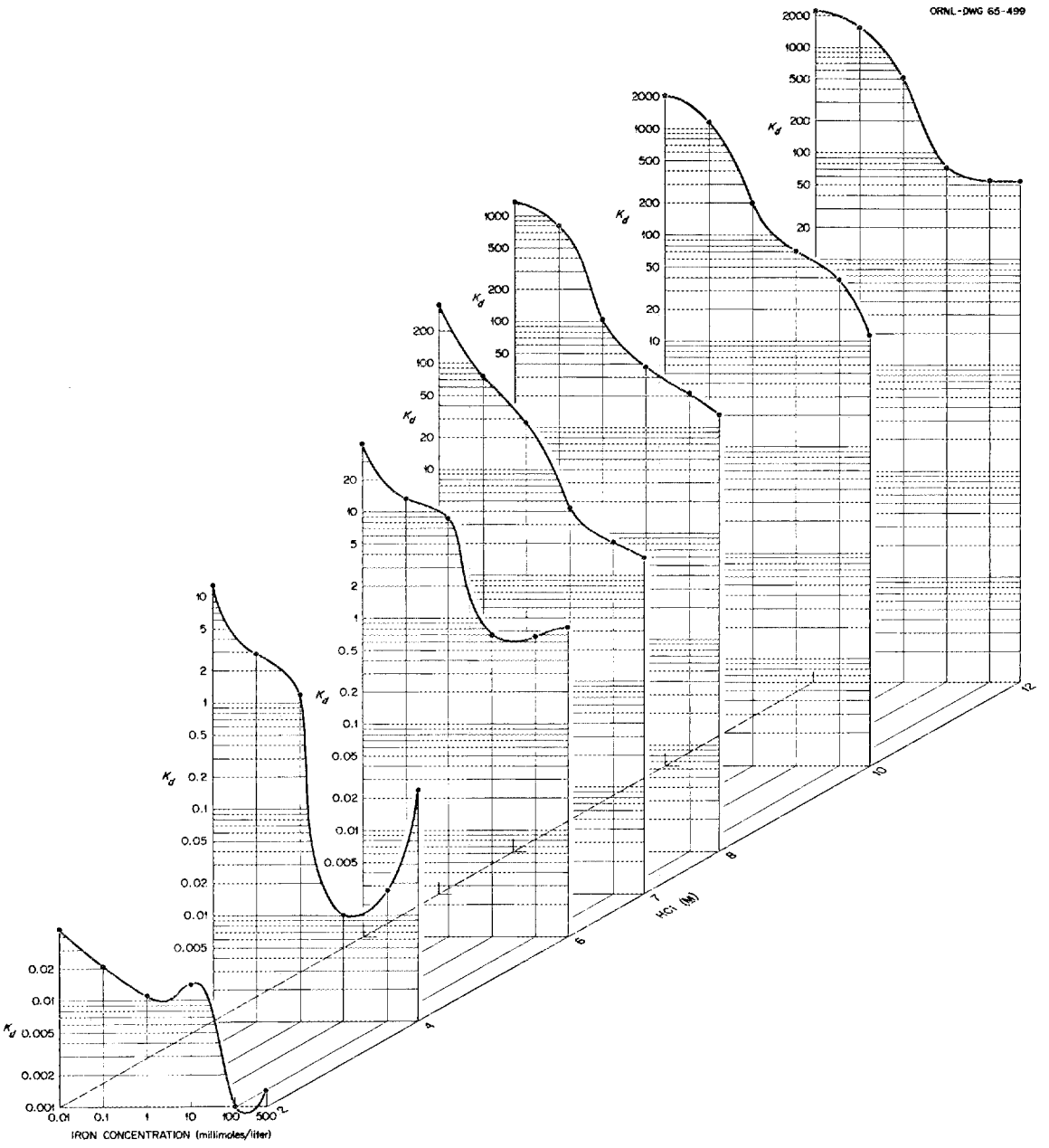


Fig. 4. Effect of HCl Concentration on Distribution Coefficients of Iron in Bis-(β-chloroethyl)ether.

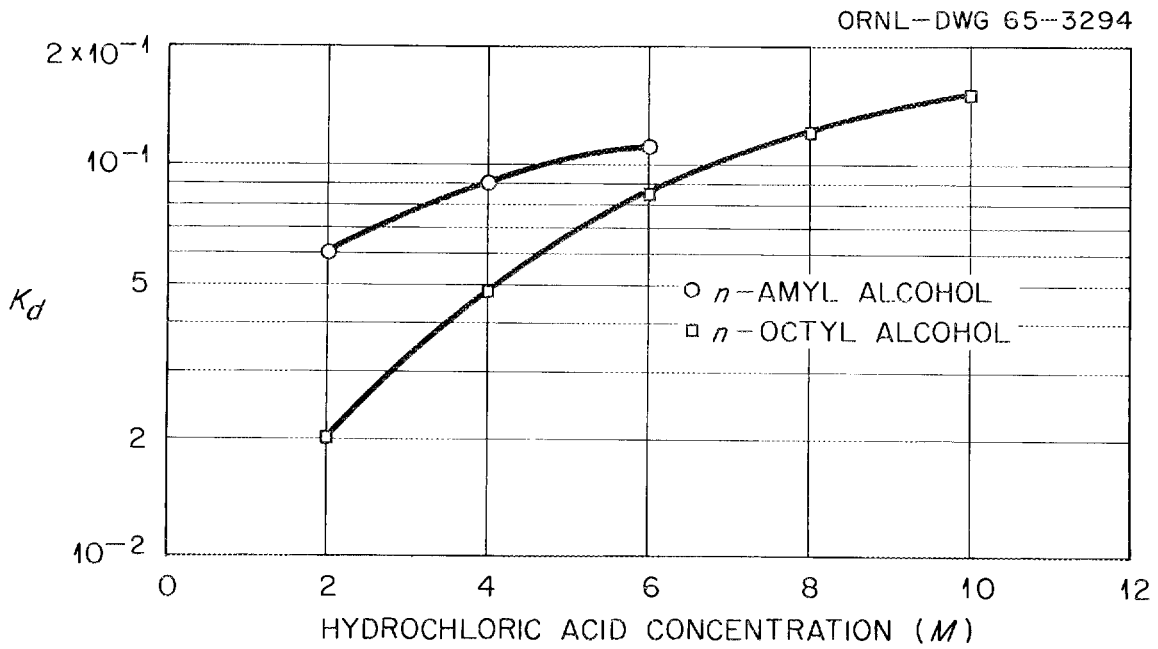


Fig. 5. Effect of HCl Concentration on Distribution Coefficients of Indium in n-Amyl and n-Octyl Alcohol.

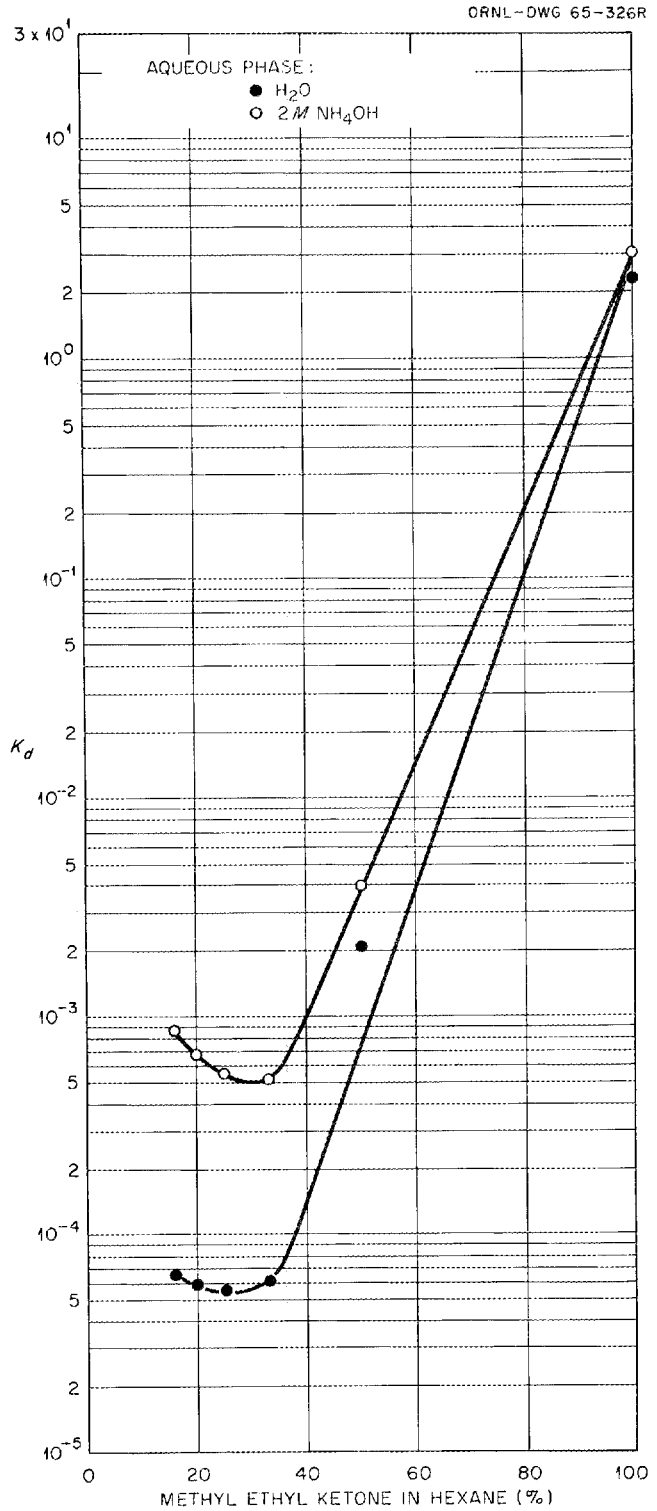


Fig. 6. Effect of Hexane-diluted Methyl Ethyl Ketone Concentration on Distribution Coefficients of Technetium-99m.

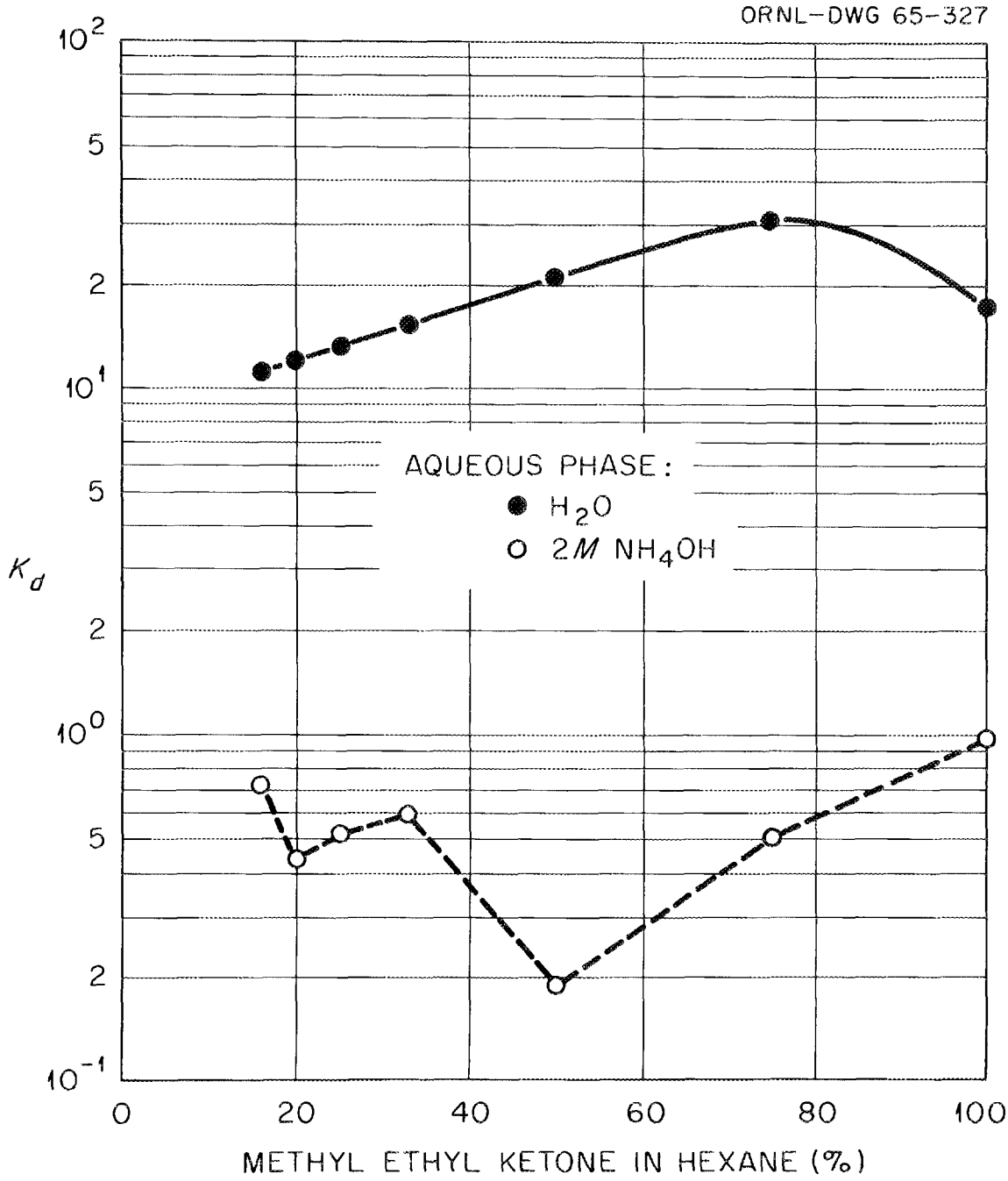


Fig. 7. Effect of Hexane-diluted Methyl Ethyl Ketone Concentration on Distribution Coefficients of Iodine-131.

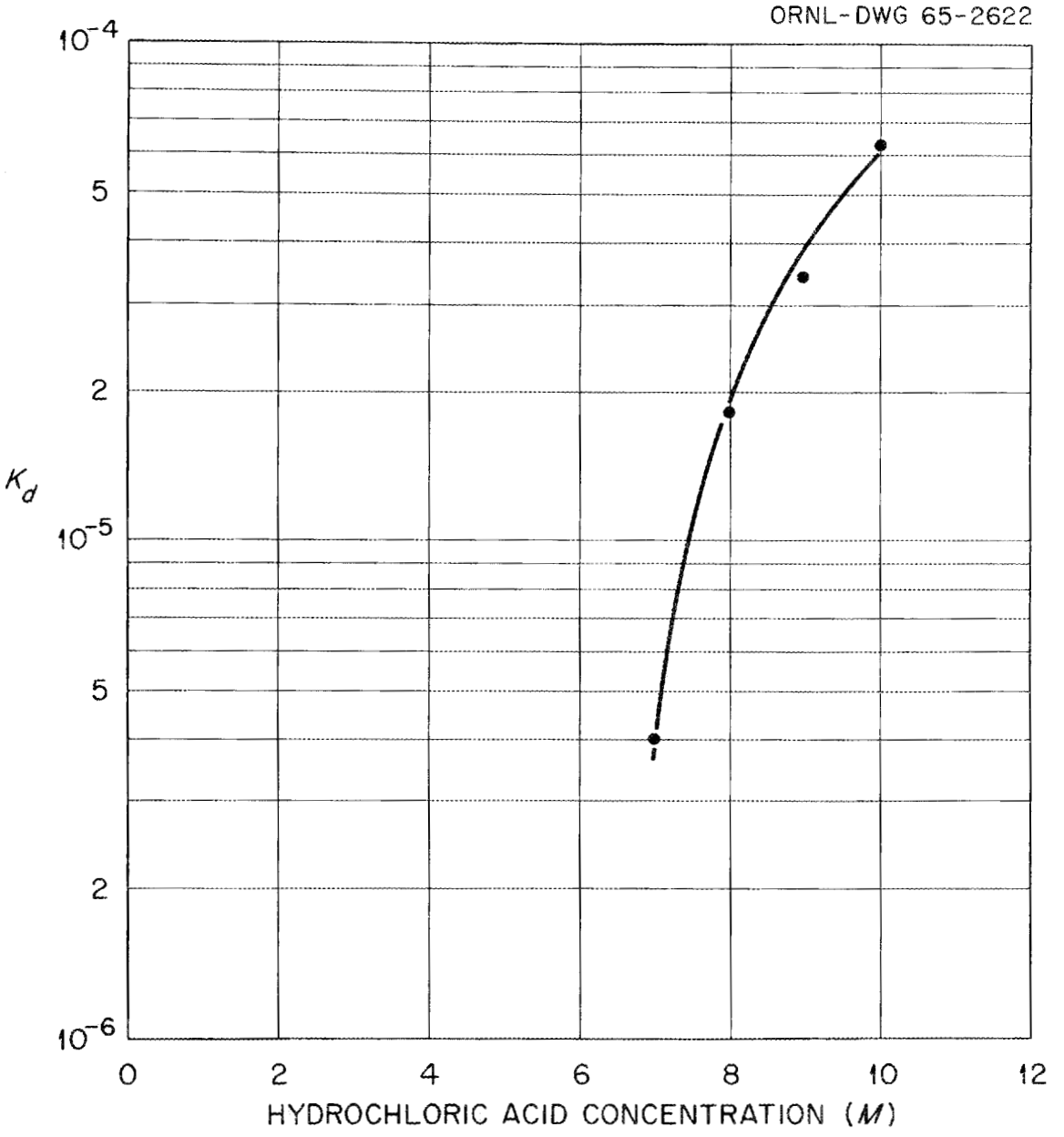


Fig. 8. Effect of HCl Concentration on Distribution Coefficients of Manganese-54 in Bis-(β-chloroethyl)ether.

