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TRANSURANIUM QUARTERLY PROGRESS REPORT

FOR PERIOD ENDING FEBRUARY 28, 1963

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CHEMICAL TECHNOLOGY DIVISION

METALS AND CERAMICS DIVISION

ENGINEERING AND MECHANICAL DIVISION

REACTOR CHEMISTRY DIVISION

PLANT ENGINEERING, ORGDP

TRANSURANIUM QUARTERLY PROGRESS REPORT FOR PERIOD ENDING FEBRUARY 28, 1963

Compiled by
W. D. Burch

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OAK RIDGE NATIONAL LABORATORY
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1. ABSTRACT

The development of separation processes for the transuranium elements, process equipment development, HFIR target fabrication development, design of the TRU Facility, design of development facilities, corrosion studies, and analytical research and development studies are reported here.

Process Development. -- The principal effect of alpha radiation in Tramex feed is a loss of acid at the rate of about 0.18 moles per liter per day at activity levels of 10 w/liter, which is the proposed operating level. Since acid in the feed must be held between 0.01 and 0.1 M in order to obtain both a stable feed and sufficient extractability of the transplutonium elements, the loss of acid by radiolysis means that adjusted feeds will be stable for only a few hours unless acid is replenished. Initial feed adjustments were demonstrated on a 1 to 2 liter scale in laboratory glassware by simple distillation of excess water and acid. Hydrogen chloride gas bubbled through 10 M LiCl increased the acidity to equilibrium values of 1.35 M at 60°C and 0.4 M at 120°C. Tests are being made to determine if some mixture of air and HCl will give the desired concentration---0.1 M HCl. Careful investigation of the effect of $AlCl_3$, $LiCl_3$, and HCl concentrations in Tramex feed on americium, europium, and acid distribution coefficients is in progress. Concentrations must be very carefully controlled. For example, the americium distribution coefficient between 0.6 Alamine 336·HCl--diethylbenzene and 10 M LiCl--0.1 M $AlCl_3$--0.02 M HCl is about 4. This distribution coefficient decreased to 1 by either increasing the acid concentration to 0.135 M or by decreasing the $AlCl_3$ plus LiCl salt concentration to 9.5 N.

A correlation has been noted between the dielectric constant of the solvent used to dilute 2-ethylhexylphenylphosphonic acid (2-EH(ØP)A) and distribution coefficients of americium and curium between 1 M 2-EH(ØP)A and 1 M HCl. The californium distribution coefficient decreased from 45 for heptane diluent (with a dielectric constant of 1.9) to 3 for toluene diluent (with a dielectric constant of 2.4). Americium distribution coefficients with these two diluents were 0.37 and 0.028, respectively.

Complete separation of americium and californium was demonstrated in a test in which americium was preferentially stripped into 1.9 M HCl from a column of 2-EH(ØP)A adsorbed on powdered glass.

Since aluminum is more soluble as the acid-deficient aluminum nitrate than as neutral aluminum nitrate, solubility as a function of acid deficiency was determined. At 25°C the solubility of $Al(NO_3)_3$ is 2.5 M. At 0.5 M acid deficiency the aluminum solubility is increased to 3.64 M. A test run of the anion exchange americium-curium recovery process was made with simulated feed adjusted to 2.8 M Al which was 0.5 M acid deficient. There were no difficulties with iron precipitation, and europium recovery (used as a stand-in for americium-curium) was normal.

Analysis of used resin from the Building 4507 americium-curium recovery program indicated residual americium-curium on the resin was less than 0.1% of the material processed. Large amounts of aluminum and considerable amounts of chromium, copper, iron, nickel, palladium, and rhodium were found on the spent resin.

Analysis of feed solution indicates that the eight plutonium-aluminum alloy rods processed in Building 4507 contained between 0.45 and 0.90 μg of californium. By extrapolation, the balance of the TRU rods (which have been irradiated longer) should contain 100 to 150 μg of californium.

Scouting tests indicate that it may be possible to separate americium and curium from most lanthanides by either extracting the lanthanides into quarternary amines or sorbing them on anion exchange resin from dilute carbonate solutions. Group separation of actinides and lanthanides is not achieved since the heavier actinides have larger distribution coefficients than the lighter lanthanides.

Elution from anion exchange resin with 4.4 M $LiNO_3$ gave poor californium-einsteinium-fermium separations compared with the excellent americium-curium separation reported last quarter. This was a result of an unexpected reversal in elution positions of the transcurium elements.

Dense, coarse particles of PuO_2 were prepared by precipitating $Pu(OH)_4$, washing, drying at 150°C, and firing at 1200°C. The glassy solid had a density of 10.99 (96% of theoretical), and most particles were 1 to 3 mm in size.

Chemical Process Equipment Development. -- Tantalum disconnects formed on tubing and burnished by rolling were satisfactorily leak tight

even after several makes and breaks. Tests of equipment handling and maintenance concepts and of manipulators are in progress in the processing cell mockup. Calculations were made and conditions selected for irradiation of 0.1 g and multigram quantities of Am^{241} in the ORR. A three-pulsed-column complex was erected to test complete transuranium element processing flowsheets. Scrub tests with europium tracer using dispersed phase coalescers at intervals throughout the sieve plates gave HETS in the range of 18 to 30 in.

Materials Evaluation Studies. -- Protective coatings were tested for damage by exposure to Co^{60} gamma radiation in air and H_2O at 40 to 50°C. Polyamide cured epoxy systems exhibited superior radiation resistance while immersed in deionized water. Some coatings appeared usable after 4.8×10^9 r in water or 6×10^9 r in air.

Target Fabrication Development. -- Research has continued on development of various methods required for the fabrication of HFIR target rods. A method was devised for achieving a continuous metal matrix containing a uniform dispersion of 6-μ-diam oxide particles. The successful production of large plutonium oxide particles ranging from 50 to 100 μ in size will render the procedure unnecessary for the present.

Welding problems continue to present themselves. Acceptable welds can be made, but the frequency of bad welds would not be tolerable. A programed power supply with precise control is required to achieve reproducible welds.

Several methods are being considered for producing a primary leak seal and for obtaining the required strength in the end closure. During this quarter, the possibility of using a braze joint to accomplish this has been studied with encouraging results.

The mockup area was prepared, and installation of some equipment for the plutonium target production was started.

The work on the design of equipment for the TRU Facility continued on schedule. All the major items in cubicle 3 were designed and approved for construction. However, because of shortage of funds, no additional equipment will be built during this fiscal year. The most important items

not yet being designed in cubicle 2 are the end-closure devices, collapse equipment, and the apparatus for attaching the hex can. This equipment is still under development, and there will be several months delay before the final process is selected. Design work on equipment for cubicle 1 is proceeding on schedule.

Design Studies. -- Design of all equipment for the hot development facility in cell 4 of Building 4507 is complete, and installation is proceeding rapidly. A target date of May 10 for cold startup was established. Cell piping and equipment-rack fabrication is proceeding on schedule. All equipment (tanks, tank tops, filter) to be tantalum plated was fabricated and is ready for plating.

Detailed design of the TRU chemical processing equipment was started in the ORGDP Engineering Department. The initial work was concentrated on methods for getting service lines into the cubicle and tank pits, on the hot disconnect well and piping, and on an in-cell sampler unit. A decision to eliminate all Hastelloy tubing from the process cells was made; it was based on corrosion data that showed the superiority of Zircaloy-2 over Hastelloy C in all environments and on cost data which indicated that an all-Zircaloy-2 system was probably cheaper than a combined Zircaloy-2--Hastelloy C system. Tantalum will still be used for high-temperature service.

The complete set of drawings and specifications for the TRU Processing Plant were reviewed and approved by both ORNL and the AEC-ORO. The package should be submitted for bids about mid-March. Procurement by ORNL of items to be supplied the building contractor is under way. The design of mechanical components for the building to be supplied by ORNL, such as the conveyor, equipment transfer case, and cell cubicle is proceeding on schedule.

Analytical Development. -- During this period, nine methods have been written and incorporated into the TRU section of the ORNL Master Analytical Manual. A new method for Cs^{137} analysis based on the selective extraction into TTA containing the nitro group has been developed. Rapid analysis of spectrophotometric data for multicomponent systems at non-equilibrium conditions is possible using a computer code written this month.

2. INTRODUCTION

Gram quantities of transuranium elements for research uses are to be produced in the High Flux Isotope Reactor (HFIR) and Transuranium Processing Facility (TRU) being built at Oak Ridge National Laboratory. Production of these elements in quantity will simplify research with them and make it possible to enlarge our knowledge of their chemistry, solid state physics, and metallurgy. Target material will be provided for production of still heavier and unknown elements.

Isotopes of curium, berkelium, californium, einsteinium, and fermium will result from irradiation in the HFIR of Pu^{242} and of a mixture of Am^{243} with Cm^{244}. These feed materials are being produced by long-term irradiation of 10-kg batches of Pu^{239} in a Savannah River reactor. The irradiated Pu^{239} will be processed at Savannah River to recover the Pu^{242} as decontaminated PuO_2 and the americium-curium as a solution containing about 3 kg of rare earth fission products. The rare earths will be removed from the americium-curium and the actinide oxides fabricated into HFIR targets in the TRU Facility. Also in the TRU Facility the irradiated HFIR targets will be processed to recover the heavy elements for research uses, and the recovered curium isotopes will be refabricated into HFIR targets.

Progress reports are included on the development of separation processes for the transuranium elements, process equipment development, HFIR target fabrication development, design of the TRU Processing Plant, corrosion studies and analytical research and development. This work was performed by the Chemical Technology, Metals and Ceramics, Engineering and Mechanical, Analytical Chemistry, and Reactor Chemistry Divisions of Oak Ridge National Laboratory, and the General Engineering Department of the Oak Ridge Gaseous Diffusion Plant.

Previous reports in this series are:

1. For period ending February 28, 1962 - ORNL-3290.
2. For period ending August 31, 1962 - ORNL-3375.
3. For period ending November 30, 1962 - ORNL-3408.

3. PROCESS DEVELOPMENT

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3.1 Tramex Process Development

In the Tramex process, separation of actinide and lanthanide elements is achieved by extraction of the actinides into tertiary amines from concentrated, low acid, LiCl solutions. Tramex process development studies during the report period included the effect of alpha radiation on feed stability, methods of feed adjustment, and the effect of acid and salt concentration on distribution coefficients. In a sense these are all interrelated. Acid in the feed must be held between 0.01 and 0.1 M in order to obtain a stable feed and high distribution coefficients for the transplutonium elements. The principal effect of alpha radiation in the feed is a loss of acid at the rate of about 0.18 moles per liter per day at activity levels of 10 w/liter. Feed adjustment is necessary to increase the salt concentration to about 10 M LiCl and reduce the acid to less than 0.1 M HCl. A method of continuously replacing acid lost by radiolysis is needed in order to prevent hydroxide precipitation of metal cations. Without acid replenishment, adjusted feed is stable for only a few hours.

3.1.1 Effect of Alpha Radiation on Tramex Feed

Investigation of acid loss from Tramex feed by radiolysis was continued. Tests were made with synthetic feeds of 10 M LiCl containing from 2.0 to 2.6 w of Cm^{242} alpha activity per liter. Acid-loss G values were approximately 10 to 20 molecules per 100 ev for acid concentrations between 0.4 and 1.0 M, and 2.0 to 2.2 for acid concentrations less than 0.4 M. In order to obtain satisfactory actinide distribution coefficients, the acid concentration must be less than 0.1 M, but a small amount of acid is necessary to prevent precipitation of metal hydroxides. Since a G value of 2 corresponds to an acid loss of 0.18 mole per liter per day for feed containing 10 w/liter, which is the proposed operating level, adjusted feeds will be stable for less than a day unless acid is replenished.

Three synthetic feed solutions of 10 M LiCl containing approximately 1 M HCl and 2.0, 2.4, and 2.6 w/liter of Cm^{242} were prepared. Because of

the limited availability of Cm^{242}, the solution volumes were only 1 to 2 ml. These solutions, kept at room temperature and vented to the atmosphere, were analyzed each day for acidity and alpha activity. The acid concentrations were normalized to constant activity concentration to adjust for small volume changes caused by evaporation and radiolysis.

Results of the experiment at 2.6 w/liter are shown in Fig. 1. Results of the other two tests were similar. The initial rate of acid loss corresponded to large G values of 10 to 20. Later, at acidities below 0.4 N, G values were 2.0 to 2.2. The solutions became acid deficient and metal hydroxides, including curium, precipitated. Preliminary analysis of the radiolytic gas indicated the presence of chloride and an unusually high hydrogen-to-oxygen ratio of about 6 to 1.

These investigations will continue with emphasis upon determination of the acid-loss mechanism and methods to reduce the amount of acid loss. Future work will be done with larger volumes of solution at higher activity levels in order to improve the accuracy of the data.