Elution of Carrier-Free Indium

A Bio-Rad AG-1, X-8, 50 to 100 mesh anion-exchange column (0.5-in. dia by 5-in. long) in the chloride form was charged with ~280 μc of 118-d ^{113}Sn. The 1.73-h ^{113m}In daughter was eluted with various concentrations of HCl at a rate of 1.0 ml/min for ~60 min, during which time 60 1-ml samples of effluent were collected and counted by observing the 390-kev isomeric transition photo peak with a 3 by 3 in. NaI(Tl) crystal gamma spectrometer. Small significant quantities of ^{113}Sn were found in the 0.1 to 1.0 M elution samples several weeks later. The ^{113m}In elution data, corrected for the ^{113}Sn content, are shown in Fig. 9.

The (n,γ) Burnup of Promethium-146 in Promethium-147

Promethium-146 was detected in small amounts among the uranium fission products by Roberts, Wheelwright, and van Tuyl,[16] who pointed out the possible shielding problems this could introduce with very large (megacurie) ^{147}Pm power sources.

The 1600-d half-life of ^{146}Pm precludes complete decay or decay to negligible levels within a reasonable period. To determine if the ^{146}Pm content could be depleted or reduced by re-irradiation of ^{147}Pm in a reactor, it was necessary to determine if the effective (n,γ) cross section were sufficiently large. The effective cross section for the $^{146}Pm(n,\gamma)^{147}Pm$ reaction was estimated as ≤8400 barns from the following data.

High specific activity ^{146}Pm was prepared by the $^{146}Nd(p,n)^{146}Pm$ reaction in the ORNL 86-Inch Cyclotron from 100 mg of ^{146}Nd (96% isotopically enriched as Nd_2O_3) which produced a yield of 13 μc per 8-hr irradiation. The 23-Mev protons (from the 84-μa beam) were degraded to ~12 Mev by the use of aluminum absorbers to reduce the production of other promethium isotopes and to optimize ^{146}Pm production.

After decay of the short-lived radioisotopes, ^{146}Pm was separated and purified by elution with 0.4 M α-hydroxyisobutyric acid using a Bio-Rad 50W column. A gamma-spectrometric determination showed ^{146}Pm with no other radioisotopes present. The ratio of the prominent 0.460- and 0.740-Mev lines, used as an index of the radiochemical purity at each step, was essentially constant (e.g. 2.035 at one step and 2.037 at another step). A 2.10-μc sample of ^{146}Pm was irradiated for 72 hr in hydraulic tube No. 1 of the ORR in the following neutron flux:

Thermal	1.22×10^{14} n/cm^2.sec
1.5-2.0 Mev	8.35×10^{12} n/cm^2.sec
3.0 Mev	3.8×10^{12} n/cm^2.sec

The gamma spectrum of the irradiated ^{146}Pm was followed for several weeks until the prominent 11.1-d ^{147}Nd radioisotope [produced by the (n,γ) reaction on the inactive neodymium present] had decayed to negligible quantities. The gamma-spectrometric assay showed 1.54 μc of ^{146}Pm, hence a loss of 0.54 μc.

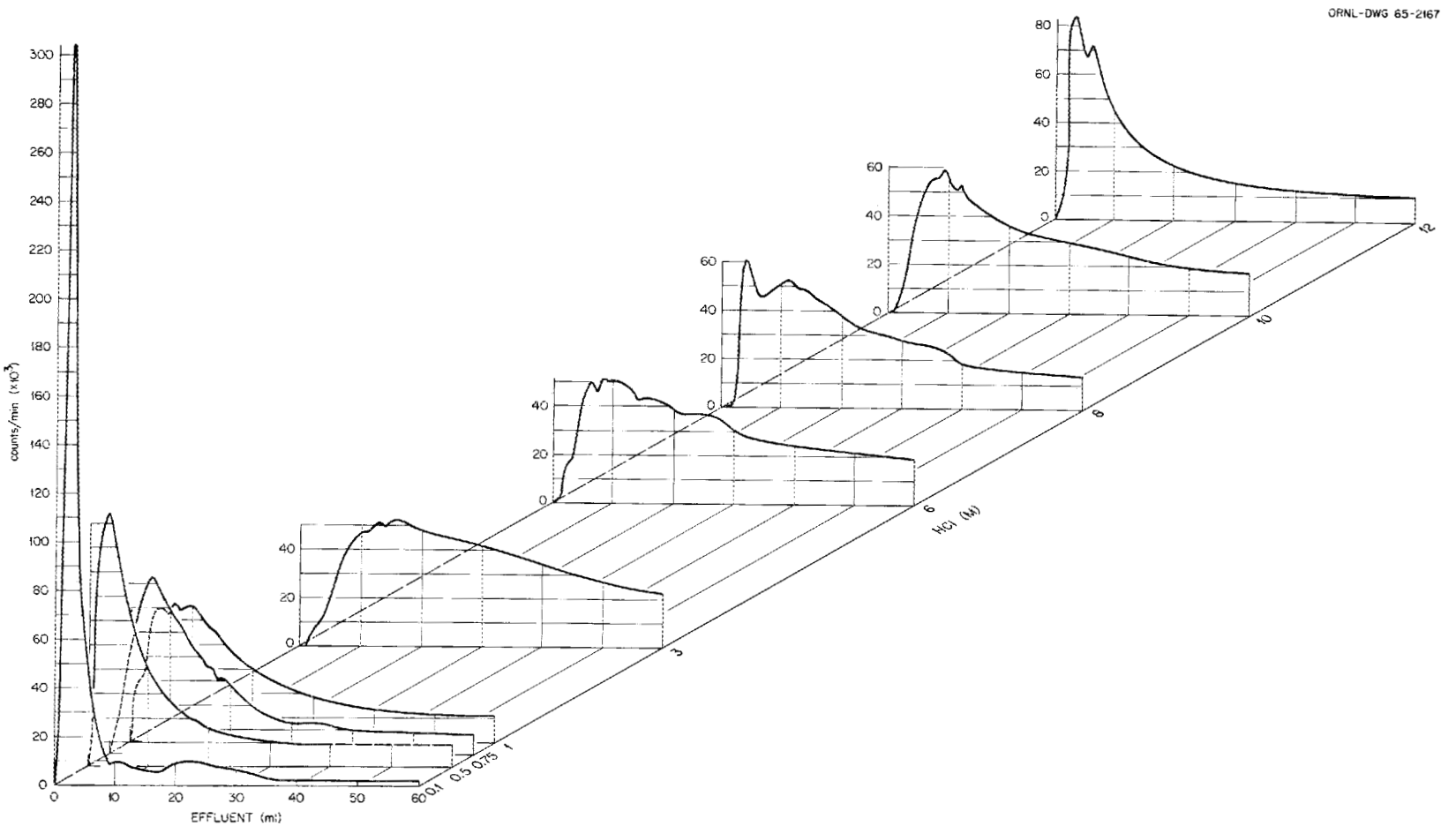


Fig. 9. Effect of HCl Concentration on Elution of Indium-113m.

Preliminary analyses have been made on the system shown in Fig. 10 using the ISOCRUNCH Program[17] which was converted to the CDC-1604-A computer. The computations, shown in Table 7, were made for the LITR, ORR, and HFIR to determine the reduction of ^{146}Pm by (n,γ) burnup, assuming an aged ^{147}Pm source (i.e. no ^{148m}Pm) and an irradiation in a well-thermalized flux.

Table 7. Reduction of promethium-146 by (n,γ) burnup in the LITR, ORR, and HFIR

Flux, $n/cm^2 \cdot sec$	Irradiation time	Atoms			$^{147}Pm/^{146}Pm$ ratio	Increase in ratio
		^{148m}Pm	^{146}Pm	^{147}Pm		
0	0	0	1	1×10^6	10^6	--
2×10^{13}	166.0 days	2.34×10^3	0.836×10^{-1}	0.833×10^6	10^7	10.0
2×10^{13}	333.0 days	1.08×10^3	0.689×10^{-2}	0.693×10^6	10^8	99.4
2×10^{13}	499.0 days	0.901×10^3	0.576×10^{-3}	0.578×10^6	10^9	996.5
2×10^{14}	16.3 days	3.32×10^3	0.932×10^{-1}	0.927×10^6	10^7	10.1
2×10^{14}	32.5 days	3.08×10^3	0.881×10^{-2}	0.859×10^6	10^8	102.6
2×10^{14}	49.0 days	2.85×10^3	0.798×10^{-3}	0.795×10^6	10^9	992.5
2×10^{15}	39.0 hr	3.45×10^3	0.945×10^{-1}	0.936×10^6	10^7	10.1
2×10^{15}	78.0 hr	3.23×10^3	0.893×10^{-2}	0.877×10^6	10^8	101.8
2×10^{15}	117.0 hr	3.02×10^3	0.843×10^{-3}	0.821×10^6	10^9	1026.8

To achieve, by decay, a ratio of $^{148m}Pm/^{146}Pm$ which is <1, the number of half-lives of ^{148m}Pm (41 d) which must be expended is shown in Table 8.

Table 8. Number of half-lives of promethium-148m necessary to reduce $^{148m}Pm/^{146}Pm$ ratio to <1

Reactor	$^{147}Pm/^{146}Pm$ ratio	Decay periods of ^{148m}Pm, half-lives
LITR	10^7	15
	10^8	18
	10^9	21
ORR	10^7	15
	10^8	19
	10^9	22
HFIR	10^7	15
	10^8	19
	10^9	22

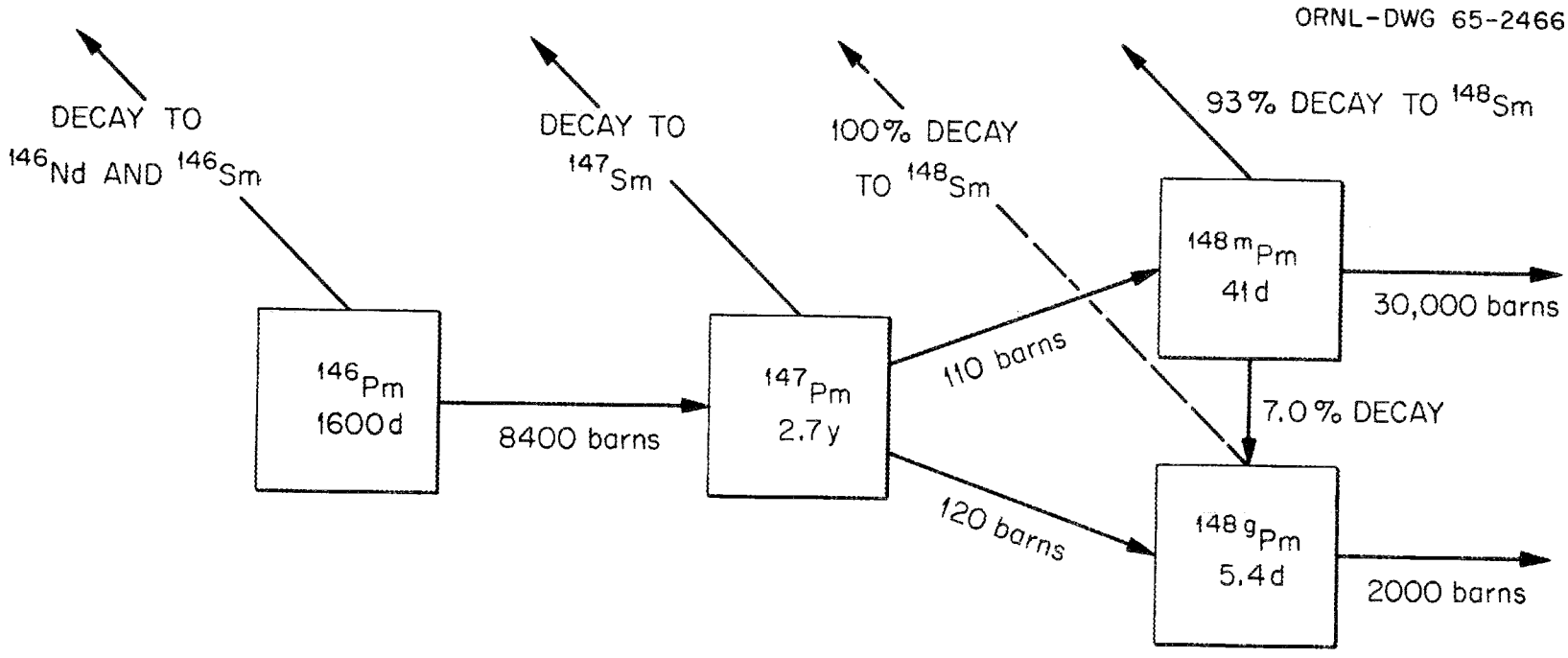


Fig. 10. Diagram of Neutron Activation and Decay of Promethium-146, Promethium-147, and Promethium-148.

The specific gamma-ray emission (Γ), in r/mc.hr at 1 cm, is given in Table 9. The specific gamma emission of ^{148m}Pm is about twice that of ^{146}Pm, so that an additional half-life must lapse when one considers the relative dose rates.

Table 9. Specific gamma-ray emission (Γ) of promethium-146 and promethium-148m

Nuclide	Photon, kev	Intensity (I), quanta/decay	Γ[d], r/mc.hr at 1 cm	I x Γ, r/mc.hr at 1 cm
^{148m}Pm[a]	98	0.07	0.46	0.03
	188	0.01	1.00	0.01
	286	0.10	1.64	0.16
	309	0.05	1.80	0.09
	412	0.18	2.43	0.44
	431	0.08	2.58	0.21
	498	0.07	2.95	0.21
	548	0.91	3.30	3.00
	627	0.94	3.70	3.48
	723	0.31	4.30	1.33
	913	0.18	5.30	0.95
	1011	0.20	5.80	1.16
				11.07
^{146}Pm[b]	460	0.69	4.10	2.83
	740	0.49	2.90	1.42
				4.25
^{146}Pm[c]	453	0.65	3.86	2.51
	745	0.30	4.40	1.32
	749	0.35	4.44	1.55
				5.38

[a] C. W. Reich, R. P. Schuman, J. R. Berreth, M. K. Brice, and R. L. Heath, Radioactive decay of promethium-148 and promethium-148m, Phys. Rev. 127: 192-204 (1962).

[b] I. M. H. Pagden, R. Jakeways, and F. C. Flack, The decays of promethium-143, promethium-144, and promethium-146, Nucl. Phys. 48: 555-60 (1963).

[c] E. F. Funk, Jr., J. W. Mihelich, and L. F. Schwerdtfeger, Radioactive decay of promethium-143, promethium-144, and promethium-146, Phys. Rev. 120: 1781-9 (1960).

[d] J. J. Pinajian, Ph.D. Thesis, Purdue University, 1955 (unpublished).

Computer Programming for Isotopes Calculations

The calculation of reactor yields or thermal outputs of isotopes usually requires lengthy and often repetitive computations which can be readily accomplished using a computer program. Yields for different flux values and irradiation times can be obtained in a few minutes of computer time while calculation by hand may take weeks. Also, compilations on yields or power outputs of isotopes can easily be kept up to date by changing the necessary input data as better cross-section values, half-lives, etc. become available.

Two computer programs developed to handle reactor yield calculations are ISOTOPES and ISOCRUNCH. Program ISOTOPES[18] was developed to calculate the optimum time of neutron irradiation for maximum yield, the specific activity of the product isotope in curies per gram of target material, and the combined specific activity of the target and product isotopes for any general first-order reaction or decay problem. The program was written in FORTRAN-63 language for the Control Data 1604-A Computer. Program ISOCRUNCH was developed by adding optional programs to the basic CRUNCH[19] code and modifying the input and output data formats. ISOCRUNCH can be used to compute the amount of each isotope in a reaction and decay chain for any specified neutron flux and time, to sum the contributions of various chains to the same isotope, to plot the yield of an isotope vs time for a given flux, and to find the optimum time for maximum yield of an isotope in a chain. The basic theory and use of ISOCRUNCH has been reported[17] with sample problems and the FORTRAN calling program included.

Computer program ISOPOWER was developed to calculate the power output of radioisotopes in different units (watts/g, watts/cm^3, and curies/g). The program will sum the contributions to the total power output from all isotopes in a decay chain and from different chains of a mixed radioisotopic power source. The results can be printed out and plotted as power output vs time curves on an associated plotter. The program will be described in a report to be published with a sample problem section and a listing of the FORTRAN II calling program.

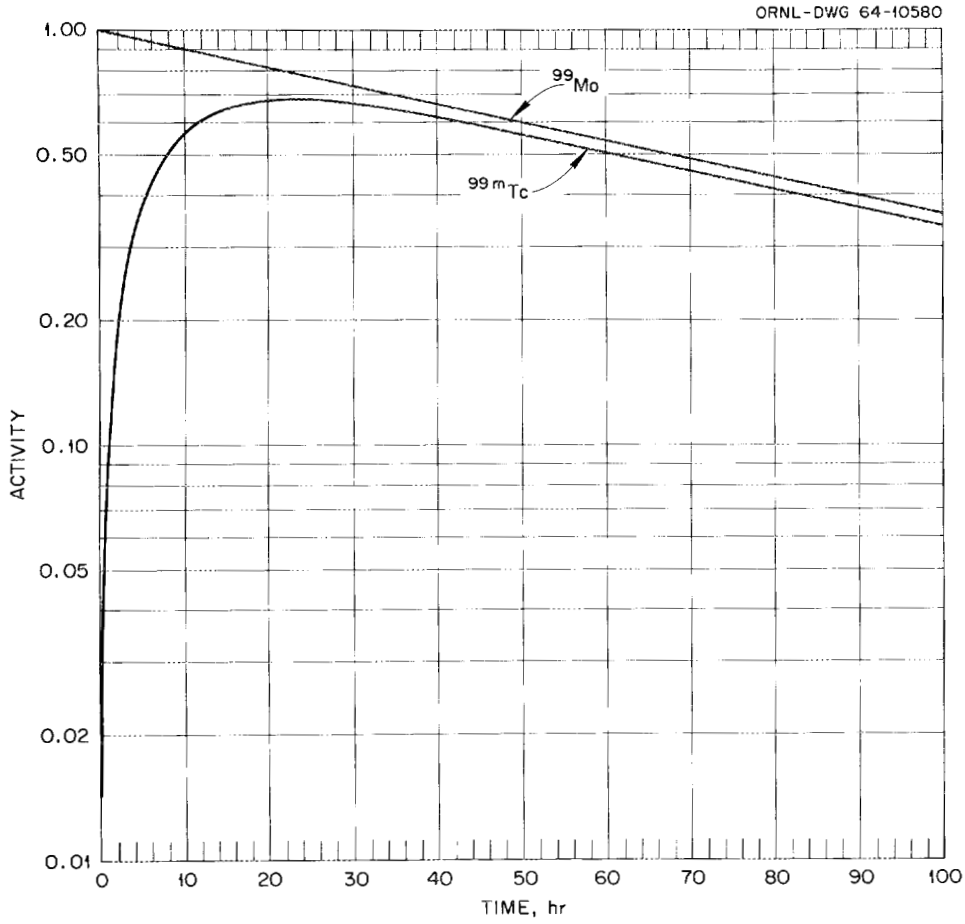
Two computer programs[20] have been written; one of which calculates the fraction of a radioactive isotope remaining after a specified period of time in different units (sec, min, hr, days, and years). A single output page contains the necessary decay factor corrections for all the radioisotopes listed in the ORNL Radio and Stable Isotopes Catalog. The other computer program was written to calculate the growth of the radioactive daughter and decrease of parent. The exact solution to the equation,

$$\lambda_B N_B = \frac{\lambda_A N_A T_A}{T_A - T_B} \left(1.0 - e^{[(T_B - T_A)\lambda_B t]/T_A}\right) ,$$

where

N_A and N_B = number of parent and daughter atoms, respectively,
λ_A and λ_B = decay constant of parent and daughter, respectively,
T_A and T_B = half-life of parent and daughter, respectively, and
t = time,

was programmed to give the output in two forms — graphic and tabular — e.g., the output for ^{99m}Tc in Fig. 11.



Time, hr	Activity of ^{99m}Tc
.00	.00
2.00	0.176
4.00	0.311
6.00	0.415
8.00	0.495
10.00	0.554
12.00	0.598
14.00	0.630
16.00	0.652
18.00	0.666
20.00	0.674
22.00	0.678
24.00	0.678
26.00	0.675
28.00	0.670
30.00	0.665
32.00	0.655
34.00	0.646
36.00	0.636
38.00	0.626
40.00	0.615
42.00	0.604
44.00	0.593
46.00	0.582
48.00	0.571
50.00	0.560
52.00	0.549
54.00	0.538
56.00	0.528
58.00	0.517
60.00	0.507
62.00	0.497
64.00	0.487
66.00	0.477
68.00	0.467
70.00	0.458
72.00	0.448
74.00	0.439
76.00	0.430
78.00	0.421
80.00	0.413
82.00	0.404
84.00	0.396
86.00	0.388
88.00	0.380
90.00	0.372
92.00	0.365
94.00	0.357
96.00	0.350
98.00	0.343
100.00	0.336

Fig. 11. Computer Program Output in Graphic and Tabular Form for Technetium-99m.

RADIOISOTOPE TECHNOLOGY

Process Improvement

Carbon-14

The ^{14}C process was reviewed to determine yields from irradiated Be_3N_2 and isotopic purity of ^{14}C produced under controlled conditions. The principal ^{14}C compound evolved during dissolution of the Be_3N_2 in H_2SO_4 was methane, indicating that the ^{14}C produced by the $^{14}N(n,p)^{14}C$ reaction in the Be_3N_2 target is in the form of Be_2C. Forms of carbon in the target other than carbide are released as CO and CO_2. These gases can be separated from the ^{14}C methane by reaction with cobalt oxide at high temperature before the ^{14}C methane is converted to CO_2. After a caustic scrubber was installed in the process gas stream ahead of the high temperature converter and the converter medium was changed from copper oxide to cobalt oxide, the ^{14}C yield increased ~50%.

Isotopic purity of ^{14}C increased to an average of 90% from ~80% before the process change. Since any normal carbon present as Be_2C in the target material cannot be separated in processing, future gains in isotopic purity can be made only by target purification or increased neutron irradiation time.

Phosphorus-32

An interfering precipitate which hindered various ^{32}P tagging procedures was observed when the HCl solution of orthophosphoric acid was adjusted to pH 8. Chromatographic analysis revealed the presence of orthophosphoric acid and interfering metal phosphates.

Simulated ^{32}P process runs using unirradiated sulfur failed to yield a precipitate when the residual solution (HCl) was adjusted to pH 8-9, which indicated that the interfering precipitate was introduced by the irradiation of containers or reagents used in processing. The procedure was modified by rinsing the aluminum irradiation can with molten sulfur rather than treating it with concentrated HNO_3, and by substituting a quartz distillation vessel for the platinum vessel previously used during the distillation procedure. Since initiation of these two process changes, the occurrence of precipitate at pH 8 has been reduced below limits of detection.

Xenon

As uses of ^{133}Xe increase, especially in medical diagnosis, product purity becomes more important. Reduction of atmospheric gas contamination and oxides of nitrogen to ~10% of total xenon present was accomplished by increased vacuum integrity and the addition of a high temperature calcium trap. Prior to this improvement, the atmospheric gases were present to the extent of 80-90% on a routine basis. New process equipment is being designed to reduce contamination to <5% on routine products.

Xenon-131m (12 d) has been produced by the $^{129}I(n,\gamma)^{130}I \xrightarrow{\beta^-} {}^{130}Xe(n,\gamma)^{131m}Xe$ reaction. The usual method for the preparation of ^{131m}Xe is by collection of the beta decay product of ^{131}I; however, the irradiation of ^{129}I is less costly. In some applications, ^{131m}Xe can be used in place of ^{133}Xe.

Tritium

Loading ^{3}H on uranium traps to remove ^{3}He has been done without controlling the efficiency of uranium tritide conversion. Since this purification step represented ~50% of the ^{3}H loading and processing efforts, any improvement in the loading efficiency has a large effect on cost associated with the ^{3}H process. Installation of a monitor in the discharge line from the traps permits uranium to be loaded to near capacity without loss of ^{3}H by overloading.

Iodine-131

Designs for modifying the present ^{131}I processing cells to make them more adaptable for the changing ^{131}I production requirements have been started. Prototype equipment for two proposed ^{131}I production methods was fabricated and tested. The first process, called the dry process, involved melting the uranium-aluminum target and recovering the ^{131}I and ^{133}Xe from the gases given off during melting. A quartz vessel placed in an RF heating coil was used to make several experimental runs with simulated and actual target rings. During initial runs, >90% of the iodine released during melting plated out on the walls of the melting vessel and did not reach the recovery traps. Flowing helium through the vessel during melting to carry the iodine improved the recovery ~75%, but the precise control needed for a satisfactory recovery was not desirable for production operation; therefore, equipment development for this process was discontinued. During three of the test runs, experiments were conducted to obtain an idea of the losses that might be expected due to the plating of iodine on the stainless steel process lines. The iodine in the carrier helium was passed through a trap containing type 316 stainless steel raschig rings of known surface area on which ^{131}I had plated to the extent of 0.14 curie/in.^{2} of exposed surface.

The second proposed process required a pressurized dissolver in which the target ring is dissolved in a closed system. After complete dissolution, the gas is bled off slowly to recover ^{133}Xe and the solution is processed to recover ^{131}I. A prototype dissolver was designed and fabricated and several test runs with simulated target rings were made at pressures up to 1400 psig to determine temperature and pressure data for different dissolving conditions.

Dissolution of Noble Metals by Alternating Current

A simple procedure has been developed for the dissolution of the noble metals in HCl by using half-wave and full-wave alternating current electrolysis. The rates of solution for Au, In, Ir, Pd, Pt, and Rh were investigated along with those of several other metals resistant to HCl. Tests were also conducted on the dissolution rate of gold to determine the effect of varying the frequency of the alternating current.

The equipment is easily constructed from two 10-amp variacs, a 12-amp silicon diode, and electrical leads. Only the primary windings of the second variac are used. Three turns of No. 6 copper wire are wound around this coil to produce 0-10 volts as a supply to the electrodes. The silicon diode is connected in an alternate leg of the system and supplies half-wave instead of full-wave current. The metal to be dissolved forms one of the electrodes and graphite the other.

Dissolution rates are a function of acid concentration, current density, and frequency of applied current. In most cases, the fastest rate will be produced by the most concentrated acid, the highest current density, and a low cycle of alternation. The dissolution rates in 9 M HCl using 60 cps full-wave and half-wave current and current densities are listed in Table 10.

Table 10. Rates of dissolution for various metals in 9 M HCl using 60 cps alternating current and current densities

Metal	Dissolution rate, mg/hr.in.^{2}		Current density, amp/cm^2	
	Full-wave	Half-wave	Full-wave	Half-wave
Gold	103	998	0.62	0.46
Cadmium	2550	5172	0.62	--
Copper	2426	2078	0.77	0.38
Indium	3657	2070	0.62	0.62
Iridium	22	0	0.46	--
Palladium	744	1596	0.54	0.31
Platinum	66	0	0.69	--
Rhodium	172	0	0.46	0.31
Tungsten	62	25	0.46	--

The acid concentration of the electrolyte has a marked effect on the rate of dissolution. Every metal except iridium dissolves at a faster rate in 12 M than it does in 3.2 M HCl. A number of metals dissolved at a greater rate when half-wave instead of full-wave current was applied. The use of half-wave current increased the dissolution rate for gold in 9 M HCl by a factor of 10. Cadmium and palladium dissolution increased by a factor of 2 and copper, indium, and tungsten dissolved at approximately the same rate. Platinum, iridium, and rhodium did not dissolve in HCl using the half-wave method.

The effect of varying the frequency of the alternations in the current supplied to the electrodes was studied. The dissolution rate of gold in 9 M HCl was investigated using alternating currents of 45, 60, and 100 cps. At 45 cps the dissolution rate was 1872 mg/hr.in.^{2}. The rate dropped to 998 mg when the frequency was increased to 60 cps and dropped further to 235 mg when 100 cps current was applied.

Agitation during dissolution became necessary as the metal in solution approached stoichiometric quantities. Metals that dissolved slowly exhibited very little change in the rate of solution due to agitation. Metals that had a fast rate of solution definitely dissolved at even faster rates when the electrolyte was agitated. Without agitation, depletion of the acid in the immediate vicinity of the metal being dissolved slowed the rate.

The rates of dissolution for some of the metals were increased by the addition of fluoride to the bath. A solution of 1.4 M HF in 3 M HCl was sufficient to increase the rate for iridium metal from 33 to 42 mg/hr·in.^{2}. The rate for platinum increased from 17 to 23 mg and the tungsten rate increased from 62 to 397 mg/hr·in.^{2}.

Tantalum metal did not dissolve in HCl. The addition of 1.4 M HF, however, enabled the dissolution to proceed at a rate of 82 mg/hr·in.^{2}.

Crystallization of Cesium Tetraoxalate[21]

Cesium tetraoxalate proved to be a useful intermediate compound for converting cesium alum and other cesium concentrates to useful chemical compounds from which many other compounds can be readily prepared.[22] Cesium tetraoxalate was crystallized from cesium-rich solutions with >90% yield under optimum conditions. In converting the cesium alum to tetraoxalate, a yield of 70% was achieved with <1% cocrystallization of the Al^{3+}. Any cesium remaining in solution was recycled and over-all process losses were low. Cesium tetraoxalate is sufficiently stable under radiation to be used as an intermediate in processing fission product ^{137}Cs.

Cesium tetraoxalate was calcined at 450°C to Cs_2CO_3 which was readily converted to cesium glasses or cesium salts. The process was demonstrated at FPDL and accepted for routine use for preparing multikilocurie quantities of $^{137}CsCl$ and cesium glasses. The FPDL fission product cesium recovery and purification process in which cesium tetraoxalate was used is shown in Fig. 12.

During 1963 and 1964, ~700,000 curies of ^{137}Cs as cesium alum was converted to useful source compounds by the cesium tetraoxalate process. During continuous operation, cesium alum has been converted to cesium chloride at a rate >0.05 megacurie/week. All $^{137}CsCl$ products made in 1963 and 1964 were prepared by this process.

Radioisotope Fuels Environmental Test Laboratory

The conceptual design has been completed for a facility to test isotopic power supplies under simulated space and deep ocean conditions. The proposed building, called the Radioisotope Fuels Environmental Test Laboratory, will have ~25,000 ft^2 of floor space and will contain a 15-ft-dia by 25-ft-high vacuum chamber for full system tests, four smaller vacuum chambers for component testing, and a 4-ft-dia by 10-ft-long pressure chamber capable of handling pressures equivalent to ocean depths of 30,000 ft. The proposed building will also contain a hot cell for fueling

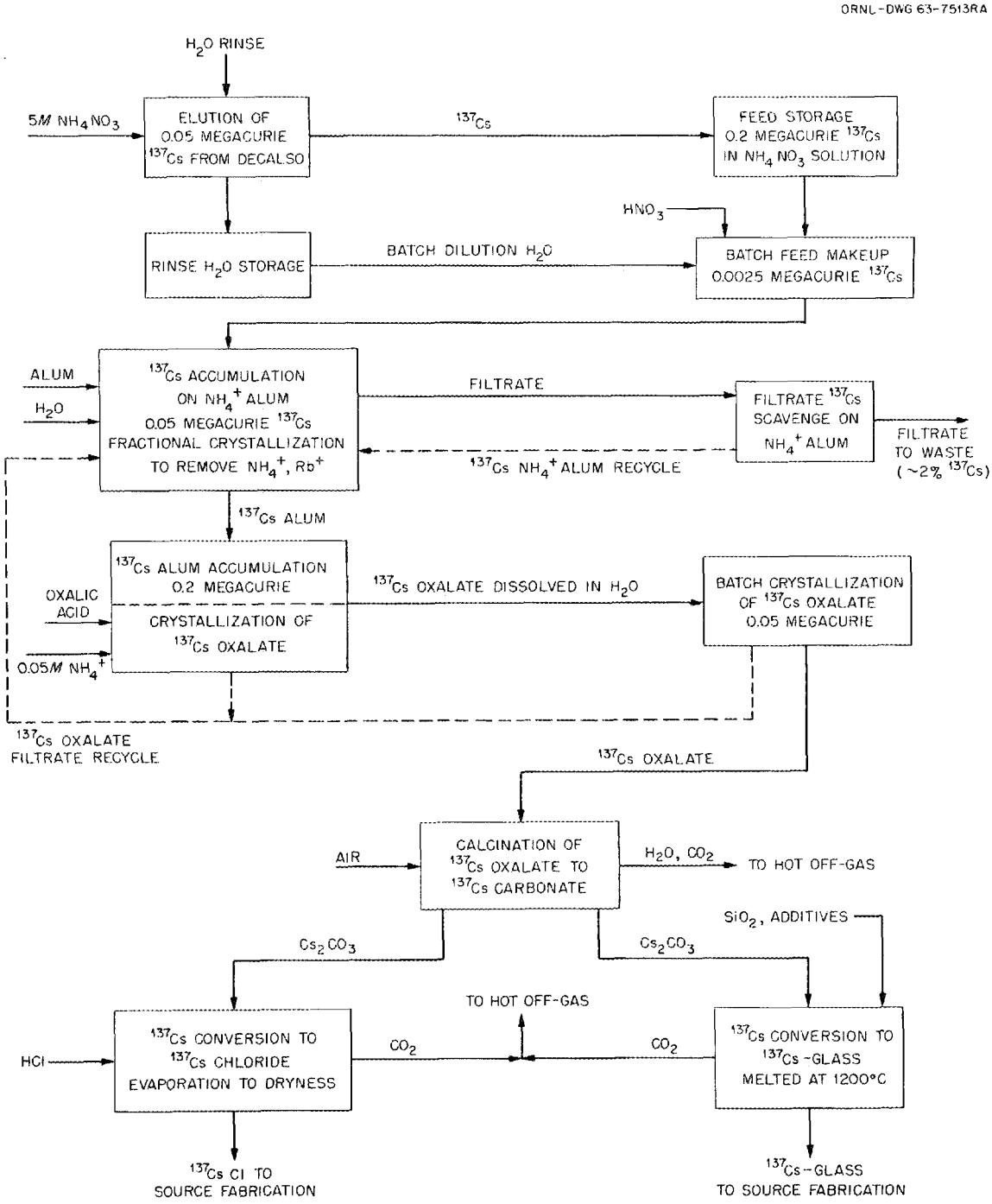


Fig. 12. FPDL Cesium-137 Processing.

experimental devices, and facilities for handling both shielded and unshielded isotopic power units. Tests required for flight approval and facilities for developing new conversion devices and handling techniques will also be provided in the new laboratory.