3.1.2 Tramex Feed-Adjustment Studies

Additional tests on a 1 to 2 liter scale in laboratory glassware were made in order to demonstrate the feasibility of feed adjustment by simply distilling off excess water and acid. Solutions of 2 M HCl containing varying amounts of LiCl and $AlCl_3$ were distilled to salt concentrations of 4 to 10 M LiCl and 0.2 to 0.5 M $AlCl_3$. Figure 2 shows the approximate concentration of the residual HCl as a function of the final total LiCl plus $AlCl_3$ salt concentration. Maximum salt concentrations that can be obtained without incurring solution instabilities were also determined. The following salt concentrations were obtained by this method:

LiCl (M)	$AlCl_3$ (M)
10	0.2
9	0.4
8	0.5

Since Tramex feed will lose acid by radiolysis, it will be necessary to replenish this acid in some manner to prevent precipitation of aluminum

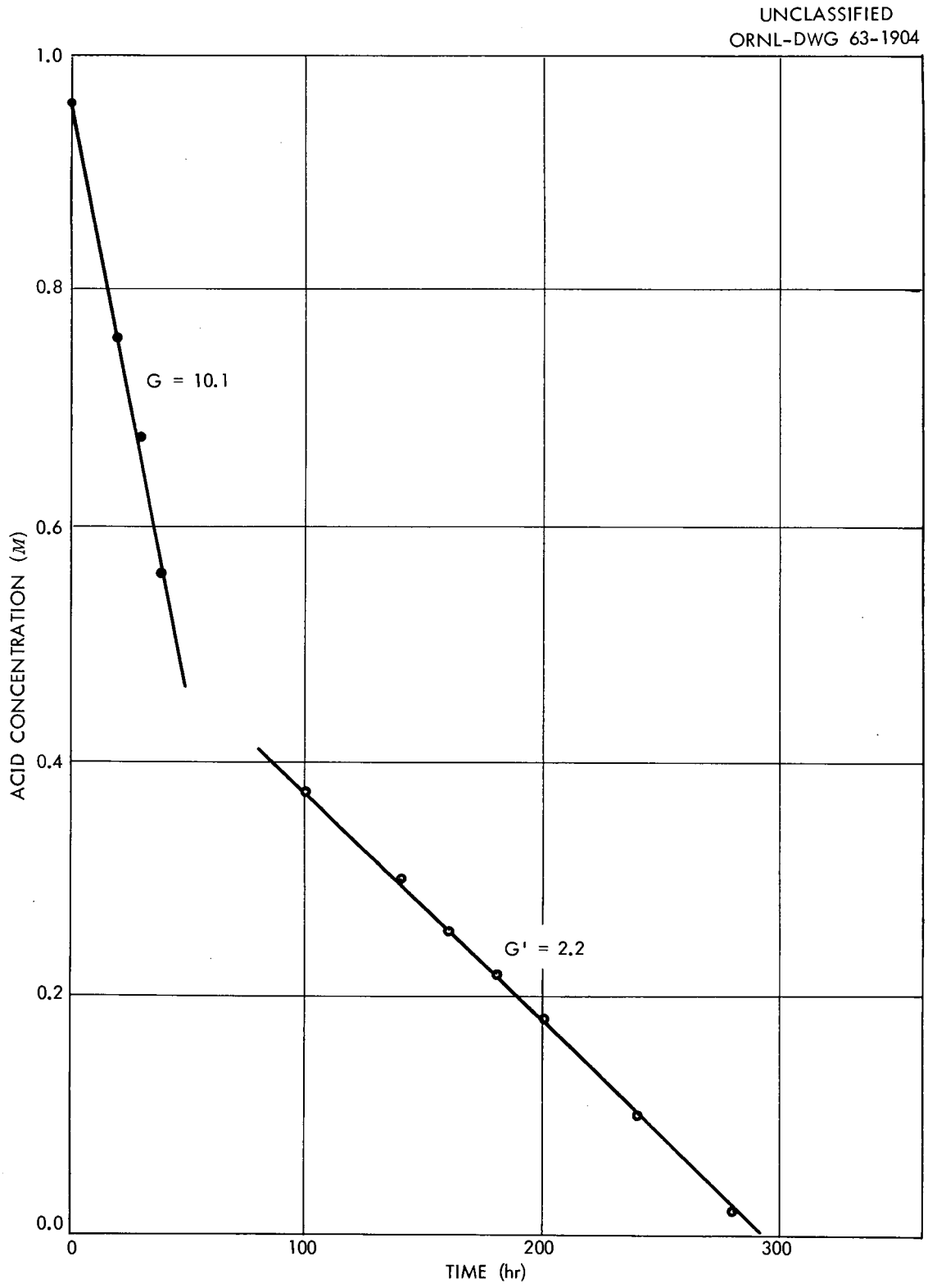

Fig. 1. Acid Depletion by Radiolysis in 10 M $LiCl_3$ Containing 2.6 w/liter Cm^{242}.

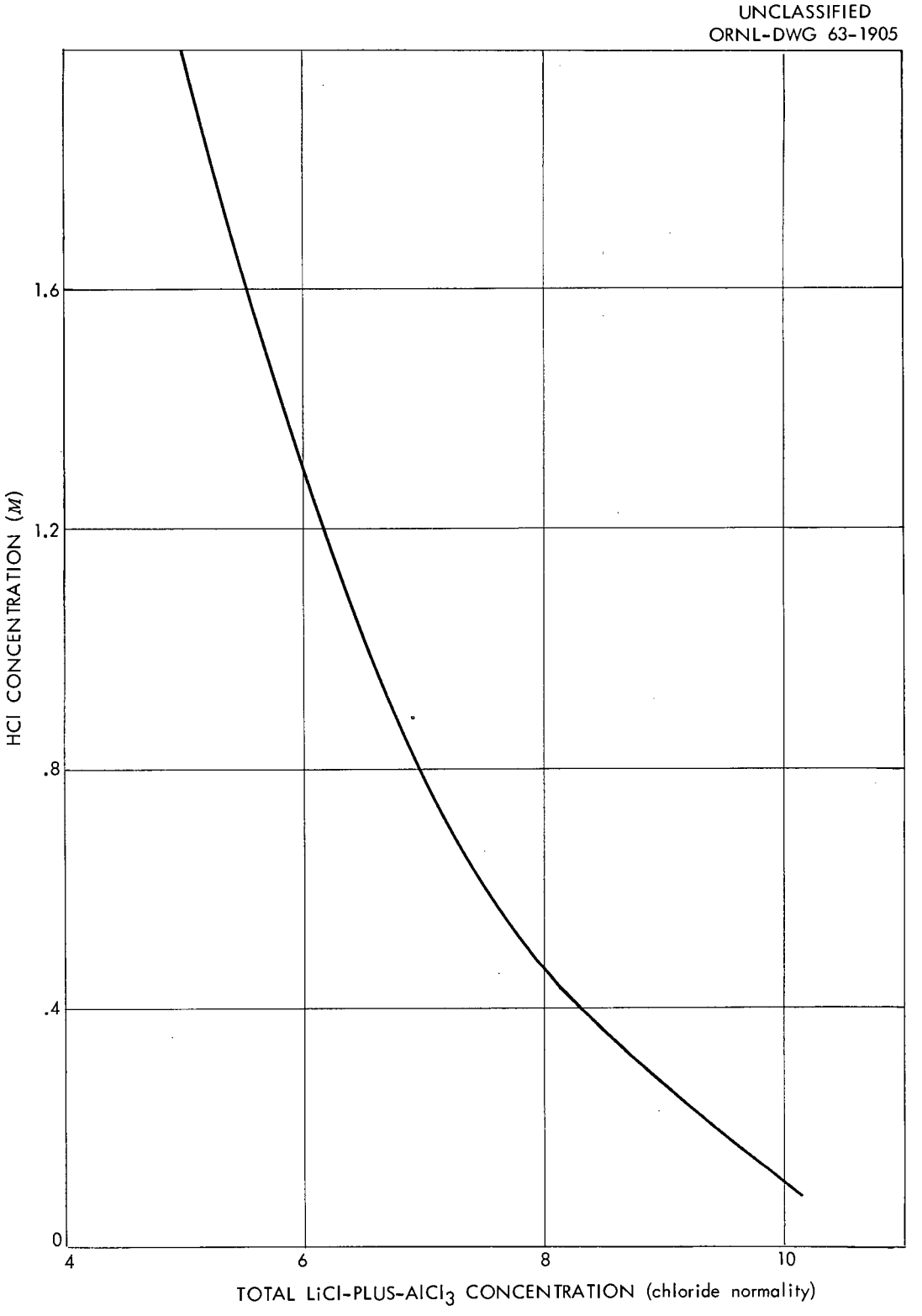

Fig. 2. Residual Free Acid in Mixed $LiCl-AlCl_3$ Solutions at the Boiling Point.

and actinide hydroxides. Bubbling HCl gas through an adjusted feed will introduce HCl without diluting the feed. Unfortunately, equilibrium acid concentrations when pure HCl was used were found to be 1.35 M at 60°C and 0.4 M at 120°C, both too high for satisfactory extraction.

Diluted HCl gas bubbled through the feed will adjust the acidity to the desired value if the HCl partial pressure in the gas is equal to the HCl vapor pressure over the feed. Work to determine the vapor pressure of HCl over 10 M LiCl containing up to 0.1 M HCl has been started. If the vapor pressure is very low, the volume of mixed gas necessary to replace acid lost by radiolysis may be too large for practical application.

3.1.3 Effect of Salt and Acid Concentration in Tramex Feed on Extraction

Difficulties have been noted in the Tramex system with duplicating distribution-coefficient data. The coefficients change rapidly with small variations in acid or salt concentration, and the analysis of feeds with the necessary degree of precision is quite difficult. A careful investigation of these variables is being made to aid in understanding the system and to develop methods for determining when actual Tramex feeds are satisfactorily adjusted. Since it was found that the hydrochloride salt of Alamine 336 extracts HCl from the aqueous phase, distribution coefficients were determined for HCl between the hydrochloride salt of 0.6 M Alamine 336 in diethylbenzene and feeds containing 9, 10, and 11 M LiCl over an acid range of 0.01 to 2.0 M.

With well-characterized feeds, distribution coefficients were also determined for americium and europium over the above range as a function of acid concentration. The amount of acid extracted by the HCl salt of Alamine 336 from feeds containing 9, 10, and 11 M LiCl and from acid solution only is shown in Fig. 3. In Fig. 4 the HCl distribution coefficients are plotted on a logarithmic scale against the acid concentration in the organic phase. Straight-line parallel functions were obtained for 9, 10, and 11 M LiCl below organic acid concentrations of about 0.3 M. Distribution coefficients appear to vary regularly with both acid and salt concentration. Results were similar when americium and europium distribution coefficients were measured and plotted in the same manner (Figs. 5 and 6). Americium distribution coefficients were obtained for

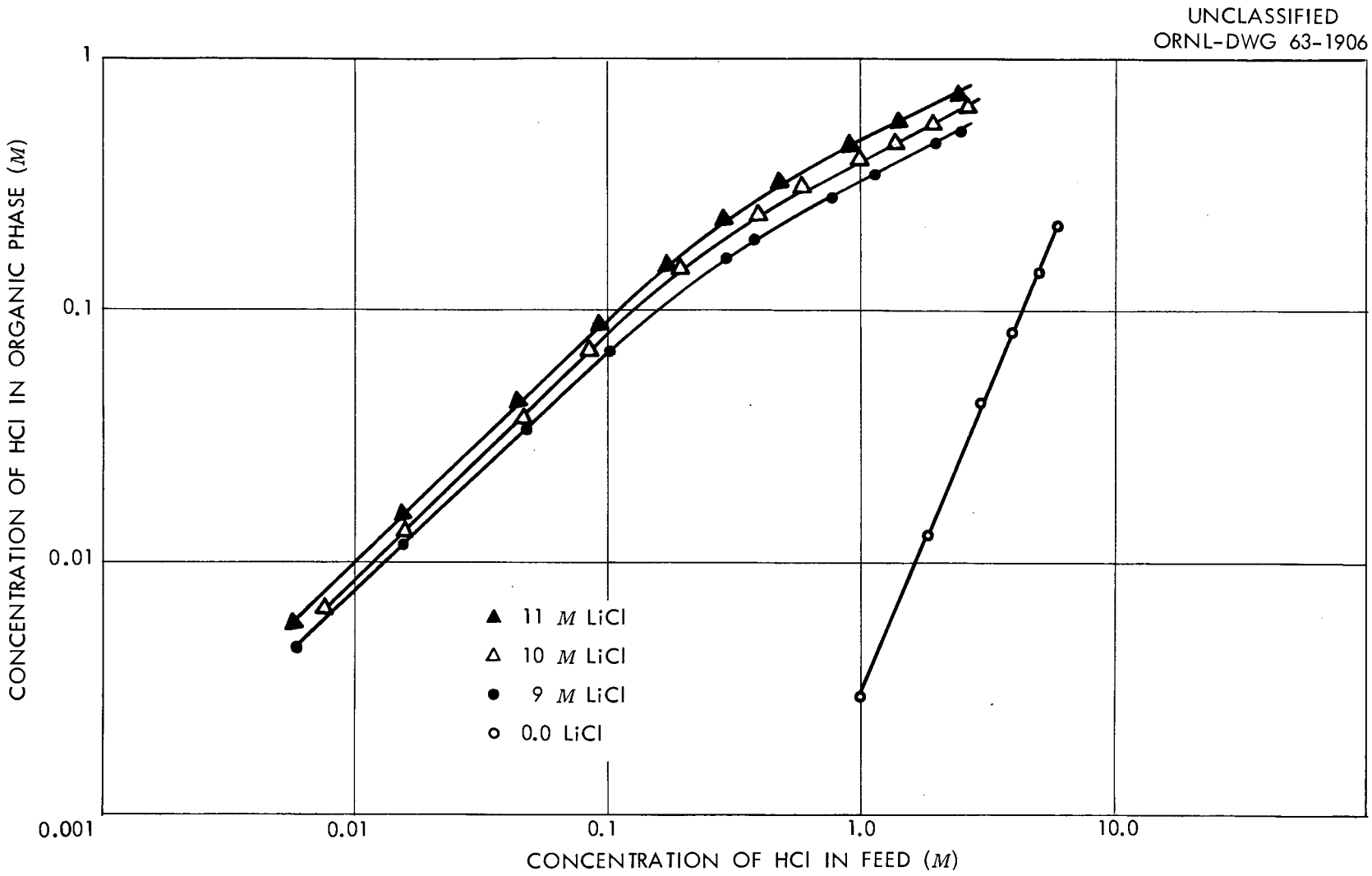

Fig. 3. HCl Extraction by Equal Volumes of the HCl Salt of 0.6 M Alamine 336 from Aqueous Feeds Containing LiCl and HCl.