Cooperative Programs

Cooperative Test of Cobalt-60-Powered Thrustor*

A ^{60}Co-powered space Thrustor prototype was tested in a hot cell in cooperation with Marquardt Corporation and U. S. Air Force personnel. Heat is produced in heat exchanger plates by absorption of the gamma radiation from 50,000 curies of ^{60}Co metal in the form of 74 fuel pins. The heat is then transferred to hydrogen gas flowing between the plates and exiting through a rocket nozzle. The maximum temperature achieved during the tests was 1280°F under no-flow conditions at the exit nozzle. The fuel pins operated at only 50°F lower than the heat exchanger exit temperature due to inadequate insulation.

Cooperative Test of Strontium-90-Powered Thermoelectric Unit

The largest $^{90}SrTiO_3$ pellets ever prepared were fabricated for an RCA thermoelectric generator test device which required 83,000 curies in a single unit. Green pellets of 4.06-in. dia were pressed from calcined $^{90}SrTiO_3$ powder and sintered at 1400°C. The pressed pellets had diameters of ~3.5 in. and densities averaging 3.8 g/ml. Eight pellets of ~15,000 curies each were prepared; six of them were used in the final source. After storage the pellets developed cracks from internal thermal stress but none were damaged beyond use. The pellets were doubly encapsulated in welded capsules to supply power for the converter. The fuel capsule was loaded remotely into the generator which operated in a vacuum chamber for >1400 hr at a surface temperature of 1100°C. The over-all thermal-to-electrical efficiency of the thermoelectric generator was 4.6% and there was no degradation of converter efficiency during the test.

Krypton-85 Enrichment System

Work has started on the installation of a thermal diffusion process for isotopically enriching ^{85}Kr. The system will have six 36-tube electrically heated columns arranged in two 3-column cascades, a shielded loading-unloading station, a cooling tower for column heat disposal, and remotely operated valves, pumps, and controls. With ~5% feed material about 56 days will be required for batch operation through the two cascades to give an annual production rate of 3000 curies of 45% ^{85}Kr or correspondingly greater amounts of lesser enrichments. Installation of the equipment in an existing cell block with 2 ft of concrete shielding will be completed during the first quarter of 1965.

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*Marquardt Corporation tradename.

Source Development

Technetium-99

Oxide-free ^{99}Tc metal was electrodeposited onto various metal foils from a dilute aqueous solution of ammonium pertechnetate in acidic ammonium oxalate with a yield of >99%. This procedure should be useful for large-scale preparation of ^{99}Tc metal because of high yield, high purity, and simplicity of operation, and for the preparation of beta sources which are used in the medical profession[23] to coagulate blood and inhibit the formation of scar tissue from keloid-forming material. The 0.29-Mev beta rays from ^{99}Tc penetrate deep enough to stop blood flow but, because of their low energy, do not cause significant radiation damage. Technetium-99 need not be recalibrated frequently because of its 10^5-y half-life. It has a simple decay scheme, decaying 100% to the ground state by beta emission.

Oxides of technetium have been electroplated from acid solution by Perrier and Segre[24] who first demonstrated many of its chemical properties, by Glass and Bleidner[25] in their work on electrochemistry, and by Motta, Larson, and Boyd[26] in their studies of tracer chemistry. Rogers[27] used oxide deposition to separate technetium from molybdenum and rhenium. Matsura and Yumoto[28] prepared pure technetium oxide beta sources on thin foils by deposition from an alkaline solution.

Motta, Larson, and Boyd[26] prepared the metal by electrodeposition from a 1 M H_2SO_4 solution containing a small quantity of fluoride which increased their yield from 25 to 75%. Although Eakins and Humphries[29] were able to plate the metal from 4 M H_2SO_4, the addition of fluoride did not improve their yield and deposition stopped after ~25% of the technetium had been plated. This was due to an accumulation of a brown-colored reduction product of the ammonium pertechnetate in the bath and required continuous addition of peroxide to convert this compound back to the pertechnetate.

The acid concentration necessary for the deposition of metal rather than oxide was interrelated to the oxalate content of the electrolyte. If the oxalate concentration were reduced, the acid concentration had to be increased for metal deposition. Technetium-99 recovery on copper was >99% complete with a metal purity of 99.0 and 99.3% as determined by polarographic and coulometric analyses, respectively. Table 11 shows the H_2SO_4 concentration required to produce an adherent ^{99}Tc metal or oxide film on various metals from a 0.7 M oxalate solution.

A satisfactory current density for deposition was 1.3 amp/cm^2. Current densities >1.3 amp/cm^3 produced a metal deposit which did not adhere to the cathode when dry.

Table 11. Sulfuric acid concentration necessary to plate technetium-99 metal and oxide

Metal	H_2SO_4 concentration, molarity for metal	for oxide
Copper	0.45	0.32
Nickel	0.87	0.58
Aluminum	1.14	0.71
Silver	1.41	0.87
Gold	1.41	0.87
Stainless steel	1.41	0.87
Platinum	1.90	1.14

Examination of metallic deposits under a 750-power microscope showed that technetium was deposited on the surface of the foil in the form of microscopic spheres ranging in size from 0.00008 to 0.0007 in. in diameter (Figs. 13 and 14). These spheres were built up in layers, with the lower layer completely covering the foil and subsequent layers forming pyramids of metal commonly called "trees." As the thickness of the deposit increased, the voids were filled and new layers of pyramids formed. The spheres were easily broken from the tops of the pyramids by paper wiped across the surface of the deposit which picked up a reading of ~20,000 dis/min. Readings from subsequent wipes decreased until the fifth reading was <10 dis/min above the background reading.

The purity of ^{99}Tc metal (produced by hydrogen reduction of the oxide) was improved by lowering the reduction furnace temperature from 1000°C to 700°C. Metal produced prior to this purification contained ~10% oxygen and 1,000 to 100,000 ppm silicon. The cooling cycle after reduction was changed to maintain hydrogen flow over the metal until room temperature was reached. The new cycle produced >99% ^{99}Tc metal with an oxygen content of <1%.

Americium-Beryllium-Curium

A method for fabrication of neutron sources composed of a mixture of ^{242}Cm, ^{241}Am, and beryllium was developed and tested. This unique system permits fabrication of the source under low neutron yield by encapsulating a pressed pellet composed of ^{241}Am oxide on powdered beryllium metal, followed by neutron irradiation in a reactor to produce ^{242}Cm by the $^{241}Am(n,\gamma)^{242}Am \xrightarrow{\beta} {}^{242}Cm$ reaction. Neutron yields of 10^7 have been obtained using 0.010 g of ^{241}Am. Some gamma activity is associated with the neutron yield due to fission products from the ^{241}Am. Figures 15 and 16 show the energy curves of ^{241}Am-Be-^{242}Cm sources.

Iridium-192

High specific activity ^{192}Ir radiographic sources were prepared by irradiating thin ~2-mil iridium metal ribbons and then fusing them into spheres

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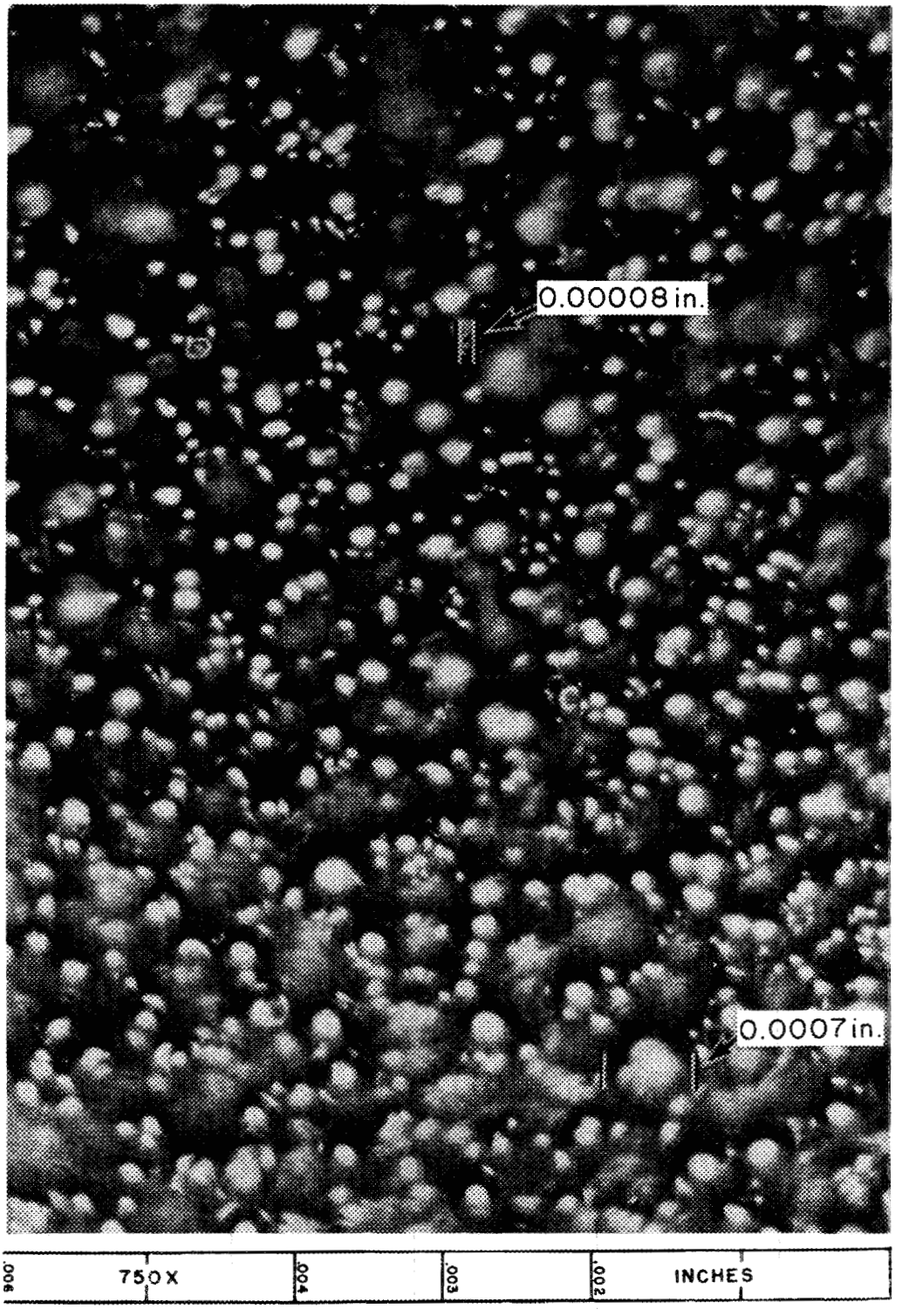

Fig. 13. Photomicrograph of Electrodeposited Technetium-99, Reduced 8%.

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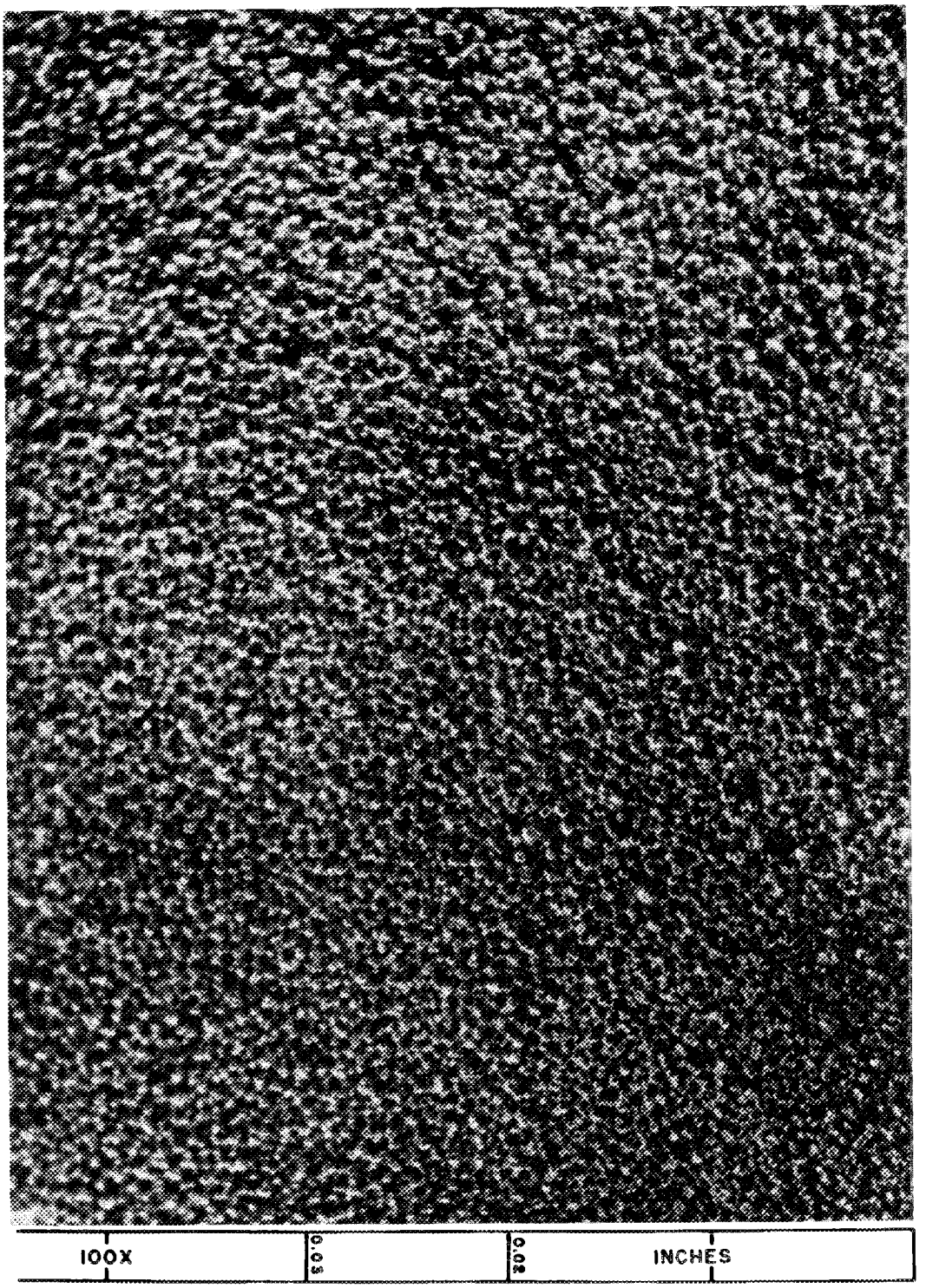

Fig. 14. Photomicrograph of Electrodeposited Technetium-99, Reduced 8%.

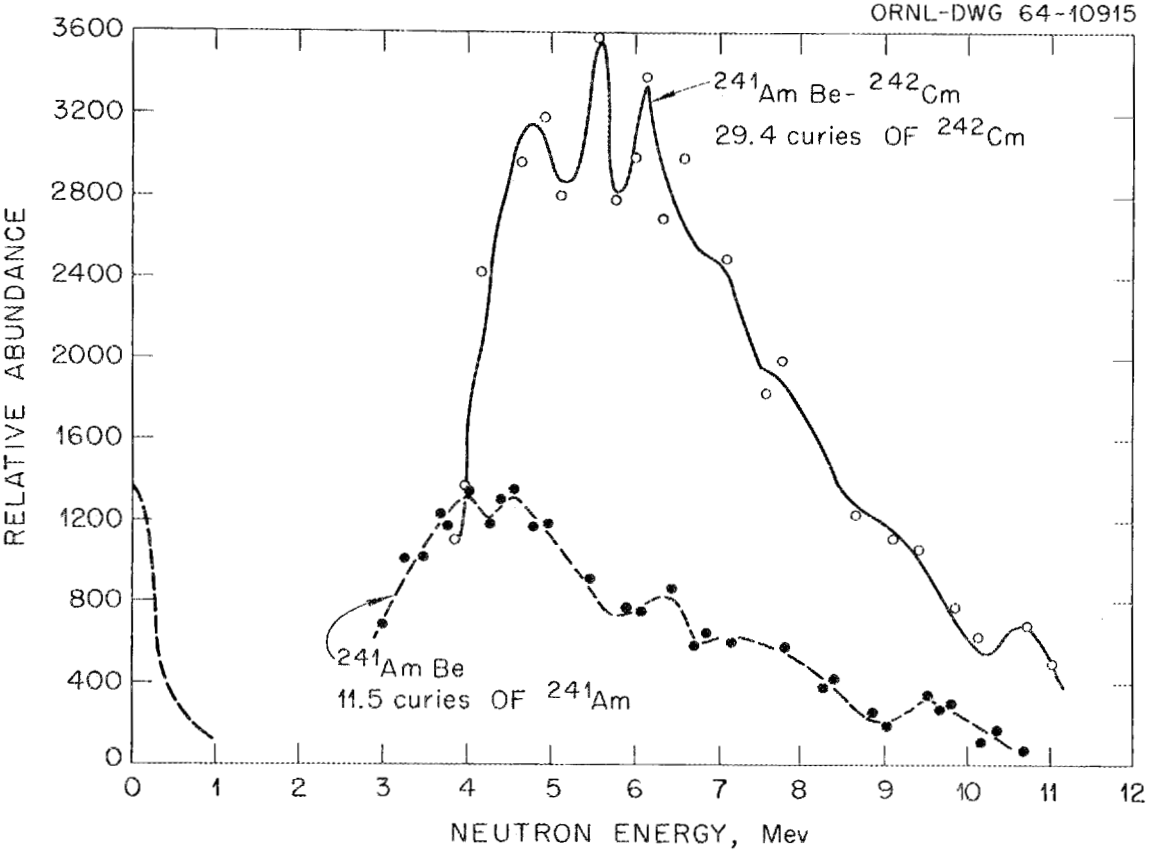

Fig. 15. Neutron Spectrum from ^{241}Am-Be-^{242}Cm and ^{241}Am-Be Sources.

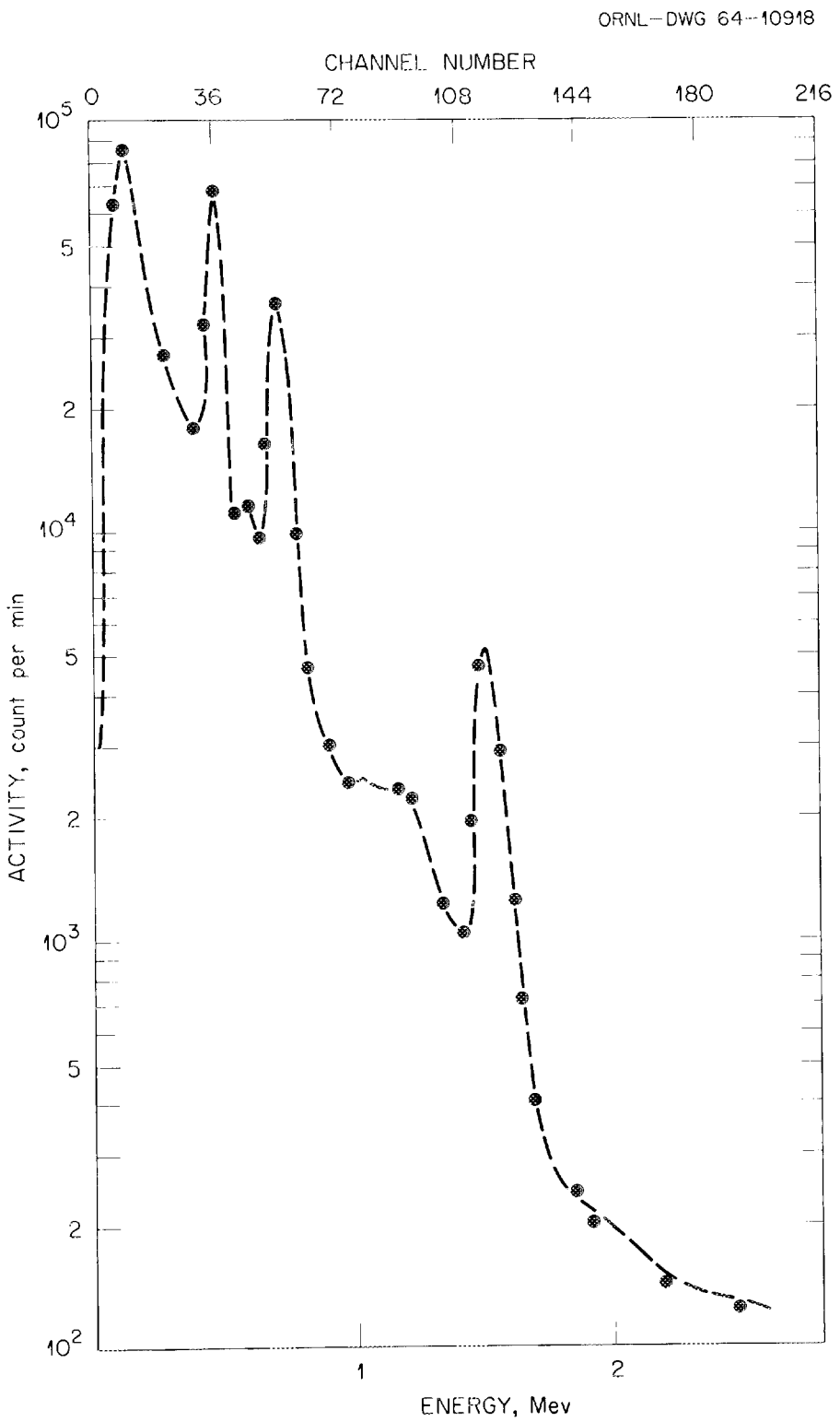

Fig. 16. Gamma Spectrum from a 5.13-curie ^{241}Am-Be-^{242}Cm Neutron Source.

of ~1/16-in.-dia size. Iridium-192 specific activities as high as 800 curies/g have been obtained by this method. Burnup of ^{192}Ir is of greater significance than decay in long term irradiation to produce high specific activity sources, thus the yields achieved by irradiation of ordinary pellets in high neutron flux reactors are limited. By reducing the mass of iridium to thin sections, one can obtain high utilization of the ^{192}Ir produced at or near the surface of the target. Since the radiographic spherical source is formed after irradiation, a homogeneous concentration of ^{192}Ir in the form of a small point source is obtained (Fig. 17).

Ceramic Sources

A technique was developed, tested, and placed in operation for the preparation of fired ceramic forms containing various radioisotopes. Fabrication of the ceramic into any geometric design is carried out before addition of radioisotopes. Cold fabrication reduces cost and permits a great deal of latitude in the choice of fabrication methods. Adsorption of the quantity of the element that will exchange with the ceramic can be controlled precisely. Based on the weight of the ceramic and the known adsorption capacity, the activity concentration can be varied to provide the desired amount of radioisotope. The ceramic is then fired at its fusion temperature to form a stable glass structure. As a result of firing, the ion-exchange properties are exchanged for increased chemical stability.

Promethium-147

A 70-curie ^{147}Pm source, to be used in determining the damage to solar cells by ~80-kev electrons, was prepared for space-radiation simulations. The energy spectrum obtained is shown in Fig. 18. The source was fabricated from potassium silicate and promethium oxide (which was distributed over ~25 cm^2) reacting at ~1100°C to form a glass. A total potassium silicate and promethium oxide weight equivalent to ~25 mg/cm^2 was obtained. A 5 mg/cm^2 Pyrex overlay was fused to the surface to reduce smear contamination to a low level. The electron flux above 25 kev was ~4.6×10^{10} electrons/cm^2·sec at a distance of 2 cm.

A low energy ^{147}Pm beta source was prepared by sorbing ^{147}Pm onto the surface of a prepressed pellet of aluminum silicate. The pellet was then fired to incorporate the promethium into the silicate as a glass. This pellet was placed in an aluminum holder with an open window 3/16 in. in diameter to measure the density of shock waves in air.

A 10-mc ^{147}Pm beta source producing K x rays from tin was prepared as a "sandwich" source between 2-mil tin foil. After encapsulation in aluminum with a 10-mil window, the K x-ray intensity was 2.08×10^8 photons/min at 1 cm. The characteristic x-ray peak was 62% of the total bremsstrahlung produced.

Phosphorus-32

About 200 ^{32}P sources (Fig. 19) were prepared to be used in a blood irradiator to study the synergistic effect of zero gravity and radiation on white

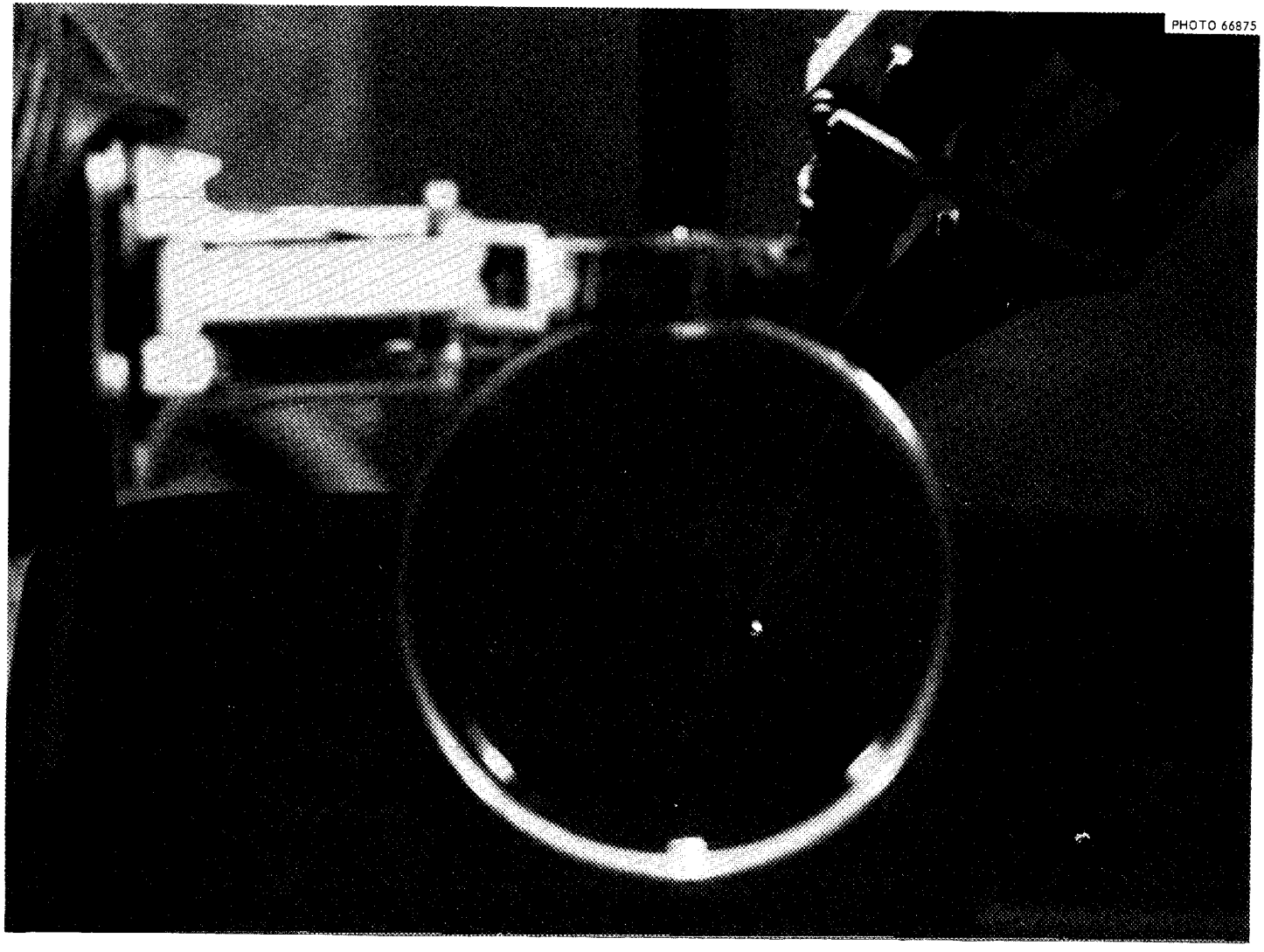

Fig. 17. Iridium-192 Radiographic Source.

ORNL DWG. 65-3190

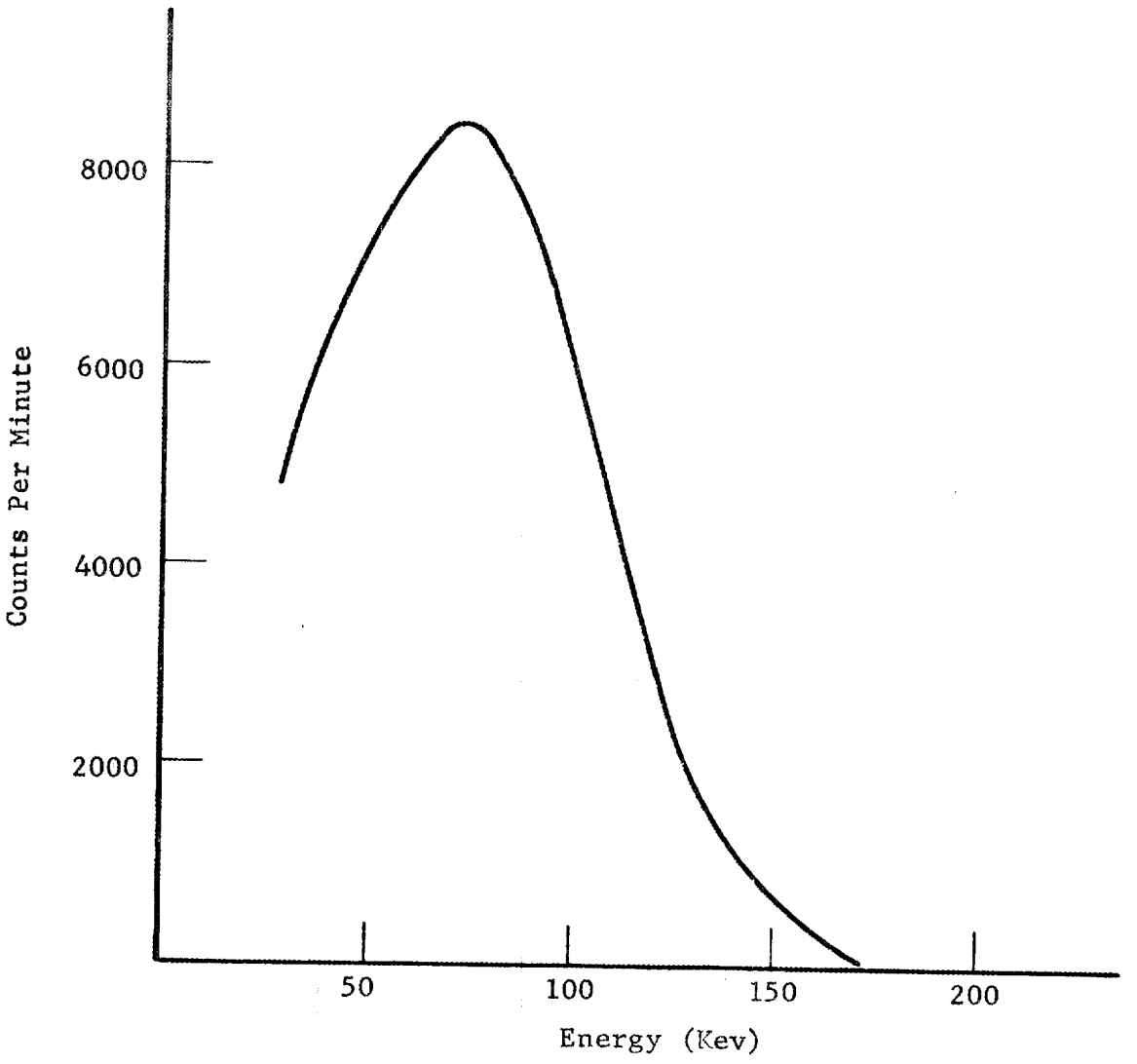

Fig. 18. Energy Spectrum of Promethium-147 Source.