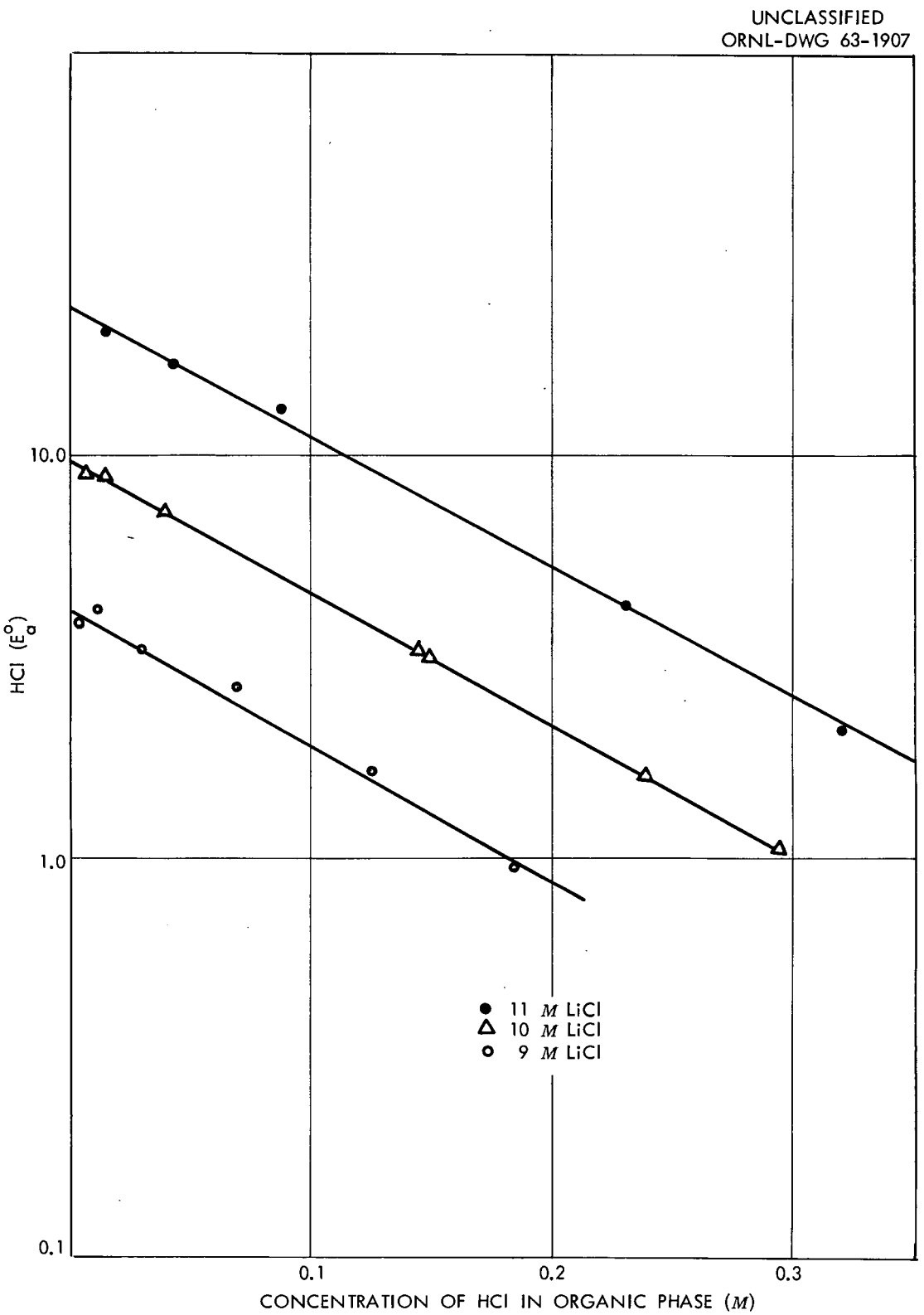

Fig. 4. Distribution Coefficients for HCl between the HCl Salt of 0.6 M Alamine 336 and LiCl Feeds.

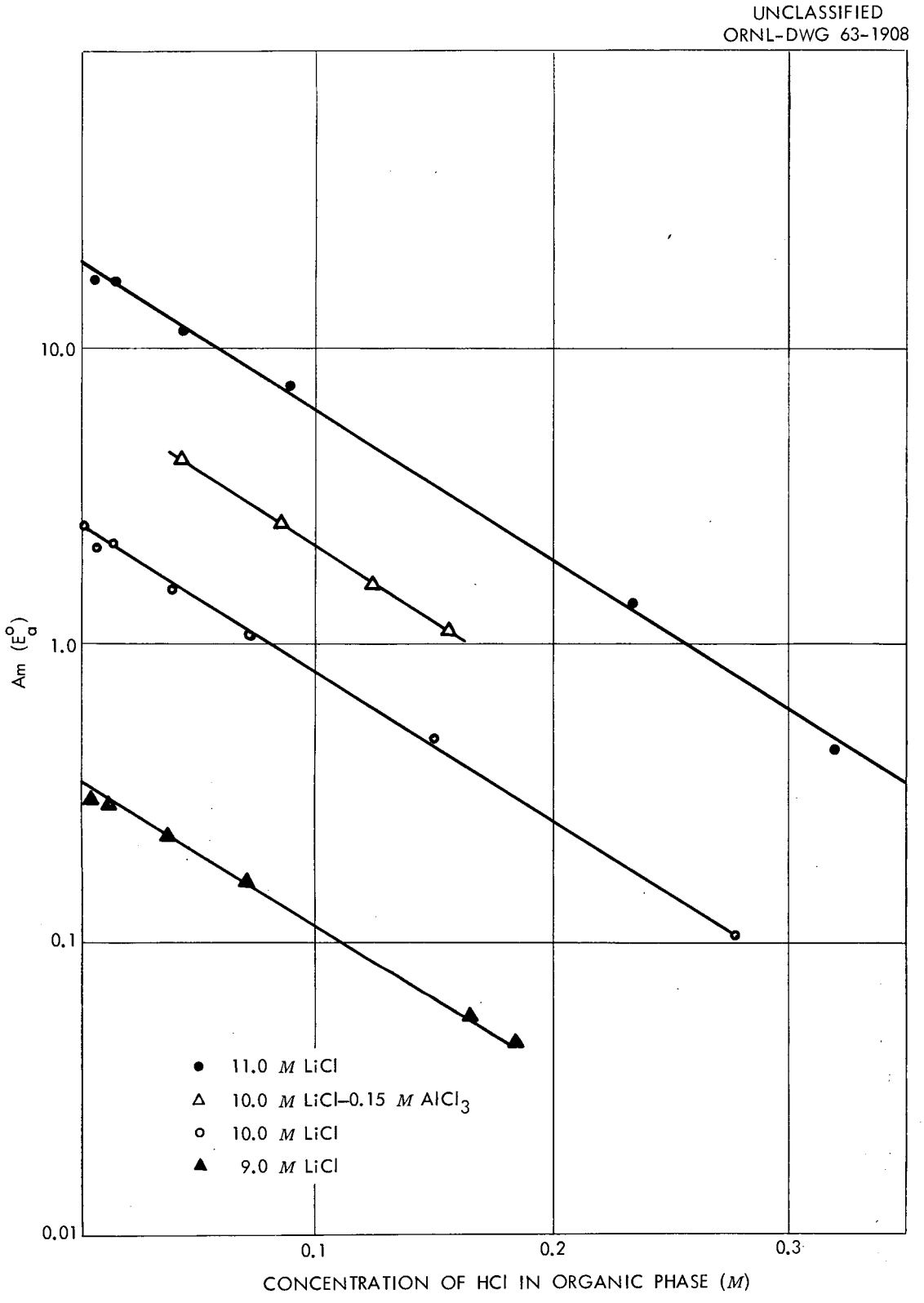

Fig. 5. Distribution Coefficients for Americium between the HCl Salt of 0.6 M Alamine 336 and LiCl Feeds.

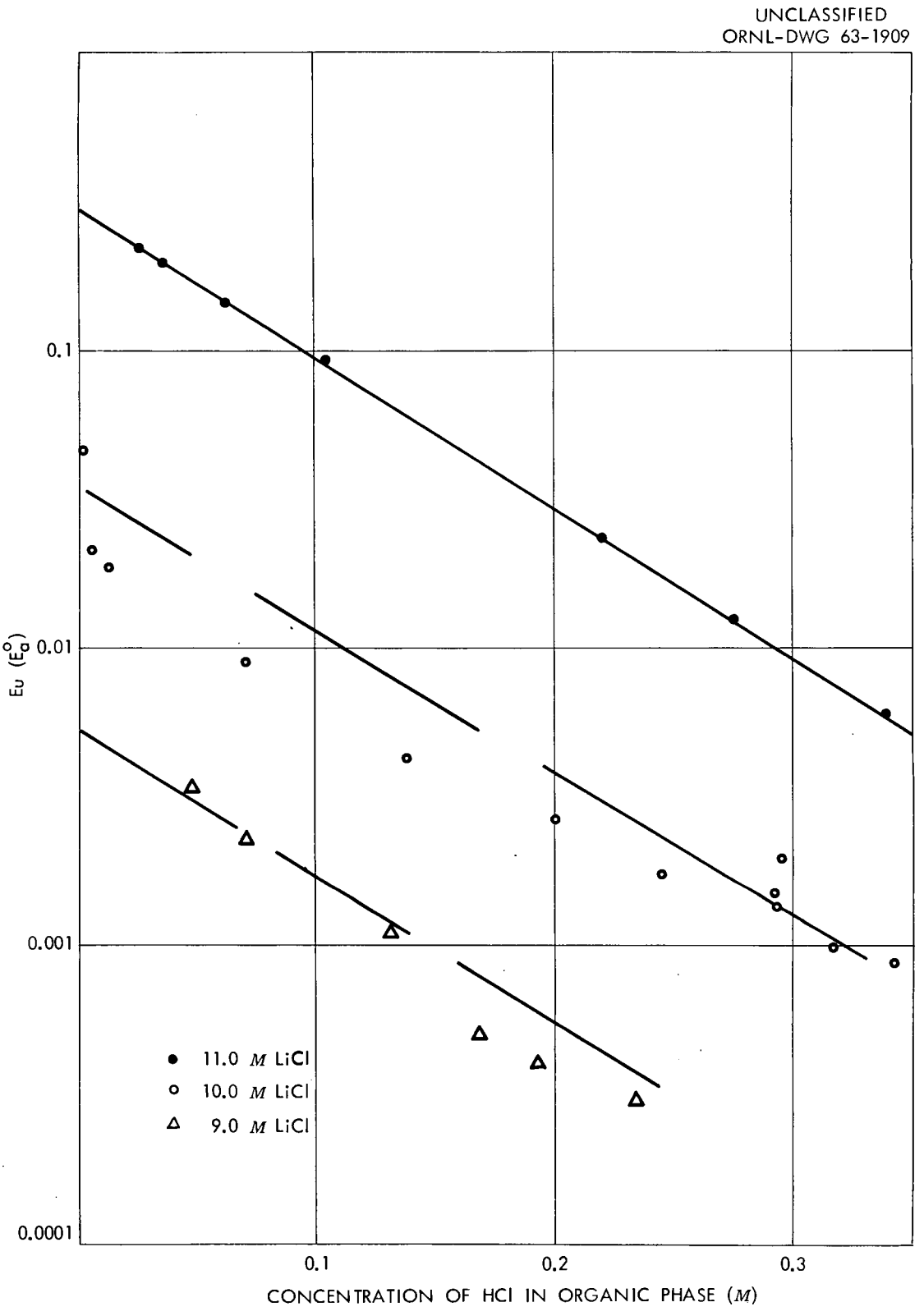

Fig. 6. Distribution Coefficients for Europium between the HCl Salt of 0.6 M Alamine 336 and LiCl Feeds.

10 M LiCl feed containing 0.15 M $AlCl_3$ as well as for 9, 10, and 11 M LiCl feeds. The position of the 10 M LiCl--0.15 M $AlCl_3$ curve (Fig. 5) demonstrates that there is almost no difference between the salting strength of LiCl and $AlCl_3$ when normalities are compared.

In order to determine the precision with which feeds can be duplicated and distribution coefficients can be determined by the methods employed in this study, the series containing 0.15 M $AlCl_3$ was duplicated starting with new feeds. Listed below are americium distribution coefficients obtained and acid analysis of the organic phase for the two sets of data:

HCl Organic Phase		E_A^O for Americium	
Set No. 1	Set No. 2	Set No. 1	Set No. 2
0.042 M	0.043 M	4.27	3.87
0.086 M	0.081 M	2.58	2.44
0.124 M	0.125 M	1.65	1.60
0.156 M	0.156 M	1.15	1.06

In order to obtain this degree of reproducibility, synthetic feed solutions were carefully prepared by volumetric combinations of 1 N HCl solution and saturated solutions of $AlCl_3$ and LiCl. Numerous analyses have demonstrated that saturated LiCl is 13.85 M and saturated $AlCl_3$ is 3.05 M at 25°C. Organic-acid analyses were made by plotting pH-vs-caustic titration curves of the solvent dissolved in acetone. Reproducibility of this method is about ±0.002 M acid. Distribution coefficient data for americium indicates that measurements made by counting are less accurate than those made by organic-acid analyses for a single sample.

Considerable scatter was obtained for europium distribution coefficient data for 9 and 10 M LiCl. This is apparently due to lack of precision when counting at the low activity levels used. Curves indicating probable values are shown. These were drawn by comparison with europium data for 11 M LiCl and with americium data.

The degree of precision necessary for Tramex-feed adjustments is apparent from these data. For example, proposed Tramex feed will contain 10 M LiCl, 0.1 M $AlCl_3$, and 0.02 M HCl, giving an americium distribution

coefficient of about 4. Data given in Fig. 5 indicates that the americium distribution coefficient will be decreased to 1 by either increasing the acid concentration to 0.135 M or by decreasing the salt concentration to 9.5 N.