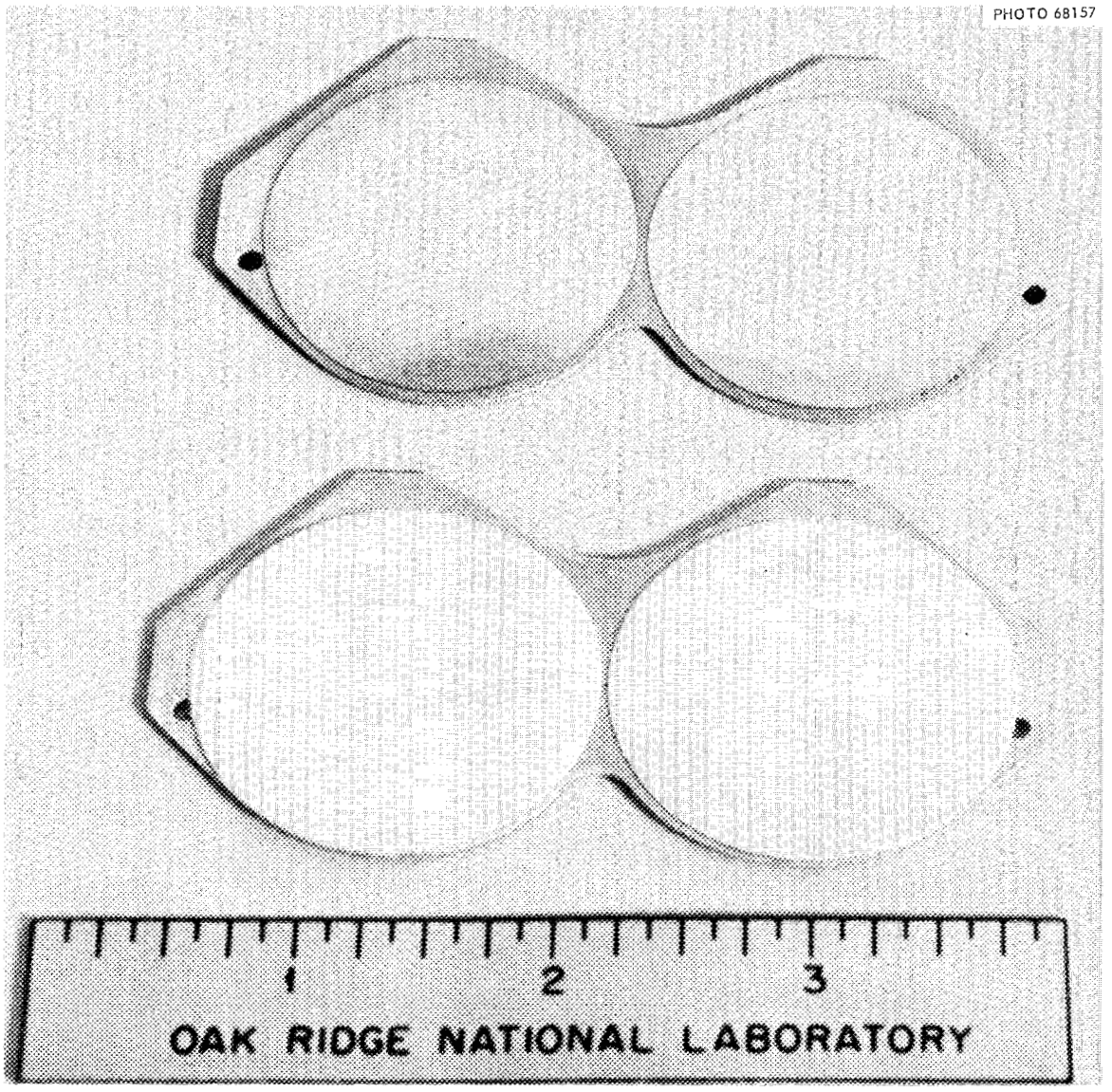

Fig. 19. Phosphorus-32 Sources.

blood cells. Since the experiment was designed for in-capsule space flight, high integrity sources were required. To obtain uniform activity over the surface of the source, ^{32}P was incorporated into film-forming media such as silicates and polyvinyl alcohol. This technique, while providing a high degree of uniformity, vacuum integrity, and good adherence, was not satisfactory under conditions of high moisture. A compromise in the uniformity of ^{32}P distribution over the surface of the source permitted fabrication of high integrity sources by bonding filter paper to platinum plaques and then saturating with ^{32}P solution. After drying, the paper was saturated with dilute catalyzed epoxy which dried to form a stable coating.

Beta Sources

Use of beta sources for direct conversion to electrical power is highly dependent upon sources with high beta emission. Since self-absorption within the active layer limits such sources to thin cross sections, our experimental program has been directed toward ceramic coating, cermets, and alloys which can be fabricated into forms meeting specific requirements. Dependence upon thick capsules for safety under all conceivable conditions is not compatible with high output; at present, therefore, sources must be considered in relation to specific use and equipment. Flame spraying the rare earth oxides produces coatings with many desirable properties. These coatings are being considered for preparing large-area, high-curie electrostatic propulsion sources; however, surface roughness and low electrical conductivity may limit their usefulness.

Strontium-90

Seventeen 1-watt (160-curie) $^{90}SrTiO_3$ sources were prepared. Square $^{90}SrTiO_3$ sintered pellets (1 x 1 x 1/4 in.) were also prepared with good results. Test pellets were made from varying grades of $^{90}SrTiO_3$ to investigate variations in density as related to TiO_2 content.

A small $^{90}SrTiO_3$ beta backscatter source for measuring coating thickness was developed. Approximately 30 millicuries of $^{90}SrTiO_3$ was placed in the end of a source holder 6 in. long and 0.080 in. in diameter. Most of the radiation was emitted laterally through the 0.002-in.-thick end window. A solid state detector at one side and slightly below the source received backscatter beta radiation. A number of these sources were fabricated and placed in operation.

Preliminary work during the last year indicated that beta process irradiator concepts may provide methods of high energy transfer to reaction mixtures. A pebble bed reactor loaded with ^{90}Sr with the beta emitter fixed in a ceramic medium inert to the reagents involved in the conversion process will be tested early in 1965 (Fig. 20.)

A converter powered with a 200-mc ^{90}Sr source, prepared by pressing a 50:50 weight ratio of aluminum powder and $^{90}SrTiO_3$ into a pellet, operated for several days. When formed under a pressure of ~90,000 psi, the pellet had good mechanical strength and electrical conductivity. This source

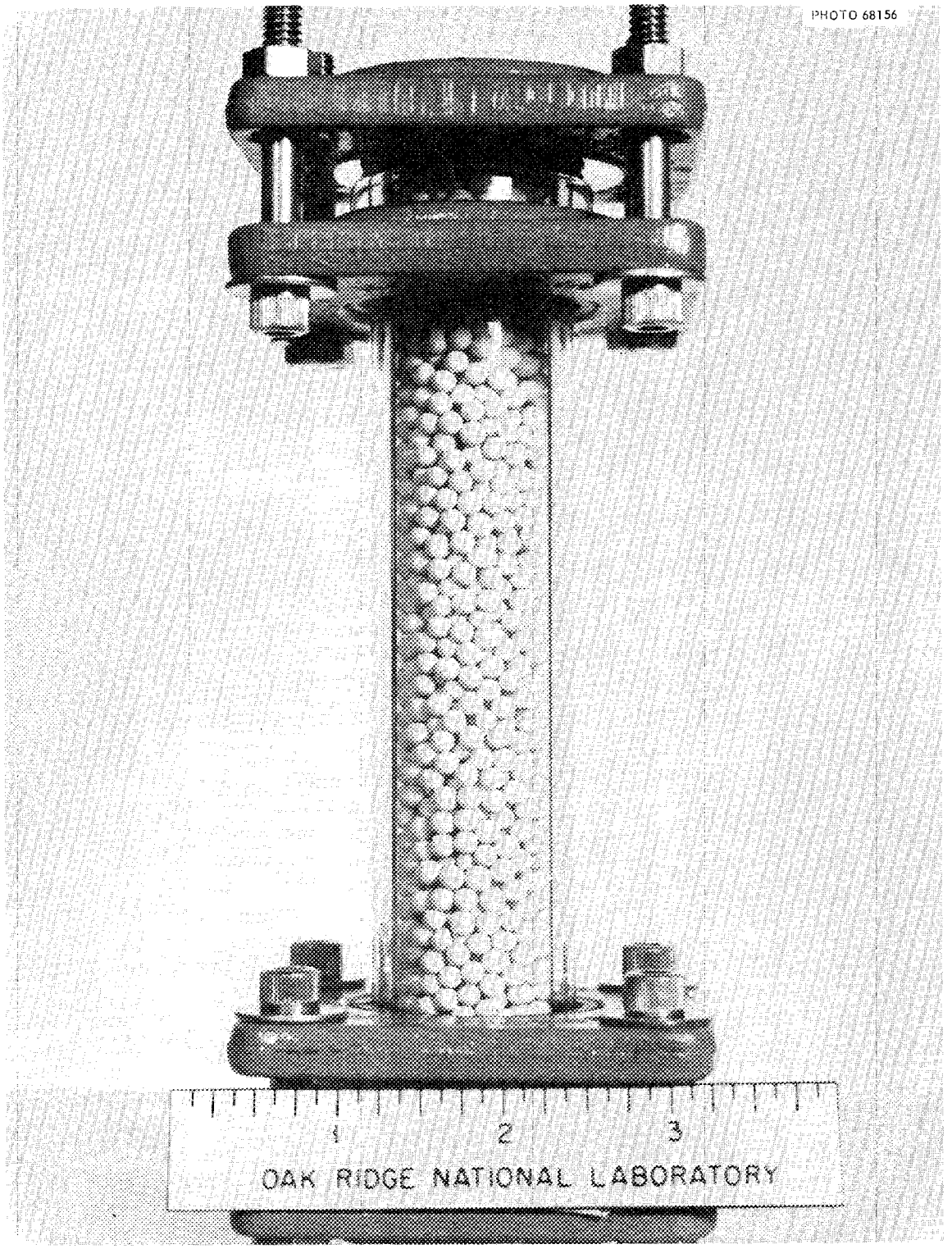

Fig. 20. Strontium-90 Pebble Bed Reactor.

was centered in a spherical cavity, ~1 in. in dia, machined from two blocks of aluminum (Fig. 21). It was held in position with a quartz bushing insulator through which an electrical connector was passed to the outside of the assembly. In a vacuum, currents of 10^{-9} amp at 240 volts were obtained from this device. A second converter, in which a quartz insulator was placed between the source and the collector, produced currents of 10^{-9} amp at 1350 volts at atmospheric pressure. When this device was placed in a vacuum, the voltage dropped due to micro discharges caused by poor geometry of the vacuum system components. Improvements in the vacuum system and converter design to reduce micro discharge are under way. Objectives of this program are to obtain information for designing converter systems that will operate in the 50-100 kv range at low current.

Low Energy X-ray Sources

A number of low energy x-ray sources were prepared for analytical and radiographic applications.

A fission-produced ^{151}Sm oxide source, encapsulated in aluminum with a 15-mil window, produced ~21-kev gamma peak and an output of 9.7×10^{6} photons/min at 1.0 cm.

A 60-mg ^{99}Tc pressed pellet encapsulated in aluminum with a 15-mil window produced, at 1 cm, 2.4×10^{7} photons/min of the characteristic $K\alpha$ x ray. Approximately 0.38% of the total beta disintegrations were converted.

A "sandwich" source of 10 mc of ^{147}Pm oxide between two 1.2-mil molybdenum foils of 7.0-mm dia was encapsulated in an aluminum holder with 10-mil window thickness. An estimated 98% of the beta radiation from the ^{147}Pm was absorbed in the molybdenum foil. The conversion efficiency of the source at 1.0 cm was 0.15%. By using 2.0-mil molybdenum foil without aluminum encapsulation, the efficiency was increased to 0.6%.

After evaporating $^{55}Fe(NO_3)_3$ onto platinum, decomposing the nitrate, and reducing the oxide to metal by heating in a hydrogen atmosphere, iron can be fused into the surface of the platinum to produce a source of very thin sections which is useful in radiography to obtain the structure of thin plant tissue in detail.

Welding Methods

Welding development work using both electron beam and tungsten-inert-gas (TIG) methods has been carried out during the year. The depth of penetration of TIG fusion welds of intermediate temperature alloys (Hastelloy, Inconel, etc.) is limited to ~100 mils unless the weld is designed for the addition of filler metal. The maximum penetration of refractory metal alloys achieved without the addition of filler metal has been ~20-30 mils with this process.

Uranium-232

A total of ~100 mg of ^{232}U was obtained from various ORNL sources. The ^{232}U and the daughter products were converted to chlorides in HCl and then

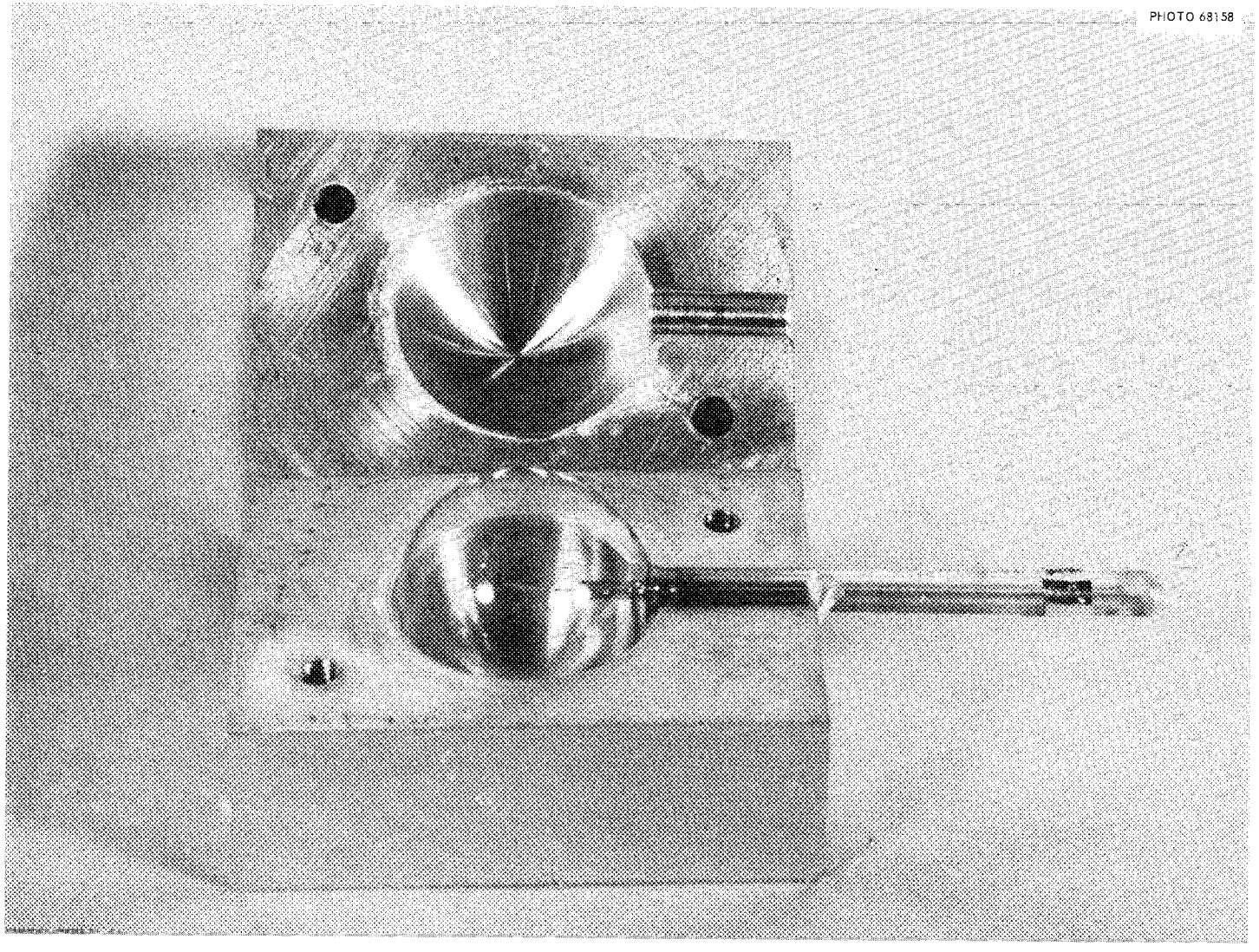

Fig. 21. Strontium-90 Power Conversion Device.

separated by ion exchange. This process provided an adequate decontamination factor (~75) for ^{228}Th and other daughters from ^{232}U with a yield of 99% of the ^{232}U. A 170-mc ^{228}Th evaporated source was prepared from this separation. Two evaporated sources, one 10 mg and one 1 mg, were prepared from another batch of ^{232}U.

Ionization Chamber Calibration

Calibration of ion chambers used for routine assay of irradiated cobalt metal and the validity of results have been investigated. Both sample and measuring geometry were important parameters in obtaining reliable results. To obtain dependable secondary standards, a series of encapsulated sources ranging from 1 to 205 curies (at encapsulation) have been built and standardized.

The ionization chambers are filled with hermetically sealed nitrogen. They contain brass electrodes and a collecting volume of 100 cm^3. By operating with a 150-volt potential across the electrodes and using 1 in. of lead for attenuation, samples ranging from 1 to 205 curies produce ion currents from 6×10^{-11} to 2×10^{-8} amp which are measured with an ORNL Q-826 electrometer. This is an AC-powered vacuum-tube instrument compensated for tube drift by placing carefully matched tubes in opposite legs of the bridge with a 0.2-volt sensitivity for full scale deflection.