3.2 Separation of Transplutonium Elements into Two Fractions in the Phosphonate System

A process[1] based upon extraction of transcurium elements from 1.0 M HCl into 1.0 M 2-ethylhexylphenylphosphonic acid [2-EH(ØP)A] in diethylbenzene has been developed for separating transplutonium elements into an americium-curium fraction and a transcurium fraction. During this report period tests were made to show the effect of various diluents on americium and californium extraction. Separation of americium-californium was demonstrated by preferential elution of americium from 2-EH(ØP)A sorbed on a column of powdered glass.

3.2.1 Effect of Diluent on Americium and Californium Extraction

Americium and californium extraction is greatly dependent upon the solvent used to dilute the 2-EH(ØP)A. Distribution coefficients between 1 M HCl and 1 M 2-EH(ØP)A decreased sharply as diluents of greater polarity were used (Fig. 7). The californium distribution coefficient decreased from 45 for heptane diluent (dielectric constant of 1.9) to 3 for toluene diluent (dielectric constant of 2.4). Americium distribution coefficients with these two diluents were 0.37 and 0.028, respectively. In a similar manner, the addition of octyl alcohol[2] (dielectric constant about 10) or tributylphosphate (dielectric constant of 7.9) to the solvent decreases the distribution coefficients.

It is believed that the more polar diluents compete with 2-EH(ØP)A dimer formation by hydrogen bonding and thus decrease the effective

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[1] D. E. Ferguson, Transuranium Quar. Prog. Rep. for Period Ending Feb. 28, 1962, p 5, ORNL-3290 (June 6, 1962).

[2] R. D. Baybarz, Separation of Transplutonium Elements by Phosphonate Extraction, ORNL-3273 (July 20, 1962).

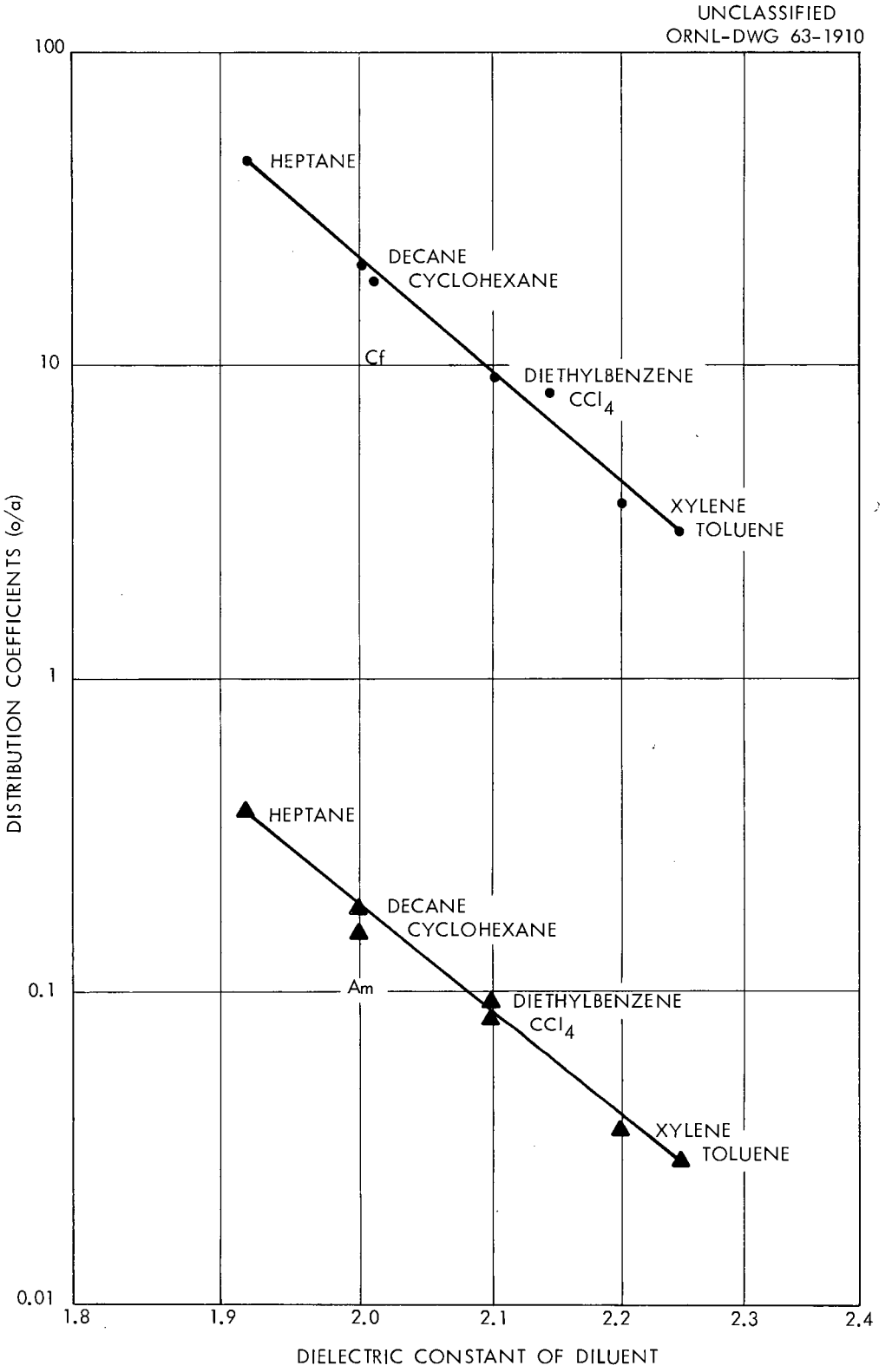

Fig. 7. Americium and Californium Distribution Coefficients between 1 M HCl and 1.0 M 2-ethylhexylphenylphosphonic Acid in Various Diluents.

concentration of the 2-EH(ØP)A dimer. Since the mechanism of actinide extraction involves hydrogen bonding with three dimers,[3] the presence of polar diluents results in less extraction of the actinides.

3.2.2 Californium-Americium Separation by Preferential Stripping from a Column of 2-ethylhexylphenylphosphonic Acid on Powdered Glass

Complete separation of americium and californium was demonstrated in a test that used preferential stripping from 2-EH(ØP)A into 1.9 M HCl. A column of 2-EH(ØP)A adsorbed on powdered Vycor glass was used, and operations were similar to those used for ion exchange resin columns.

Vycor glass powder (100 to 200 mesh) was slurried in 2-EH(ØP)A. The glass was filtered to remove excess solvent and then placed in a column 10 cm high by 0.15 cm^2 in cross section. A small volume of 1.9 M HCl containing californium and americium tracer was passed through the column at 80°C. The column was washed with 1.9 M HCl until all the americium was eluted. The californium was subsequently stripped from the column with 6 M HCl. Excellent separation was obtained, as shown by the elution curves in Fig. 8.

3.3 Laboratory Support of Americium-Curium Recovery from TRU Rods in Building 4507

Eight plutonium-aluminum alloy rods irradiated to produce Pu^{242}, Am^{243}, and Cm^{244} for HFIR targets have been processed by anion exchange in cell 1, Building 4507. Laboratory work in support of this program included studies of the effect of acid deficiency on aluminum nitrate solubility and on the anion exchange process for americium-curium recovery. The resin used in cell 1 processing was analyzed for residual americium-curium and other metal ions.

3.3.1 Solubility of Acid-Deficient Aluminum Nitrate

Aluminum concentrations near solubility limits are required for satisfactory americium-curium recovery from plutonium raffinate. Since increased aluminum solubility can be obtained with acid-deficient aluminum nitrate, solubility as a function of acid deficiency was determined. Solubility data obtained by mixing $Al(NO_3)_3$ and dibasic aluminum nitrate solutions are plotted in Fig. 9.

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[3] D. F. Peppard, G. W. Mason, I. Hucher, J. Inorg. Nucl. Chem. 18, 245 (1961).

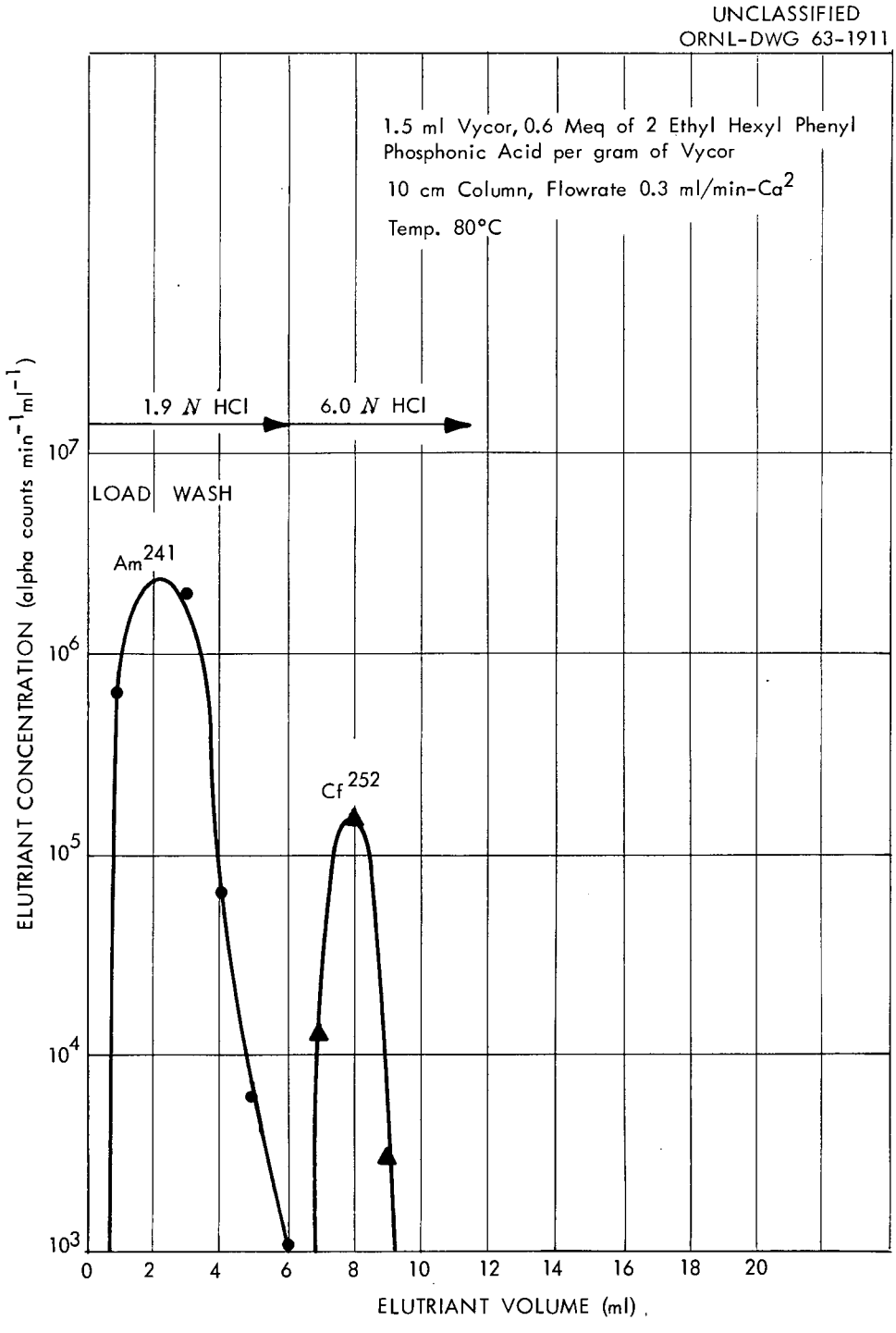

Fig. 8. Americium and Californium Elution Curves from a Column of 2-ethylhexylphenylphosphonic Acid on Powdered Glass.

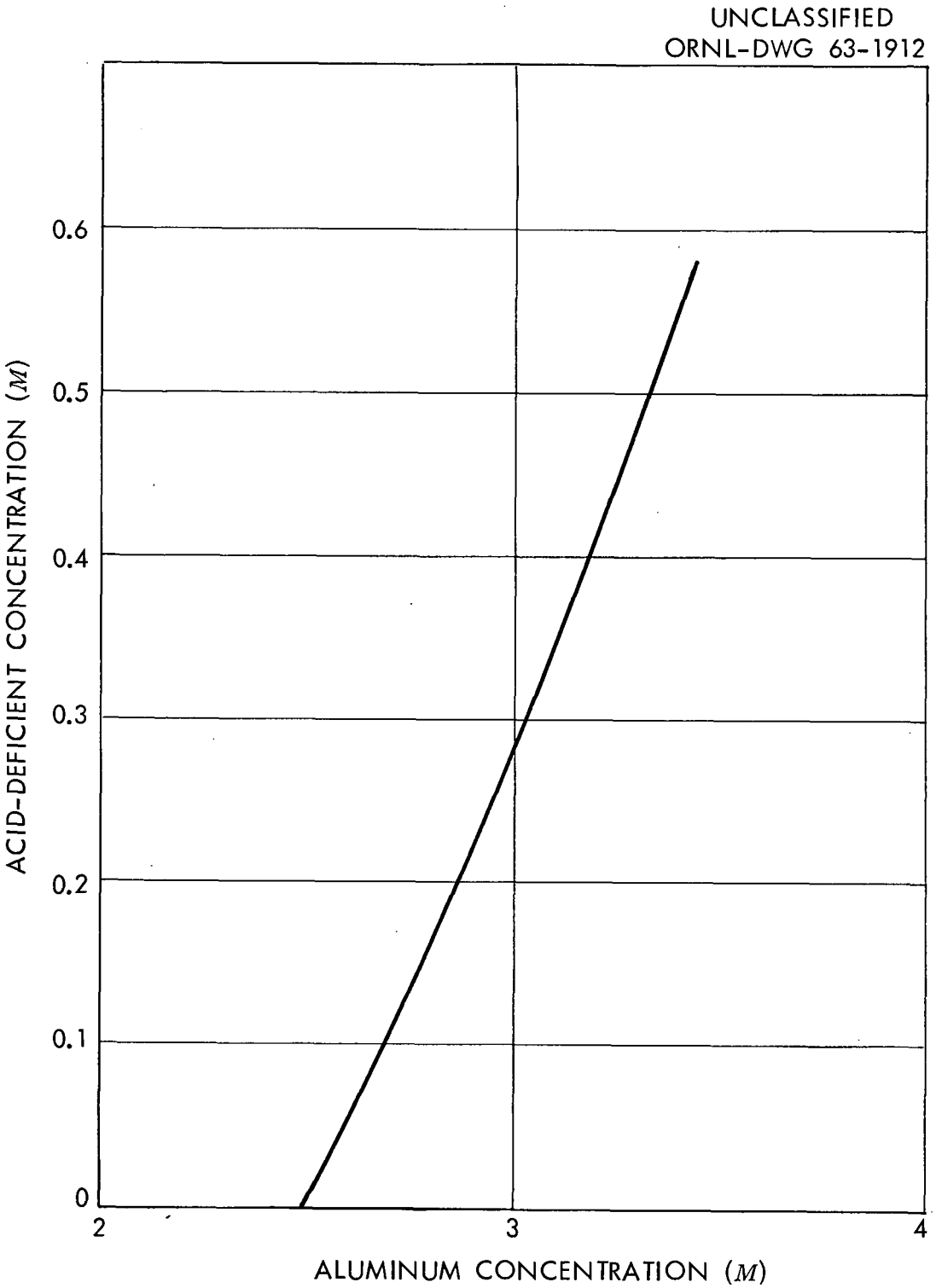

Fig. 9. Solubility of Acid-Deficient $Al(NO_3)_3$ at 25°C.

3.3.2 Effect of Acid-Deficient $Al(NO_3)_3$ on the Americium-Curium Recovery Process

A laboratory test run was made on a column of 50- to 100-mesh Dowex 1-X10 resin at 60°C to investigate iron-loading characteristics from acid-deficient $Al(NO_3)_3$ feed. The feed contained 2.8 M $Al(NO_3)_3$ which was 0.5 M acid deficient, 2 g of iron per liter, 0.5 g of rare earths per liter, and europium tracer.

Iron did not load or precipitate on the column, and only 0.8% was found in the product cut. Europium recovery was 84%, which is about normal for feed of this aluminum concentration.

3.3.3 Analysis of Used Resin from Cell 1 Processing

Resin used in the americium-curium recovery program was examined to determine possible contaminants that might interfere with actinide loading or elution behavior. Two resin samples were obtained: No. 1 was very dark resin from the top of the column, and No. 2 was lighter resin from the lower portion of the column. Each resin sample was treated with successive leaches of water, 6 M HCl, 12 M HCl, and aqua regia. Metal cations removed from the resin were determined by semi-quantitative spectrographic analysis of each leach. The impurities calculated as grams on the entire 8 liters of resin are as follows:

	Resin No. 1	Resin No. 2
Al	48-240	52-250
Ca	<0.1	<0.1
Cr	1.2-2.0	0.6-1.0
Cu	0.8-2.0	0.4-1.0
Fe	0.8-1.6	0.4-1.0
Ni	0.1-1.4	0
Pd	0.16-1.6	0.1-1.0
Rh	0.12-1.2	0.1-0.6

Most of the aluminum, iron, chromium, and nickel was found in the water leach. Palladium and rhodium were found only in the 12 M HCl leaches. No additional impurities were found in the aqua regia leaches. Gamma scans of the leach solutions were obtained, and ruthenium was the only detectable activity.

Part of each original resin sample was dissolved in alkaline permanganate solution and analyzed for americium and curium. Almost complete elution had been obtained during cell-1 operation since the alpha counts on the entire resin column were equivalent to less than 0.1% of the americium and curium processed. Attempts to determine trace impurities in these solutions by spectrographic analysis were unsuccessful because of potassium and manganese interference.

3.4 Californium Content of TRU Rods

Analysis of feed solution indicates that the eight Pu-Al alloy rods processed in cell 1, Building 4507, contained between 0.45 and 0.90 μg of californium. By extrapolation, this indicates that the balance of the TRU rods (which have been irradiated longer) contain 100 to 150 μg of californium.

3.4.1 Ion Exchange Separation of Transplutonium Elements from Cell-1 Feed

Three individual runs were made in order to separate and purify the transplutonium elements from samples taken while processing irradiated plutonium-aluminum alloy in cell 1, Building 4507. The objective was to decontaminate a sample of the transplutonium elements sufficiently so that glove-box procedures could be used to determine the californium content.

The first two samples (runs CF-1 and -2) were available in the form of a 7 M HNO_3--0.5 M $Al(NO_3)_3$ solution containing about 10^9 alpha counts per minute per milliliter (roughly 0.03 w/liter). The material was treated by a two-step anion exchange process with Dowex 1-X10 resin, 100 to 200 mesh. The columns were 25-ml polystyrene burettes, one of them jacketed for a water bath held at 80°C. The volume of the resin bed was 10 ml. The overall flowsheet is given in Fig. 10, and the results for all runs are listed in Table 1. Improvement is noticed in the second run due to better washing of the precipitate, higher acid strength for dissolution of the residue, and better timing on taking the product cut.

The third run, CF-4, utilized feed that was available in 2.6 M $Al(NO_3)_3$ solution, slightly acid deficient. A different flowsheet employing a single ion exchange column, which had been developed previously,[4] was

[4] M. H. Lloyd and R. E. Leuze, "Anion Exchange Separation of Trivalent Actinides and Lanthanides," Nuclear Sci. and Eng. 11, 274-7 (1961).

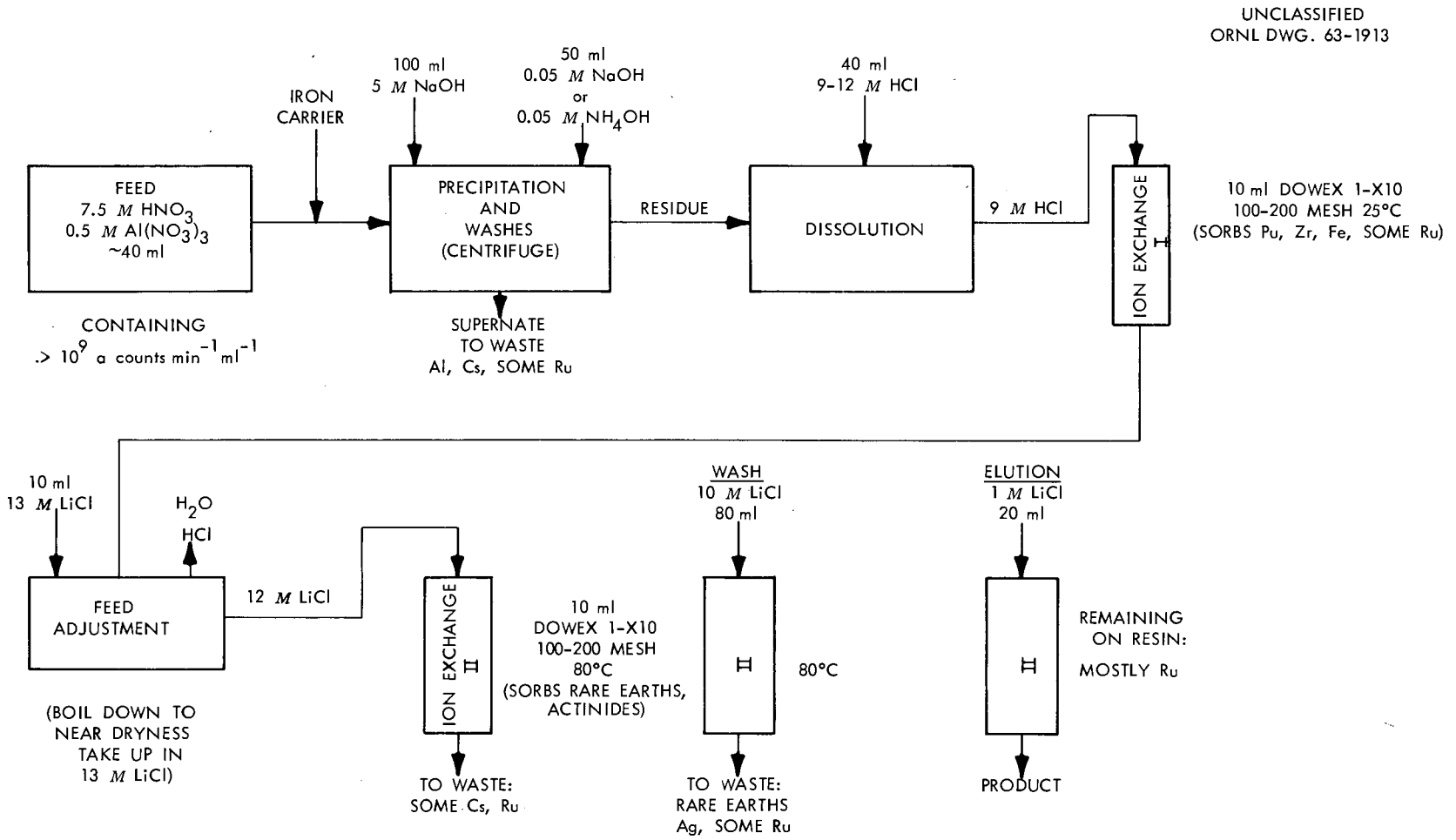

Fig. 10. Two-Column Flowsheet for Transplutonium Element Separation.

Table 1. Results of Ion-Exchange-Separation Runs

	Run CF-1	Run CF-2	Run CF-4
Feed			
Gross α activity, counts min^{-1} ml^{-1}	1.2×10^9	0.98×10^9	1.8×10^9
Cm^{242}, % of counts	38	-	38
Cm^{244}, % of counts	60	-	60
Pu^{238}, % of counts	2	-	2
HNO_3, M	~7	~7	0.9 acid def.
$Al(NO_3)_3$, M	~0.5	~0.5	2.6
Product			
Conc., counts min^{-1} ml^{-1}	1.2×10^9	1.8×10^9	4.6×10^9
Total activity, mc	15	21	31
Recovery, %	34	63	65
Material balance, %	101	103	87
Decontamination factors			
Gross γ	78	980	120
Cs^{134}	>34	>2800	>680
Ru^{106}	>40	290	>250
$Zr\text{-}Nb^{95}$	~1	4.4	?
Ce^{141}	>150	450	2.6
Pu^{238}	1300	560	>5

used. This column was also jacketed but contained 200- to 400-mesh Dowex 1-X10, which had been converted to the nitrate form. This time a resin bed of 15 ml was used. The fine resin particles and viscous solutions resulted in extremely slow flow rates, about 0.02 ml min^{-1} cm^{-2}. Unlike the previous work at tracer levels, this process gave no separation from rare earths, the product reading 5 r/hr at 10 cm. This difference is probably a result of different resin and process variables and not a result of different activity levels. No further effort is planned on this material or process.

The loading and elution curves for runs CF-2 and CF-4 are shown in Figs. 11 and 12. It is interesting to note the isolation of gamma-spectrographically-pure $Cs^{134-137}$ in the raffinate and Ag^{110m} in the first LiCl wash of the last run.

3.4.2 Californium Isolation from Transplutonium-Element Cut

Decontaminated transplutonium elements from the first two runs were processed to make two independent determinations of californium. The transplutonium elements were sorbed on a 10-ml column of Dowex 50-X8 (200- to 400-mesh) resin and were separated by chromatographic elution with 0.4 M ammonium alpha-hydroxyisobutyrate at pH 4.2 Elution curves are plotted in Fig. 13. Californium eluted in the first 8 ml of elutriant. Both alpha-pulse analysis and spontaneous fission counting were used to identify the californium. The fraction of transplutonium alpha counts due to Cf^{252} were 3×10^{-7} and 6×10^{-7} for the two determinations. The difference in these two values probably indicates the accuracy of the determinations. Based on these numbers, the eight Pu-Al alloy rods processed in cell 1, Building 4507, contained between 0.45 and 0.90 μg of californium. The balance of the TRU rods (which have been irradiated longer) should contain 100 to 150 μg of californium.

3.5 Actinide-Lanthanide Separations in Carbonate Solutions

Actinides and lanthanides can be extracted in quarternary amines or loaded on strong base anion exchange resin from dilute carbonate solutions. There was no appreciable extraction into primary, secondary, or tertiary amines. It may be possible to separate americium and curium from most of the lanthanides by either extracting the lanthanides into quarternary amines or sorbing them on anion exchange resin. Group separation of actinides and lanthanides is not possible since the heavier actinides have greater distribution coefficients than the lighter lanthanides. The practical application of this method is limited by the low solubilities of actinides and lanthanides in the dilute carbonate solutions.