A set of secondary standards for ion-chamber calibration was prepared from irradiated cobalt with varying specific activities. A sensitive ion chamber with potentiometric readout was used and was calibrated with NBS ^{60}Co standard solutions and checked with standards calibrated by 4π coincidence counting.

The pieces used for standards were of three different types selected from ion-chamber measurements made in duplicate. Six pairs were 20 mm by 1 mm discs, two pairs were 10 mm by 2 mm discs, and one pair was 10 mm by 10 mm right cylinders. The results of ten replicate measurements on one piece indicated that the activity of pairs selected should not differ by more than 3%.

Disintegration rates measured ranged from 5×10^6 to 3.8×10^7 dis/min. The results were converted to curie units using dilution factors and defined disintegration rate for a curie (3.7×10^{10} dis/sec). Duplicate dilutions were made and agreement was excellent. The maximum difference between duplicate analyses was 2%.

Ion-chamber measurements were made in triplicate on each of the nine pairs of secondary standard pieces. Data from the standards were used to draw calibration curves for four electrometer sensitivity ranges. The maximum activity that can be measured on each of the four ranges is 7.8, 23.9, 80.0, and 238 curies, respectively.

The ion-chamber reading for the cylindrical sample was 80% of the value predicted by the calibration curves. Measurements on the 10 mm by 2 mm discs, stacked to simulate a cylindrical source, gave similar results

which showed that source dimensions and measuring geometry must be considered when making ion-chamber assays. The curves produced are valid for discs up to 20 mm in dia and 2 mm thick, measured with a face toward the ion chamber. However, correction factors can be established for other geometries and source dimensions.

Since oxidation of irradiated cobalt metal creates a contamination problem and changes the activity of the source in an unpredictable way, a series of stainless steel encapsulated sources ranging from 1 to 205 curies was built. Compensation for source dimensions and measuring geometry was achieved by using the calibration curves to standardize the encapsulated standards. The activity content is ~20% higher than the assigned value. As the source decays, the relationship between true and assigned curie content remains constant, and conventional decay corrections can be used.

To check the accuracy of ion-chamber assays for ^{60}Co, a large teletherapy source was built from twenty 20-mm-dia by 1-mm-thick discs. Ion-chamber assay indicated that the activity ranged from 60 to 107 curies per piece. The total for the twenty pieces was 1672 curies. A calorimetry method developed by Posey, Butler, and Baker[30] was used to determine the thermal output of the source (26.5 watts). This corresponds to an activity of 1720 curies after using a conversion factor of 15.36 watts/kilocurie reported by Arnold.[31] The ion-chamber value is lower than the calorimetric value by less than 3%. The good agreement indicates that accurate ^{60}Co assays can be made from ion-chamber measurements.

Fission Products Development Laboratory Operations

During 1964, 1,200,000 curies of $^{90}SrCO_3$ was received. The unloading procedure was modified to remove >99.9% ^{90}Sr in low acidity solution ready for direct processing. Under the earlier procedure, up to 8% ^{90}Sr had been recovered in highly acid solution which required considerably more processing. Total $^{90}SrTiO_3$ produced during 1964 was 1,175,000 curies. In a one-month period of continuous ^{90}Sr processing, 460,000 curies of $^{90}SrTiO_3$ was prepared.

The first large-scale shipment (500,000 curies) of ^{144}Ce in the form of mixed rare earth sulfates was made from the HAPO to the FPDL in May. The unloading procedure was identical to that of $^{90}SrCO_3$ except for the concentration of the acid used to rinse the cask. No problems were encountered and the only significant difference between ^{90}Sr and ^{144}Ce handling was the lack of any detectable pressure buildup in the ^{144}Ce cask. Casks loaded with similar curie amounts of $^{90}SrCO_3$ usually increase in pressure at a rate of ~0.5 psi/day.

A shipment of fission product rare earth solution containing 49,000 curies of ^{147}Pm was received. The ^{147}Pm solution was accidently discharged in a cell during transfer and drained into a waste holdup tank. Temporary equipment was set up to recover this material which was processed by oxalate precipitation of the rare earths. Total

amount of ^{147}Pm that was eventually recovered in suitable form for ion-exchange separation was 36,400 curies. About 18,000 curies was purified by ion exchange and eluted with DTPA.

A total of 372,000 curies of ^{137}Cs was received in eight Shielded Transfer Tank shipments. Mainstream ^{137}Cs crystallizations were conducted with only two shutdown periods — one for a failed agitator and the other for major revisions in the crystallizer system.

Cesium-137 is retained when 3,000 gallons of Hanford fission-product waste (9 M $NaNO_3$) is passed through a shielded transfer cask containing 400 gallons of Decalso.[32] Four transfer casks, each containing 0.05 megacurie of ^{137}Cs, are then transferred to ORNL by rail. The ^{137}Cs is eluted from each cask with 1200 gallons of 5 M NH_4NO_3 and becomes feed material for cesium processing at the FPDL. The major advantage of this method is that cesium can be concentrated in a solid form for transporting from Hanford to ORNL.

A single order for 215,000 curies of encapsulated ^{137}Cs sources in the form of rectangular plates was completed for shipment to Brookhaven National Laboratory. Fifty-seven sources were prepared — 48 from $^{137}CsCl$ and nine from ^{137}Cs glass. The sources, each doubly encapsulated in welded stainless steel containers, will be used in a gamma irradiator. Seven other ^{137}Cs gamma irradiation sources were fabricated totaling 20,000 curies. Other ^{137}Cs shipments included 12 teletherapy units (21,000 curies), 109 miscellaneous sealed sources (600 curies), 19 unencapsulated pellets (2100 curies), and 23 shipments of bulk $^{137}CsCl$ powder (35,000 curies).

One hundred ninety-nine ^{137}Cs sources, ranging in size from 20 to 500 mc, were prepared by sorbing ^{137}Cs on an inorganic ion exchanger (Decalso), melting the Decalso into platinum cups, and then melting inert Decalso on top of the active material.

General Maintenance

Routine building maintenance was carried out at a level comparable to previous years. The servicing of manipulator arms and booting continued to be the largest single item needing maintenance. No major structural changes were made. Panels were installed to partially enclose the area over the cell block and the crane bay was painted to improve visibility. Locker room facilities were expanded to accommodate more personnel.

No full plant shutdown was necessary during 1964. During the crystallizer system shutdown, an extra tank was modified to serve as a third crystallizer in the primary crystallization stream, providing an extra scavenging step on the ^{137}Cs waste stream. During one shutdown, the process control cell was opened for the first time since 1959 to be repaired.

An additional Shielded Transfer Tank was made available for ^{137}Cs shipments and provided significant advantages in scheduling. All of the tanks were coated with a new type of protective paint which decreased decontamination time.

Waste Disposal

Solid waste disposal continued to present a major problem. More than 2000 gallons of contaminated solid waste was removed from the manipulator cells during the year. A method of sealing individual waste packages in heavy plastic before removing was developed. This additional containment of radioactive material lessens contamination on the waste carrier and provides better protection during the burial operation.

Much of the equipment from the $^{137}CsCl$ preparation cell was removed remotely. Entry to this cell for final decontamination and installation of new equipment is scheduled for the first quarter of 1965. While this cell is out of service, final processing of $^{137}CsCl$ is being done in temporary equipment in other cells.

Both process control manipulator cells were decontaminated and repaired. One was repaired in conjunction with the revisions to the ^{137}Cs system. In the other, the inner pane of the shielding window cracked during a routine hot oil flush, so the cell had to be entered to repair the window. This cell had been operating for over four years since its last decontamination.

RADIOISOTOPE SAFETY

Source Testing

Source Classification System

The major accomplishments during 1964 have been the completion of the Source Classification System and a draft of the test procedures for determining the class of a source. Tables 12 and 13 are a summary of the proposed system. Work has begun on a rating system to determine the class of sources for specific applications.

Table 12. Proposed ORNL source classification system structural characteristics

Class	Internal pressure, psig	Puncture resistance, ft/sec onto 1/8-in. dia pin	Crushing force, lb for 1 hr	Shear strength, lb for 1 hr	Impact force		Vibration	Abrasion
					<2 lb, ft-lb	>2 lb, ft free fall		
I	30	14	200*	40**	20	10	10 cps at 10 g for 12 hr	No requirement
II	100	23	200	100	30	15	10 cps at 10 g for 12 hr	50 μ Al_2O_3 abrasive for 250 rev. of 10 in. grinding lap
III	1,000	31	2,000	1,000	60	30	No requirement	Same as II
IV	3,000	44	20,000	10,000	120	60	No requirement	No requirement
V	10,000	62	50,000	25,000	240	120	4 hr at 5-500 cps resonance and 0.001-0.1 in. double amplitude	No requirement

*For 15 min.
**For 30 min.

Table 13. Proposed ORNL source classification system environmental characteristics

Class	Operating temperature resistance, °F	Operating temperature range, °F	Thermal shock range, °F	Temperature-humidity cycling
A	400 for 1 hr	32 to 212	212 to 32 then to -76	200°F—100% RH to 35°F—5% RH 2 hr cycle time for 48 hr
B	750 for 1 hr	-40 to 212	212 to 0 then to -76	Same as A
C	1700 for 1 hr	-320 to 500	500 to 32 then to -76	No requirement
D	2550 for 1 hr	-76 to 1700	1700 to -76	No requirement
E	4500 for 1 hr	32 to 3000	3000 to -320	No requirement

One hundred twenty sources representing 37 commercial designs were classified according to the proposed Source Classification System to determine if sources designed by different companies for similar applications would give consistent results. These trial classification results, given in Table 14, show that most (70%) multicurie source capsule designs (teletherapy, radiography, and neutron sources) fell in the Class III structural category, while most (85%) gaging type sources, which normally contain <1 curie, were in the less demanding Class II.

Table 14. Trial classification of representative commercial sources

Application	Class	Number of designs
Teletherapy	IIIB	2
Teletherapy	IIIC	1
Gages	IIIB	2
Gages	IIC	4
Gages	IIB	5
Gages	IIA	2
Radiography	IIIC	9
Radiography	IIIB	2
Radiography	IIC	2
Radiography	IIB	3
Neutron	IIIC	3
Neutron	IIA	2

While these results are consistent with the intended use of the capsules, the temperature classes of the sources were less consistent. The same wide variation in temperature resistance of sources intended for similar applications was noted in earlier phases of the Source Safety Testing Program when >300 sources were tested to obtain values for the Source Classification System.

Source Application Rating System

The inspection of old sources, particularly those which had been exposed to accidents during use, was continued to obtain background information for the Source Application Rating System. The results of these investigations follow:

1. A ^{137}CsCl gage source in a stainless steel capsule with a back-soldered threaded closure used to measure density of solid rocket propellant was in a rocket propellant explosion and fire. Examination of the source revealed that the solder had melted, but the screw closure remained intact with no transferable activity on outside surfaces of the source.

2. Eight ~1-curie ^{60}Co sources (with non-threaded soldered closures) which had been in use for ~5 years for gaging the melt level in blast furnaces were sent to ORNL for inspection. The lead housings and solder seal of four had melted from overheating and all four were leaking. The other four showed no evidence of leakage. Photomicrographs of the capsule material did not show any interaction between ^{60}Co metal and stainless steel capsule.

3. Three ^{60}Co gage sources, similar to those described above and used in the same way for ~2-1/2 years, were returned by another steel production company. The solder joint on one looked corroded, and the source leaked when given the vacuum leak test; the other two did not. No transferable activity was found on the outer surfaces of the three sources by use of a standard smear test.

4. Five beta gaging sources, each containing ~50 mc of ^{90}Sr, were received after they had been in a fire. The source capsules were 1/2 in. in dia by 1/4-in.-high stainless steel in aluminum housings with 1.5-mil-thick brazed copper end windows. The seal on each source was a threaded plug brazed with a silver-bearing braze material. The source compound inside the capsules was baked onto ceramic discs. Three of the sources were completely separated from their aluminum housings, one had only a glob of aluminum on one end, and the remaining source was still in the aluminum housing which had partially melted around it. Three of the sources had cracked end windows; however, based on the amount of activity found in the rubble around the sources, it appears that only a fraction escaped during the fire.

5. A multicurie ^{137}CsCl irradiation source was received for inspection after giving a positive smear test. Examination did not reveal leaks

but did reveal transferable activity on the source capsule. An autoradiograph of the seal weld showed several spots of activity, and successive radiographs made after thin layers of the weld area were removed indicated that the weldment was contaminated with small amounts of the source compound which were gradually causing leak indications during the smear tests.

Tritium Evaluation

Tritium losses from titanium and zirconium foils at elevated temperatures were determined. Titanium foils hold ^3H well until the 200-250°C range is reached when ^3H is released at unpredictable rates. Zirconium foils released ^3H at temperatures below 100°C. Results from ^3H activated phosphor indicated that ~0.5% of total ^3H was released at temperatures up to 150°C. Above 200°C the ^3H is given off at unpredictable rates.

Extended Test Programs

Certain sources are examined for aging and radiation exposure effects over an extended period by subjecting them to periodic tests, usually once each year.

Water soak tests were conducted on 2-year-old alpha calibration sources. The amounts of activity removed by these tests were insignificant as in previous tests.

Soak tests on ^3H activated phosphor paints were conducted. The ^3H which leached into the water during the soak tests increased in some cases three times as much as one year ago. Some of the paints began to flake off the aluminum backing material. Radium activated phosphor paint losses increased by a factor of two; however, no flaking was observed. The paint samples in both cases were two years old when tested.

Evaluation of Test Methods

The only criterion determining failure of a classification test is the presence of a leak in the source capsule at the conclusion of the test that was not evident prior to the test. A simple leak test has been developed[33] for use with the classification tests and for potential use as a quality control procedure in source manufacture. The source to be leak tested is immersed in ethylene glycol or isopropyl alcohol and the pressure above the liquid reduced to about 125 mm of Hg absolute; a leak is indicated by a stream of bubbles rising through the liquid. The sensitivity of the test was determined by preparing a number of calibrated leaks, measuring the hole sizes microscopically, and measuring the leak rates. Under the test conditions described above, leaks as small as 3×10^{-6} cm^3/sec can be readily detected and leaks of 1×10^{-7} cm^3/sec have been detected by subjecting the capsule to a higher gas pressure immediately before the test. The relationship between the leak diameter (D), surface tension of the immersion liquid (T), and pressure differential required to initiate bubbling (ΔP) can be expressed by the empirical equation, $\Delta P = 4T/D$. Either isopropyl alcohol or ethylene glycol can be

used as the immersion liquid because of their relatively low vapor pressures and surface tensions.

The vacuum leak test can be used only if there is a free volume in the capsule adequate to support a stream of air bubbles in the event of a leak. The test has been used successfully on samples having free volumes as small as 0.1 cm^3; however, this is considered the minimum volume at which it should be used. In cases where there is not sufficient free space in the capsule for the vacuum leak test, an alternative test can be used. This test involves adding a small amount of an easily soluble cesium or lithium salt (cesium or lithium chloride) to the non-radioactive source compound in the prototype capsule being tested and submerging the source in water in a vessel which is alternately evacuated and vented to atmospheric pressure for a period of 20 minutes. The water is then analyzed by a photometric technique capable of detecting cesium concentrations as low as 0.003 $\mu g/cm^3$. A soluble radioactive tracer ($^{137}CsCl$, $^{24}NaCl$) can be substituted for the non-radioactive tracer and standard counting methods used for analysis.

In addition to the above tests, four other simple leak tests have been investigated.

1. Smear Test

   The source is wiped on all accessible surfaces with an absorbent paper disk 25 mm in dia. No attempt is made to "scrub" the source vigorously, but it is thoroughly wiped, using moderate pressure on the paper. Although both wet and dry smear papers are used, the wet smears consistently pick up a higher count than the dry smears. In all cases in which a wet smear shows a leak, however, the corresponding dry smear also shows a leak.

2. Hot-Water Bubble Test

   The source at room temperature is quickly immersed in water that is just below the boiling point (~90°C). If a stream of bubbles emanates from the source due to the expansion of air in the source, a leak is indicated. The sensitivity of this test is estimated to be $\sim 10^{-4} cm^3/sec$.

3. Air-Pressure Bubble Test

   This test is a variation of Test 2 above. The source is placed in a pressure vessel at 540 cm Hg air pressure for 15 min and then is quickly transferred to the hot water. A leak is present if a stream of bubbles is observed.

4. Weight-Gain Test

   The source is first weighed to an accuracy of at least 0.1 g and is then placed in a water-filled pressure vessel at 1800 cm Hg pressure for 1 hr. The source is removed from the pressure vessel and reweighed. A gain in weight indicates that water has entered the capsule through a leak.

The following conclusions can be made from experience with these leak tests:

1. The smear test does not always give indication of a leaking source capsule.

2. The weight-gain test is unreliable, particularly when there are small leaks or when there may be extraneous material on the capsule that can be dislodged.