Distribution coefficients for various actinides and lanthanides between 30% Aliquot 336 in diethylbenzene and 0.5 M $NaHCO_3$ are given in

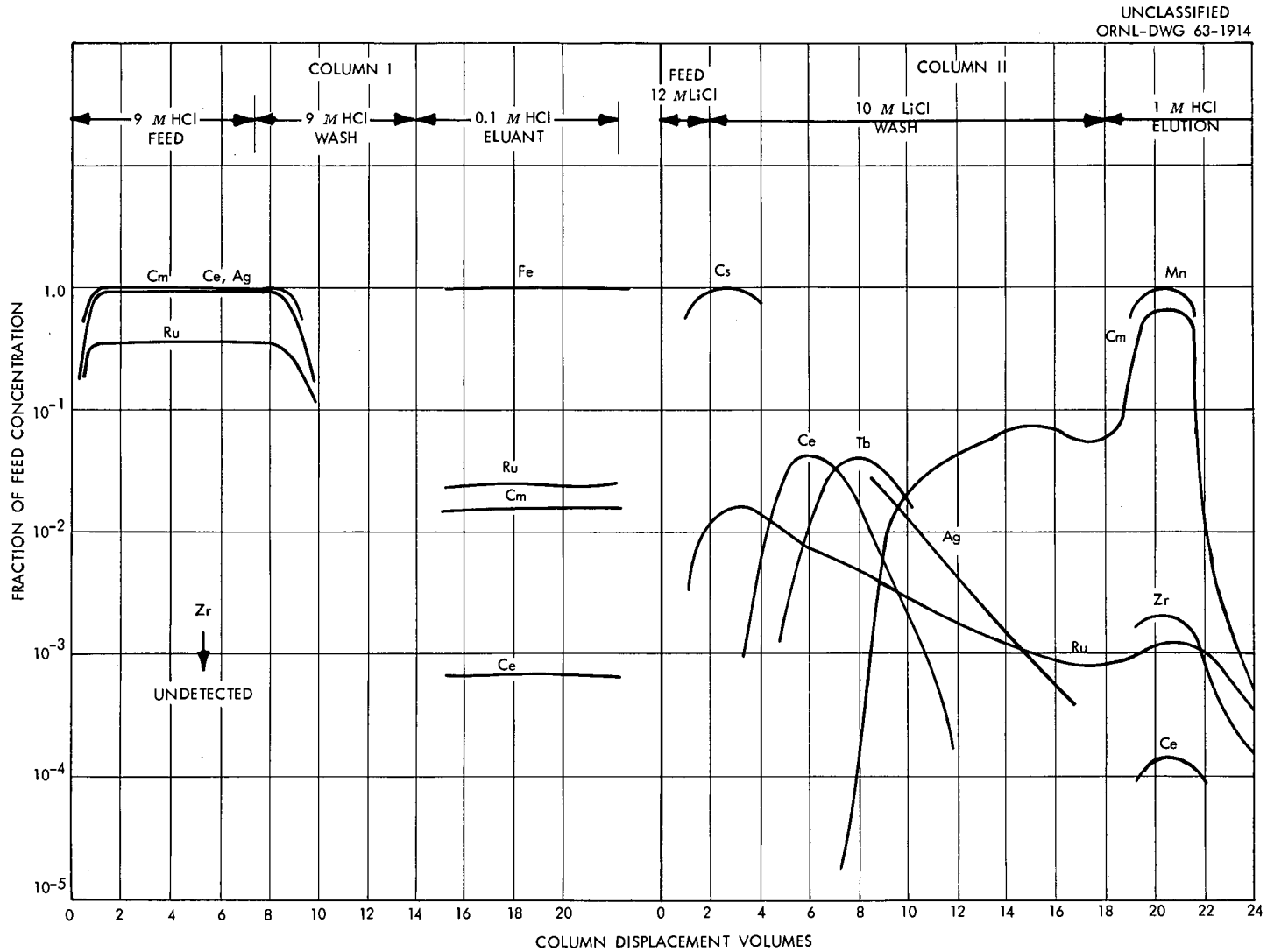

Fig. 11. Elution Curves for Run CF-20.

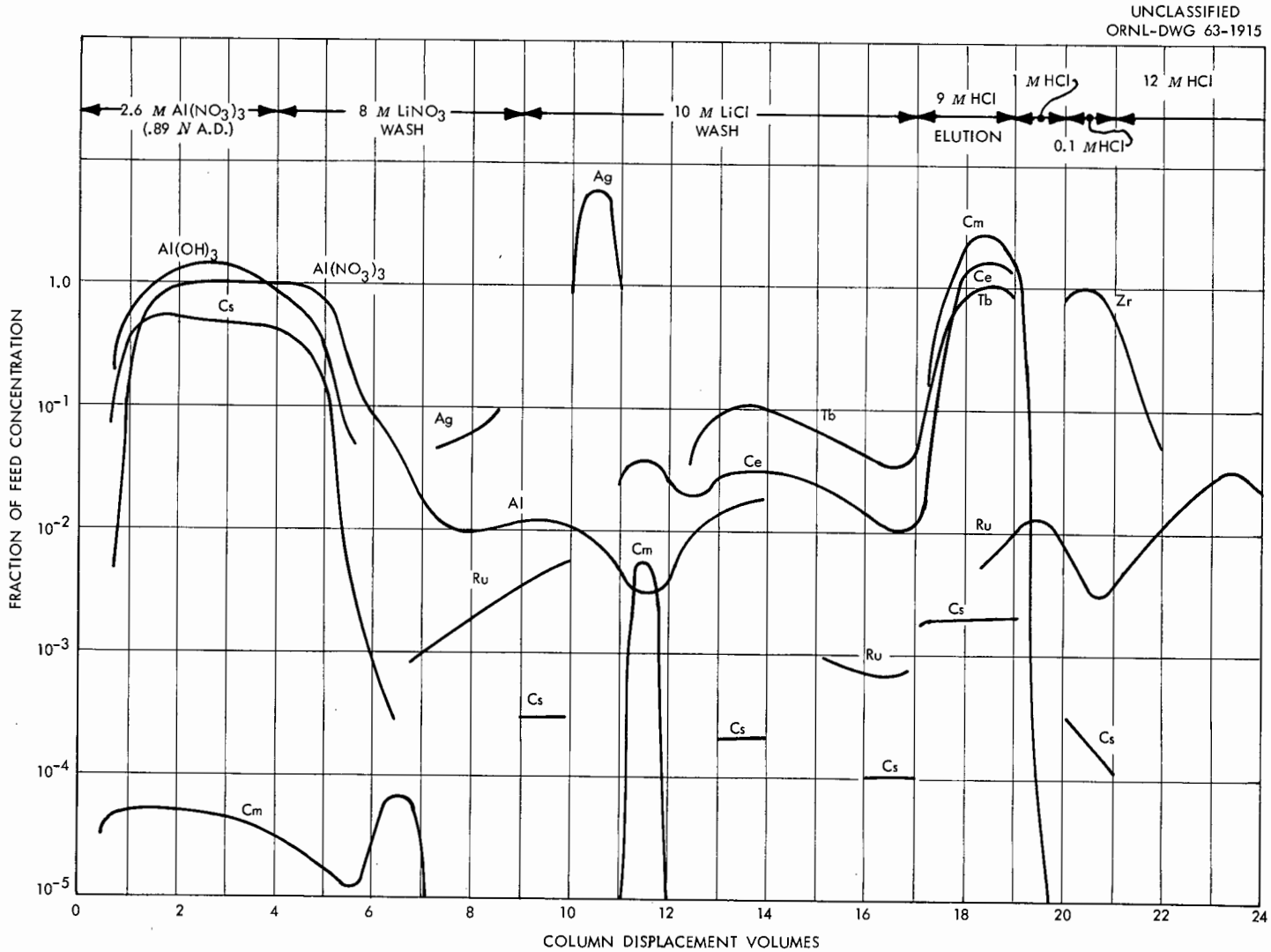

Fig. 12. Elution Curves for Run CF-4.

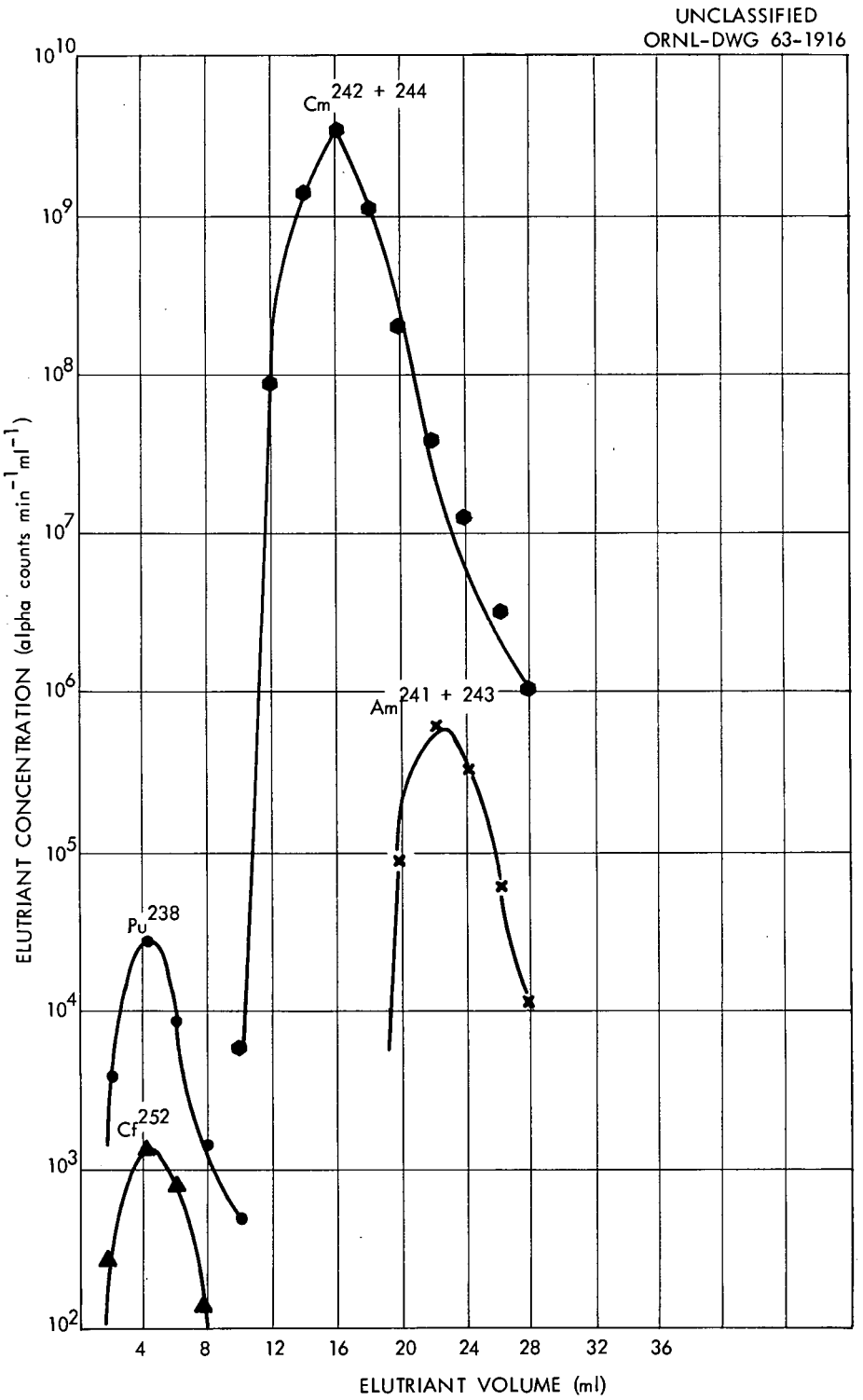

Fig. 13. Elution Curves for Californium Separation from Other Transplutonium Elements by Chromatographic Elution from 10 ml Column of Dowex 50-X (50- to 100-mesh) Resin with 0.4 M Ammonium Alpha Hydroxyisobutyrate at pH 4.2.

Table 2. This system is somewhat unusual since the trivalent actinide distribution coefficients are displaced downward. Americium often behaves much like neodymium or promethium. For this system the americium distribution coefficient is even less than the cerium distribution coefficient.

Table 2. Distribution Coefficients for 30% Aliquot 336 in Diethylbenzene vs 0.5 M $NaHCO_3$

Element	Valence	Distribution Coefficient (%)
U	6+	13.6
Np	5+	4.6
Pu	4+	2.2
Am	3+	0.52
Cm	3+	0.65
Cf	3+	2.03
Es	3+	2.51
Ce	3+	1.0
Eu	3+	4.8

The effect of $NaHCO_3$ concentration on americium, cerium, and europium distribution coefficients is shown in Fig. 14. Over the range 0.1 to 1 M $NaHCO_3$, distribution coefficients are approximately proportional to the inverse of the cube of the bicarbonate concentration. There appears to be a slight convergence of distribution coefficients at the higher concentration. Below 0.1 M $NaHCO_3$ the curves are considerably flattened, with only a slight increase in extraction noted with decreasing bicarbonate concentration. Behavior of actinides and lanthanides was essentially identical in $NaHCO_3$ solution or in Na_2CO_3 or K_2CO_3 solutions at half the molar concentration of the bicarbonate solution. Lanthanide and actinide sorption from these solutions onto a strong base anion exchange resin such as Dowex 21K was similar to extraction into the quarternary amine, Aliquot 336.

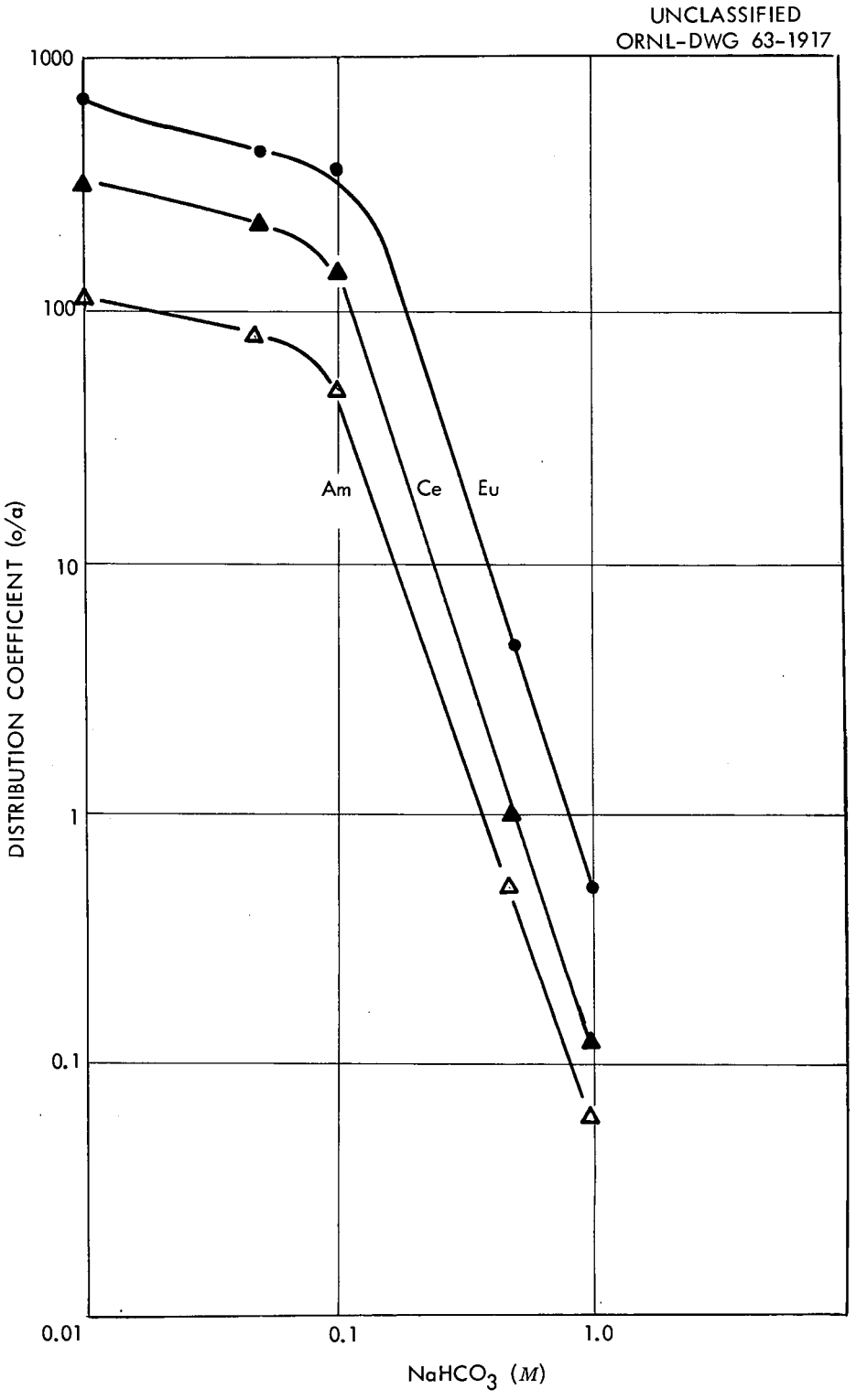

Fig. 14. Distribution Coefficients of Americium, Cerium, and Europium in 30% Aliquot-336 in Diethylbenzene as a Function of $NaHCO_3$ Concentration.

3.6 Californium, Einsteinium, and Fermium Behavior in the $LiNO_3$-Anion Exchange System

Elution from anion exchange resin with 4.4 M $LiNO_3$ gave poor californium-einsteinium-fermium separations compared with the excellent americium-curium separation reported last quarter.[5] This was a result of an unexpected reversal in elution positions of the transcurium elements.

Distribution coefficients of trivalent actinides between Dowex 1-X8 resin and 4.4 M $LiNO_3$ solution adjusted to pH 1.5 are given in Fig. 15. These were determined by batch equilibration of resin with 4.4 M $LiNO_3$ containing actinides, and the results were confirmed by noting the elution order from Dowex 1-X8 resin columns. The same relative behavior of trivalent actinides was noted when Alamine 336·HNO_3 was equilibrated with a 6 M $LiNO_3$ solution of the actinides.

Based upon previously determined distribution coefficients of Pu^{3+}, Am^{3+}, and Cm^{3+}, it was anticipated that distribution coefficients of the transcurium elements would continue to decrease with increasing radius of the hydrated ions, which increases with atomic number for these elements. However, this trend reversed with einsteinium and fermium, which have greater distribution coefficients than californium. It is hypothesized that since the f electron half shell is filled for curium, Li^+ may be more effective in partially stripping the hydration sheath from transcurium elements. This partial dehydration would enhance complex formation with the nitrate ion and thus result in a reversal of the sorption trend on anion exchange resin.

3.7 Preparation of PuO_2 for HFIR Target Prototypes

A method of preparing high-fired PuO_2 glass was investigated. Successful preparation will make it possible to grind the oxide to a specified particle size. This is important since the PuO_2 powder must be coarse enough for satisfactory preparation of Al-PuO_2 cermets with a continuous aluminum phase.

A solution 0.5 M in $Pu(NO_3)_4$ and 4 M in HNO_3 was rapidly mixed with an equal volume of 8 M NH_4OH to precipitate $Pu(OH)_4$. The precipitate was

[5] D. E. Ferguson, Transuranium Quar. Prog. Rep. for Period Ending November 30, 1962, ORNL-3408 (May 31, 1963).

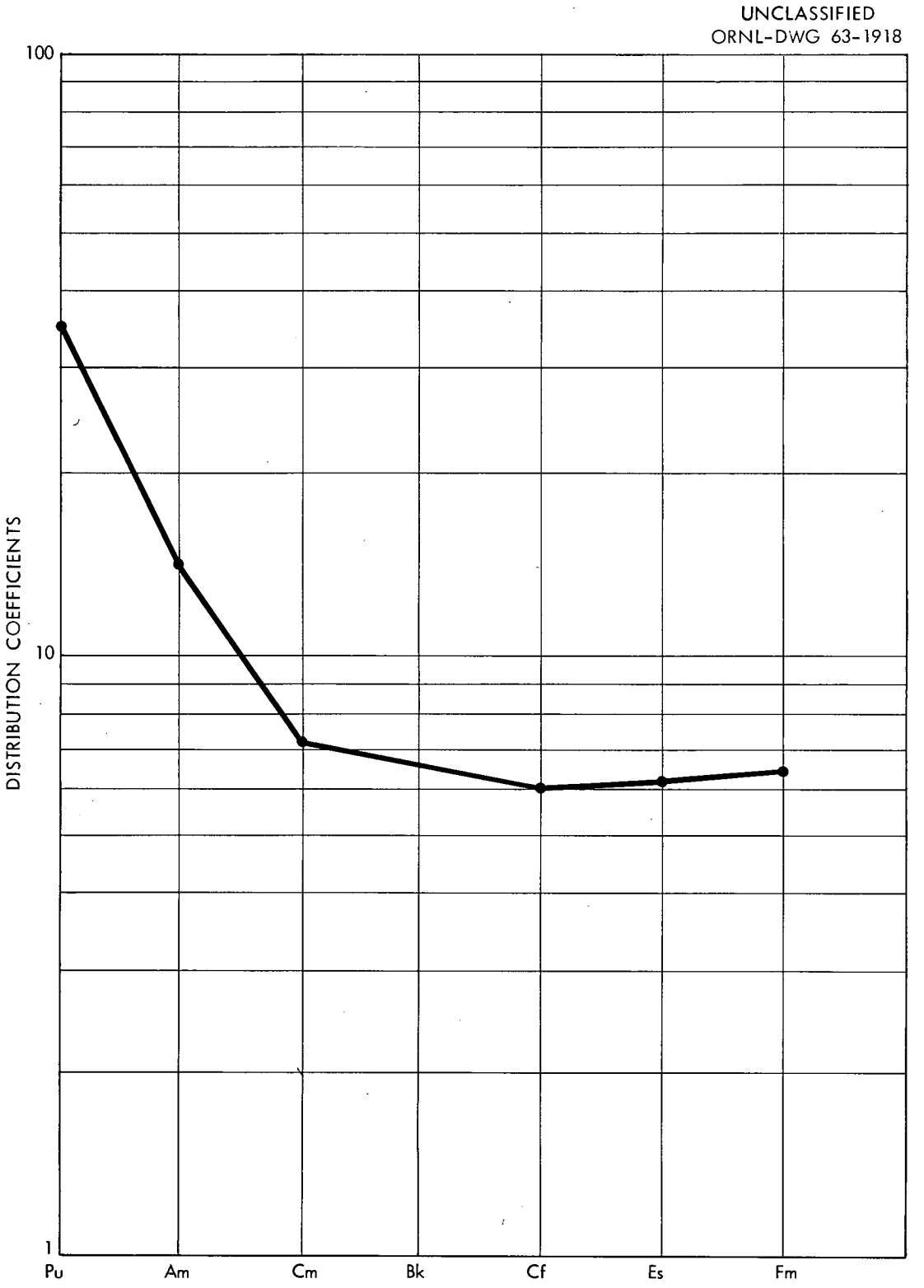

Fig. 15. Actinide Distribution Coefficients in 4.4 M $LiNO_3$ for Dowex 1-X8.

washed thoroughly with 2 M NH_4OH and dried at 150°C. Firing the dried solid at 1200°C produced a glassy solid with a density of 10.99, which is 96% of the theoretical density of PuO_2. Most of the particles were about 1 to 3 mm in diameter.

4. EXPERIMENTAL ENGINEERING STUDIES

P. A. Haas A. D. Ryon C. D. Watson

Experimental studies are being carried out as necessary to aid in the engineering design of the chemical processing cells. These include testing of protective coatings, development of process equipment components, and testing in a mockup of procedures and equipment proposed for use in the cell cubicles.

4.1 Materials Evaluation

G. A. West

A radiation-damage testing program of protective coatings proposed for the TRU Facility is nearing completion. Currently, 42 coatings have been exposed to a Co^{60} gamma source at an intensity of about 1×10^6 r/hr at 40 to 50°C in air and in deionized water. Polyamide cured epoxy systems exhibited superior radiation resistance while immersed in deionized water. A listing of the best coatings and maximum tolerable exposure follows:

Amercoat No. 66 system with Fiberglas -- to 4.8×10^9 r
Plasite No. 7155 -- to 4.5×10^9 r
Amercoat No. 66 system without Fiberglas -- to 4.5×10^9 r

In general, all coatings tested are less affected by radiation when exposed in air environments than when immersed in water. A few coatings have now attained an exposure up to 6×10^9 r in air and appear usable. Fourteen coatings, consisting of epoxies and modified phenolics, were considered usable after exposures in deionized water from 1×10^9 r to 4.8×10^9 r. Most of the failures at the higher radiation levels resulted from deterioration of film, loss of adhesion, and a marked decrease in resistance to impact and gouging. Coatings reinforced with Fiberglas fabric retained resistance to impact and gouging better than unreinforced coatings.

Decontaminations by a water flush and a nitric acid scrub at room temperature were determined for the various types of coatings.[6] A decontamination factor of 3×10^4 would reduce the contaminant used to about 0.1 mr/hr and would probably be desirable for most controlled maintenance work. The maximum decontamination factors [DF = (initial activity, mr/hr)/(activity following decontamination, mr/hr)] were as follows:

Vinyl	US Stoneware, RO221 -- with silicone -- TP 216 seal	9.33×10^3
Polyester	Prufcoat, epoxy modified polyester	2.7×10^3
Epoxy	Wisconsin Protective Coating, Plasite-9009	1.30×10^3
Modified Phenolic	Wisconsin Protective Coating, Plasite-7155	6.75×10^2
Inorganic	Amercoat-1680	0.63×10^2

4.2 Disconnect Development

T. S. Mackey

Tantalum disconnects fabricated by forming the conical sealing surface on the tubing itself were tested and leaked at unacceptably high rates. Tantalum disconnects with machined surfaces also leaked, but after burnishing the male and female cone surfaces with hardened rollers, satisfactory leaktightness was obtained even after several make and break cycles.

Ten of the commercially fabricated disconnect clamps were checked for misalignment. Measurements made at 60 lb of torque showed misalignment ranging from 1.1 to 1.8°. The acceptable limit is 0.5°. The upper clamp arm contact surface was then machined at an angle of 1.5°. Subsequent misalignment tests indicated acceptable values for all clamps except one, which gave 0.8° misalignment.

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[6] Data of A. B. Meservey *et al.*, of Chemical Technology Division, ORNL.

4.3 Cell Mockup

T. S. Mackey

Assembly of the cubicle top and component parts was completed. Fabrication of the mockup of the top of the cell has started. This will provide a means of testing equipment removal and maintenance concepts connected with the tank pit and cell cubicle. Two extended-reach master-slave manipulators were returned to the shop for adjustment after initial testing showed faulty tape and cable adjustments. Tests of manipulators, equipment-rack handling devices, and lighting-fixture handling devices are in progress.

4.4 Component Testing

T. S. Mackey

A simple means of forming bellows was used successfully for forming stainless steel test bellows. Zircaloy-2 and tantalum thin-wall tubing was ordered for fabrication of bellows from these materials for stem seals in process valves. A commercial source of tantalum bellows was located, and tantalum test bellows will be purchased if the terms are acceptable. The "Magneform" fabrication of tantalum bellows from tubing was not successful because of insufficient forming force for all conditions tested.

Fabrication of a prototype sampler pump was completed.