3. The air-pressure bubble test is only slightly more effective than the hot-water bubble test. The sensitivity of the air pressure test could probably be improved by increasing the pressure and performing the entire test in a single vessel.

4. The vacuum leak test is the most sensitive and reliable of the tests and detected leaks when all other methods failed. The test is not effective, however, when the free volume inside the source is too small to support a stream of bubbles, e.g., interstitial therapy needles. This test may not be satisfactory in the case of a large leak which permits the air to escape in one or two large bubbles that might not be seen by the observer.

5. The value of a thorough visual examination should not be discounted, for, in many instances, examination of seal areas under moderate magnification (5X-20X) clearly revealed porosity.

An attempt was made to define the hole size in a capsule containing water soluble source compounds that leaked. A hole, 0.0015 in. in diameter, was drilled in a typical $^{137}CsCl$ source capsule containing non-radioactive CsCl equivalent to a 1500-curie source. The capsule was placed in water to soak for 20 days. Water samples, taken at periodic intervals, showed cesium equivalent to 160 ± 50 μc from the total 1500 curies. Additional experiments with other hole sizes and capsule arrangements are planned.

Detection of ^{3}H activated phosphor contamination using the ultraviolet light was evaluated. Material representing as little as 64 dis/min could be easily detected.

Shipping Container Testing

Several tests have been conducted to improve the safety of ORNL radioisotope shipping containers. An ICC Specification 3AA gas cylinder was dropped from a height of 30 ft onto a concrete pad and continued to hold a vacuum of <10 microns of mercury. A package, consisting of an ICC Specification 2R container surrounded with vermiculite for shipping ^{239}Pu, successfully withstood a 1/2-hr fire test but failed the 1-hr test. Fire tests were conducted on various gasket materials to find one suitably resistant for radioisotope shipping containers. A Hastelloy-C asbestos spiral wound gasket proved satisfactory although expensive. Work on the use of intumescent paints appears encouraging.

Cesium-137 Thermoelectric Generator

A 120-thermal-watt thermoelectric generator made of ^{137}Cs borosilicate glass was returned to ORNL. The ^{137}Cs source was sealed in a uranium block insulated with multiple layers of reflective insulation with the thermoelectric converter unit on one end of the block. The block had originally been nickel plated to protect it from oxidation, but disassembly of the unit showed several areas of oxidation where the plating had apparently failed. A new container to protect the uranium block from further oxidation has been fabricated, and the conceptual design for a new insulating container has been completed.

ISOTOPES INFORMATION AND QUARTERLY REVIEW

The responsibilities of the Isotopes Information Center include: (1) producing the quarterly Technical Progress Review, Isotopes and Radiation Technology, (2) establishing a library of accessioned material relevant to Division of Isotopes Development interests (isotope applications and production) suitable for retrieval of documents and information, for publishing the Technical Progress Review, and for providing critical state-of-the-art reviews covering various subjects within the AEC areas of interest, (3) writing short brochures dealing with production and uses of isotopes, and (4) carrying out miscellaneous assignments appropriate to the Isotopes Division and Isotopes Development Center as well as AEC's Division of Isotopes Development functions.

Isotopes and Radiation Technology

During 1964, the first volume of Isotopes and Radiation Technology was completed and three issues of Vol. 2 were published. Fifty reprints of articles were made available to authors. Volume 2, No. 3, was based on a comprehensive coverage of isotope production and applications presented at the 1964 Geneva Conference. It covered ~40 papers, many of them in considerable detail.

Information Center "Library"

During the year, ~1600 documents and several hundred microfiche were accessioned. In addition, several thousand abstracts were placed on 5 by 8 in. cards for future accessioning. The Termatrex (peek-a-boo) system was inaugurated and most of the accessioned material has been indexed by punching addresses on the Termatrex cards. The part that has been indexed is now available for prompt retrieval.

Special Publications

The rough draft of a comprehensive brochure, "Isotopes in Industry," was written for the AEC as part of their "Understanding the Atom" series. It is now being edited in Washington. A number of translations of in-scope foreign articles were made available as part of the ORNL translation series.

Miscellaneous Activities

Miscellaneous activities have been quite varied.

1. Foreign journal articles of interest to DID have been reviewed for the Library of Congress contract. The contract (OID) cards will be discontinued and the Information Center will put out an expanded version of "Radioisotopes in World Industry" according to our agreement with DID.

2. About 20 Division reports were edited and published or submitted for publication. Considerable time was spent on the ORNL Radioisotopes Procedures Manual which was made available in June.

3. Submissions to Intern. J. Appl. Radiation Isotopes (JARI) were reviewed and selected.

4. Six showings of the activation analysis exhibit have been handled during the year.

5. Members of the Information Center participated at the Gatlinburg Isotopes Conference as cochairman, at the secondary radiations conference in Chicago on the program committee, at the National Federation of Science Abstracting and Indexing Services Conference in Washington on the discussion panel, at the American Society of Agricultural Engineers Meeting in New Orleans as guest speaker, and at the DID Sixth Annual Contractors Meeting in Washington on the advisory committee.

6. Numerous letters have been written to provide technical information and assistance in reply to letters of inquiry addressed to the Center.

7. A large number of foreign letters have been translated for the Information Center and for the Sales Department.

8. Approximately a dozen trips were made to lecture to various groups on isotope production and applications and technical writing. (This included participation in the ORINS Traveling Lecture Program.)

SALES REPORT -- CALENDAR YEARS 1963 AND 1964

Account No.	Isotope	1963 Sales proceeds	1963 Fund cost of sales	1963 No. of shipments	1963 Millicuries shipped	1963 Fund cost of production	1964 Sales proceeds	1964 Fund cost of sales	1964 No. of shipments	1964 Millicuries shipped	1964 Fund cost of production
Inventory Items											
3651	Carbon-14	$ 209,662	$ 102,121	179	43,688	$ 96,078	$ 213,913	$ 124,228	155	53,981	$ 106,800
3652	Cesium-137	109,239	74,267	210	87,579,432	509,592	326,846[1]	361,739	195	298,921,101	409,427
3653	Cobalt-60	41,180	59,392	16	49,410,900	57,298	110,113	148,396	18	207,442,017	53,738
3654-104	Promethium-147	25,005	127,633	148	4,585,409	51,677	27,420	59,533	138	4,602,306	38,342
3654-106	Technetium-99	9,320	8,494	35	134,201 mg	126,840	51,927	46,587	45	1,225,223 mg	48,501
3654-107	Krypton-85	79,930	94,459	172	6,983,833	12,576	80,136	78,937	168	5,455,505	13,041
3661-288	Chlorine-36	17,439	13,078	131	54.9	5,026	23,954	18,611	127	77	11,402
3661-327	Nickel-63	19,287	7,054	59	2,224	6,575	10,708	4,821	69	1,599	7,667
3661-365	Thallium-204	15,649	7,177	65	22,837	3,725	2,940	1,251	57	2,852	3,574
3662	Strontium-90	8,492	7,097	111	2,289,075	1,694,480	17,725	30,849	119	8,255,493	1,312,314
3663	Tritium	119,162	82,217	171	101,121,678	91,417	111,165	91,197	182	99,854,689	90,541
Total Inventory Items		$ 654,365	$ 582,989	1,297	134,201 mg 252,039,130.9	$2,655,284	$ 976,847	$ 966,149	1,273	1,225,223 mg 624,589,620	$2,095,347
Major Product Items											
3654-103	Cerium-144	$ 3,218	$ 1,795	80	45,252		$ 3,000	$ 2,272	66	73,258	
3654-117	Xenon-133	8,815	17,049	101	290,890		14,854	24,899	166	711,522	
3655	Iodine-131	47,759	40,362	1,577	225,800		33,606	40,796	1,254	146,907	
3656	Iridium-192	37,449	29,981	96	5,664,650		10,157	10,783	53	1,369,297	
3660	Phosphorus-32	77,674	61,577	2,170	82,141		51,337	52,148	2,001	53,526	
3661-278-9	Calcium-45 (P-1, P-2)	26,235	15,721	302	2,684		22,085	14,373	301	2,391	
3661-281	Calcium-45 (P-3)	18,898	11,312	151	419		18,989	11,462	152	424	
3661-284	Calcium-47	41,267	27,000	238	201		44,641	36,078	220	220	
3661-296	Copper-64	7,918	6,186	111	6,252		4,392	6,227	147	3,383	
3661-302	Gold-198	16,007	10,658	158	260,190		1,627	5,773	108	20,537	
3661-315	Iron-59	57,647	37,043	484	1,931		49,209	30,826	467	1,640	
3661-333	Potassium-42	11,029	7,970	688	4,893		9,680	8,652	567	4,224	
3661-356	Sodium-24	9,768	6,453	467	1,467		8,044	7,310	427	1,177	
3661-358	Strontium-85	40,362	18,778	262	808		40,207	21,992	178	804	
3661-360-2	Sulfur-35	18,747	18,902	493	16,880		17,645	11,195	462	14,008	
3661-379	Chromium-51	19,601	17,925	244	10,923		3,996	5,176	85	2,115	
3661-386	Phosphorus-33 (New)								2		
Total Major Product Items		$ 442,394	$ 328,712	7,622	6,615,381		$ 333,469	$ 289,962	6,656	2,405,433	
Development Fission Products											
3654-105	Europium-155	$ 20		1	2		$ 25		1	1	
3654-108	Iodine-129	4,445		28	2,888 mg		21,877		41	27,133 mg	
Total Development Fission Products		$ 4,465	$ 666	29	2,888 mg 2		$ 21,902	$ 13,331	42	27,133 mg 1	

Account No.	Isotope	1963 Sales proceeds	1963 Fund cost of sales	1963 No. of shipments	1963 Millicuries shipped	1964 Sales proceeds	1964 Fund cost of sales	1964 No. of shipments	1964 Millicuries shipped
	Minor Fission Products								
3654-100	Barium-140	$ 2,753		120	1,242	$ 4,387		101	1,674
3654-101	Strontium-89	6,337		126	1,791	9,006		144	3,312
3654-102	Cerium-141	515		40	174	707		29	168
3654-109	Gross Fission Products	775		27	388	702		24	256
3654-110	Zirconium-Niobium-95	4,033		127	2,400	6,673		133	3,868
3654-111	Niobium-95	2,631		58	264	2,054		40	306
3654-112	Praseodymium-143	100		2	2	900		7	18
3654-113	Neodymium-147	1,583		22	32	2,154		21	134
3654-114	Ruthenium-103	1,757		48	176	1,584		31	141
3654-115	Ruthenium-106	5,591		53	810	4,724		58	580
3654-116	Yttrium-91	2,294		47	922	2,111		36	726
Total Minor Fission Products		$ 28,369	$ 41,947	670	8,201	$ 35,002	$ 58,671	624	11,183
	Miscellaneous Development Items--Processed Materials								
3661-250	Antimony-122	$ 129		9	43	$ 80		3	14
3661-256	Argon-76	554		10	38	270		9	14
3661-260	Arsenic-76	346		22	175	440		26	209
3661-262	Arsenic-77	0		0	0	100		4	8
3661-264	Barium-131	357		9	6	420		7	7
3661-266	Barium-133	7,967		59	82	13,720		68	136
3661-270	Bismuth-210	283		8	28	275		6	25
3661-273	Cadmium-109	1,872		35	24	4,898		35	64
3661-274	Cadmium-115	100		9	29	562		13	164
3661-300	Gallium-72	582		15	164	235		8	56
3661-303	Iodine-125	10,300		123	2,789	15,075		8	15,015
3661-304	Gold-199	1,763		29	362	345		9	64
3661-305	Iodine-130	812		48	688	1,212		41	960
3661-317	Lanthanum-140	798		60	431	2,368		44	693
3661-321	Mercury-197	747		19	1,925	5,535		60	12,491
3661-325	Molybdenum-99	933		58	624	602		52	602
3661-331	Palladium-109	131		9	132	225		15	174
3661-335	Praseodymium-142	153		5	52	115		3	35
3661-337	Rhenium-186	385		24	194	459		18	144
3661-343	Samarium-153	475		18	200	425		13	157
3661-350	Silver-111	175		11	35	225		8	24
3661-367	Thulium-170	2,207		23	5,843	1,500		14	4,905
3661-370	Tungsten-187	195		7	35	50		2	2
3661-381	Selenium-75--Enriched	16,662		53	666	34,887		98	1,398
3661-382	Argon-38	220		1	0.2 cc	0		0	0
Total Miscellaneous Development Items--Processed Materials		$ 48,146	$ 38,804	664	0.2 cc 14,565	$ 84,023	$ 73,050	564	37,361

Account No.	Isotope	1963 Sales proceeds	1963 Fund cost of sales	1963 No. of shipments	1963 Millicuries shipped	1964 Sales proceeds	1964 Fund cost of sales	1964 No. of shipments	1964 Millicuries shipped
	Minor Processed Material Items								
3661-252	Antimony-124	$ 681		39	185	$ 1,411		45	280
3661-254	Antimony-125	1,406		17	14	3,000		23	29
3661-272	Bromine-82	2,434		95	653	2,851		80	713
3661-276	Cadmium-115m	3,003		43	91	2,619		26	103
3661-285	Cesium-134	839		48	677	743		27	312
3661-293	Cobalt-58	3,441		45	172	1,330		14	69
3661-295	Cobalt-60 (HSA Processed)	2,822		124	1,425	3,692		108	1,425
3661-298	Europium-152, -154	2,490		30	249	970		21	87
3661-306	Hafnium-181	545		22	365	611		24	221
3661-308	Indium-114	344		35	72	1,125		34	187
3661-311	Iron-55, -59	599		16	17	385		8	11
3661-314	Iron-55	10,757		69	314	9,274		39	315
3661-323	Mercury-203	10,519		173	10,521	28,521		165	27,371
3661-329	Osmium-191	210		6	21	80		3	8
3661-339	Rubidium-86	1,414		278	4,953	6,511		269	9,349
3661-345	Scandium-46	828		61	3,314	1,183		49	3,394
3661-347	Selenium-75 HSA	426		39	346	844		29	389
3661-348	Silver-110	853		55	655	1,349		55	415
3661-363	Tantalum-182	565		20	139	850		27	138
3661-366	Tin-113	2,519		53	72	2,288		46	64
3661-368	Tungsten-185	382		27	96	710		20	149
3661-372	Yttrium-90	136		10	46	973		12	284
3661-374	Zinc-65	3,147		165	1,407	8,922		151	4,410
Total Minor Processed Materials		$ 50,360	$ 52,981	1,470	25,804	$ 80,242	$ 52,247	1,275	49,723
					134,201 mg 0.2 cc				1,252,356 mg 0 cc
Total Radioisotopes		$1,228,099	$1,046,099	11,752	258,703,083.9	$1,531,485	$1,453,410	10,434	627,093,321

Account No.	Isotope	1963 Sales proceeds	1963 Fund cost of sales	1963 No. of shipments	1963 Millicuries shipped	1964 Sales proceeds	1964 Fund cost of sales	1964 No. of shipments	1964 Millicuries shipped
	Service Irradiations								
3658-190	86-Inch Cyclotron	$ 106,279	$ 136,425	144	142 S.I.	$ 173,381	$ 180,937	168	168 S.I
3658-193	Reactor Service Irradiations	51,140	36,878	514	511 S.I.	51,782	40,857	428	414 S.I.
3658-194	Cobalt-60 Gamma	4,141	1,597	6	6 S.I.	4,724	3,497	7	6 S.I.
3658-196	Naval Research Laboratory--LITR	76,735[2]	71,036			79,011[2]	87,115		
3658-198	Naval Research Laboratory--ORR	3,975[2]	208				72		
3658-199	NASA--ORR	3,001							
3659-200-1	Activation Analyses	5,180	3,410	9	87 Anal.	1,378	487	2	2 Anal.
3659-203	Special Services	70,066	95,895	1		87,391	69,937		
3659-204	Target Preparation--Radioisotopes	2,190	(1,451)			17,400	34,318		
3659-205	Cobalt-60 Source Selection	11,279	14,846			18,949	10,451		
3659-206	Cesium-137 Source Fabrication	40,130	25,179			49,115	38,991		
3659-209	Incoming Transport Charges--Customer Service	11	150			1,237	1,719		
3663-521	Tritium and Deuterium Target Fabrication	16,825	14,128	93		19,520	14,714	120	
Total Radioisotope Services		$ 390,952	$ 398,301	767	659 S.I. 87 Anal.	$ 503,888	$ 483,095	725	588 S.I. 2 Anal.
3631	Packing and Shipping	$ 267,885	$ 213,231			$ 246,054	$ 221,451		
3640	Isotopes Sales Department		$ 208,533				$ 199,098		
TOTAL		$1,886,936	$1,866,164	12,519	137,089 mg 0.2 cc 659 S.I. 87 Anal. 258,703,083.9	$2,281,427	$2,357,054	11,159	1,252,356 mg 0 cc 588 S.I. 2 Anal. 627,093,321

[1]Includes fund cost transfer of $225,843 from Brookhaven National Laboratory for 214,994 curies of cesium-137.

[2]Fund costs only are billed.

S.I. = Service Insertions
Anal. = Analyses

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