4.5 Americium and Californium Irradiations

S. D. Clinton

Four capsules containing 100 mg each of Am^{241} and one capsule containing 0.5 μg of Cf^{252} were prepared for irradiation in the ORR. Each of the capsules contained three cobalt foils in quartz for determining the thermal neutron flux. The capsules will be irradiated according to the following schedule:

Sample No.	Target Material	Date In	Date Out	Approximate Neutron Flux
E-12-23	Am^{241}	12-16-62	2-10-63	3.0×10^{14}
E-12-24	Am^{241}	12-16-62	2-10-63	3.5×10^{14}
E-12-25	Cf^{252}	12-16-62	1-13-63	4.0×10^{14}
F-1-2	Am^{241}	1-13-63	4-7-63	4.0×10^{14}
F-1-3	Am^{241}	1-13-63	4-7-63	3.5×10^{14}

Engineering calculations for irradiating gram quantities of Am^{241} in the ORR were completed. The 8-in.-long, 0.5-in.-OD, welded aluminum capsules will each contain 3 g of Am^{241} as pressed pellets of AmO_2-Al. The weight of aluminum in each capsule is 60 g. An ORR core piece was designed and fabricated for irradiating up to 18 capsules. The calculations are based on an average perturbed thermal neutron flux of 3.0×10^{14} neutrons $cm^{-2} sec^{-1}$, a peak-to-average thermal flux ratio of 1.2, an irradiation time of 50 days, and an initial pellet-to-capsule radial-gas gap of 1 mil. Assuming 100% release of the fission product gases, the maximum stress in the type 1100 aluminum capsule is 500 psi. Temperatures and heat fluxes in the irradiation are as follows:

Conditions	Surface Heat Flux ($Btu\ hr^{-1}\ ft^{-2}$)	Surface Temp. (°F)	Central Temperatures He Gap (°F)	Central Temperatures Air Gap (°F)
Average	438,000	192	682	789
Peak	526,000	205	793	922
20% increase in reactor power	631,000	222	928	1082

4.6 Pulse Column Tests

F. L. Daley

A three-column complex built of corrosion-resistant materials (glass, tantalum, and Teflon) was erected to test a complete flowsheet (extraction, scrub, and stripping), a single column, or any phase of the flowsheet. Each column is a 3/4-in.-ID x 48-in.-long glass pipe, with a water jacket for temperature control (Fig. 4.16). The standard column internals consist of tantalum sieve plates (1/32-in. holes, 5% free area) spaced 1/4 in.

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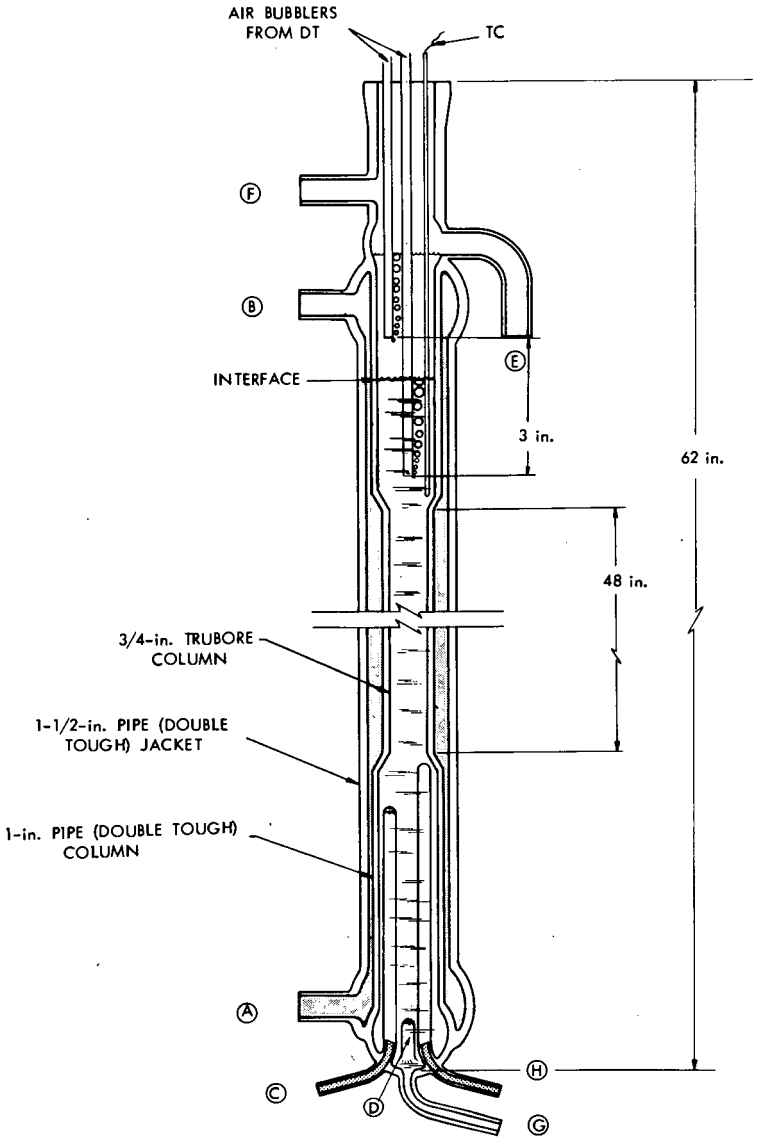
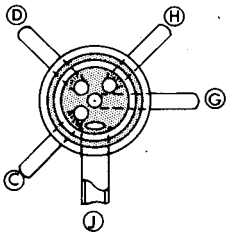




NOZZLE SCHEDULE

MARK	SIZE	WALL	SERVICE
A	1/2 in. OD	HVY.	WATER JACKET IN
B	1/2 in. OD	HVY.	WATER JACKET OUT
C	1/4 in. OD	HVY.	ORGANIC IN
D	1/4 in. OD	HVY.	ORGANIC IN (ALT.)
E	1/2 in. OD	HVY.	PRODUCT OUT
F	1/2 in. OD	HVY.	OVERFLOW
G	1/4 in. OD	HVY.	AQUEOUS OUT
H	1/4 in. OD	HVY.	THERMOWELL
J	1/2 in. OD	HVY.	PULSE

Fig. 16. Typical Pulse Column.

apart. Interface position was measured by an air-bubbler signal to a differential-pressure transmitter and controller that controlled the interface level by a pressure pot on the aqueous discharge line. Intercolumn flow of the organic phase was obtained by pulse pumping, and aqueous flow was by pump.

Shakedown tests of the system with the Tramex flowsheet revealed two minor hydraulic problems: one required increased resistance in the lines at the bottom of the column to minimize pulse loss, and the other was the limited range of intercolumn organic flow obtainable by pulse pumping.

4.6.1 Scrub Tests

A series of runs using europium tracer to measure the efficiency of the columns was made to test pulse wave and column internals. The results (Table 3) showed that the lowest stage heights (HETS) were obtained at the highest pulsed volumes tested, which were near the emulsion flooding point. A truncated sine-wave pulse was significantly better than a sine wave. Attempts to improve efficiency by inserting several types of coalescing sections (Teflon Raschig rings, graphite rod, large free area, sieve plates made of tantalum and graphite) were not successful in reducing the stage height below 16 in., which was the best for the standard sieve plates.

5. TARGET FABRICATION PROCESS AND EQUIPMENT DEVELOPMENT

D. A. Douglas

Work in the Metals and Ceramics Division falls under four broad categories: (1) development of necessary fabrication processes to produce an HFIR target rod, (2) irradiation of targets to test the adequacy of the design and the fabrication processes, (3) design of the required fabrication equipment and operation of a glove-box production line to make plutonium-bearing targets for the initial reactor loadings, and (4) the design of equipment for the remote fabrication of targets in three cubicles of the TRU Facility. Progress in these areas for the last quarter is reported below.

Table 3. Efficiency of Pulsed Columns for Tramex Scrub Section, Using 9 M LiCl--0.3 M $AlCl_3$ to Scrub Europium from 30% Alamine 336 in Diethylbenzene

Flow rate: organic, 9 ml/min; aqueous, 3 ml/min

Temperature: 50°C

Column Internals	Type of Pulse Wave	Pulsed Velocity (in./min)	HETS (in.)
Standard Ta sieve plates (1/32-in. holes, 5% free area)	Truncated sine	4.0[a]	28
	Truncated sine	4.0	27
	Truncated sine	7.5	16
	Truncated sine	8.0	19
	Sine	24.0	48
	Sine	48.0	44
10 in. of standard plates alternated with 2 in. of Ta sieve plates (3/32-in. holes, 45% free area)	Truncated sine	5.6	24
	Truncated sine	8.4	28
	Sine	26.0	40
	Sine	32.0	30
10 in. of standard plates alternated with 2 in. of Teflon Raschig rings	Truncated sine	4.0	28
	Truncated sine	7.0	25
	Truncated sine	9.0	20[b]
	Truncated sine	9.0	18
2 in. of standard plates alternated with 4 in. of graphite-rod packing	Truncated sine	6.0	25
	Truncated sine	9.0	30
5 in. of standard plates alternated with 1 in. of graphite sieve plates (3/32-in. holes, 45% free area)	Truncated sine	8.0	21
	Truncated sine	21.0[c]	19

[a]Pulse amplitude = 0.1 in.

[b]Organic freshly loaded.

[c]Pulse amplitude = 0.35 in.

Since heat-transfer characteristics are quite sensitive to the type of oxide dispersion produced, one of the most critical process development problems has been to achieve a continuous aluminum matrix with a uniform dispersion of fine oxide particles. A satisfactory solution was devised by the Powder Metallurgy Group by controlled additions and blending of superfine aluminum powders. However, a successful breakthrough by the Chemical Technology Division in producing plutonium oxide in a larger size range has enabled us to shelve this procedure and take advantage of the simpler process that large oxide particles afford.

A second critical problem has been that of obtaining end closures with sufficient strength and integrity that they will successfully survive about 18 months of reactor exposure. It has been judged that welding alone will not be sufficient for that purpose; consequently, several other means for reinforcing the end seal are being explored.

A mockup of the plutonium target line is being assembled, and design efforts on the equipment for the fabrication cubicles in the TRU Facility are progressing on schedule. Deferral of the fabrication of a part of this equipment already designed until next fiscal year may delay the mockup and testing of these items.

5.1 Target Fabrication Process Development

5.1.1 Powder Metallurgy -- (W. J. Werner)

A process has been developed for producing continuous aluminum-matrix pellets from oxide with an average particle size of 6 μ. The process involves (1) blending of the oxide with an equal volume of ultrafine aluminum (average particle size 0.03 μ, existing in the form of friable agglomerates 0.1 to 100 μ in size);[7] (2) the addition of enough vacuum-annealed, type 1100 (-325 mesh) aluminum to obtain a final composition of 16 vol % oxide; and (3) reblending and pressing.

Figure 17 shows the microstructure of a pellet produced in the aforementioned manner. A minimum agglomeration of oxide particles and many other properties of an ideal dispersion were noted. Subsequent

[7] Prepared by vapor deposition and supplied by the National Research Corporation.

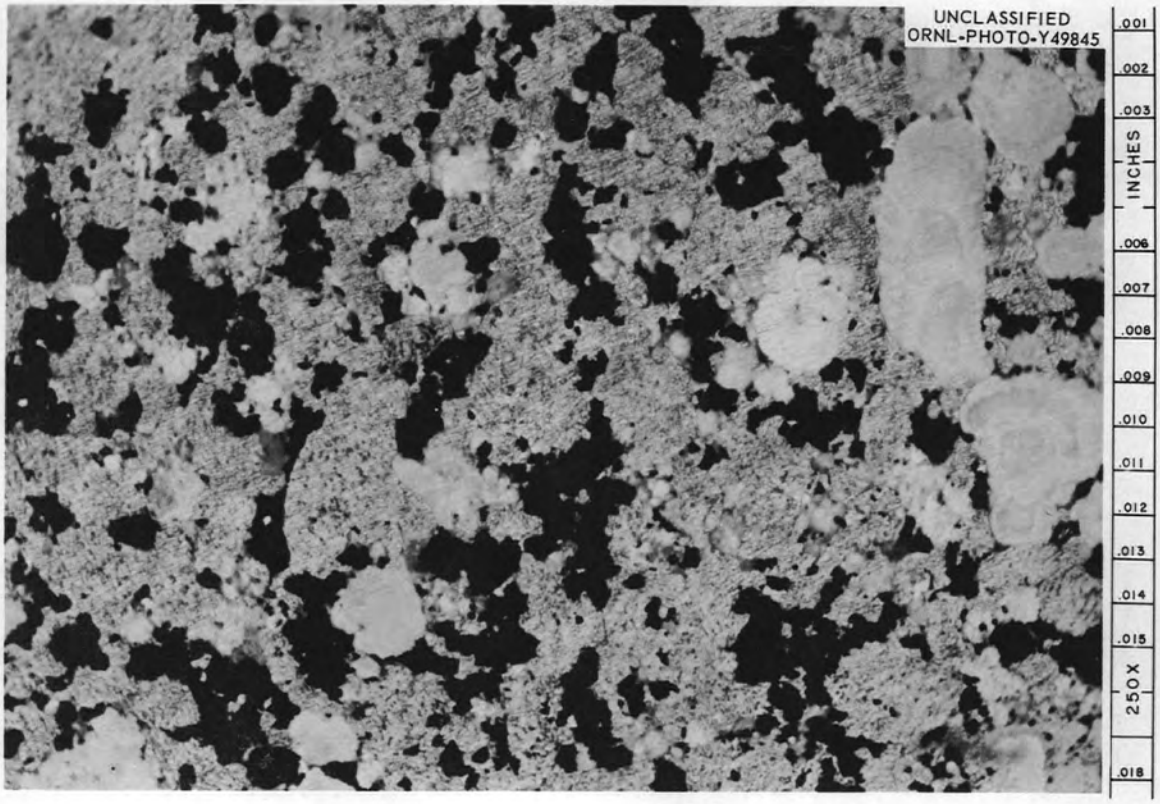


Fig. 17. Microstructure of Target Pellet Containing 16 Vol % Rare Earth, 16 Vol % Ultrafine Aluminum (0.03 μ), with Balance Being -325 Mesh Aluminum. Reduced 10%.

experiments showed that for the particular materials and concentrations used, the critical amount of ultrafines was between 13 and 19 vol %. Incidentally, this amount would increase with decreasing oxide particle size.

The initial ultrafine powder (93 wt % active aluminum metal) was pyrophoric in air at room temperature. Subsequently, oxide-stabilized ultrafines (83 wt % active aluminum metal) were received. These powders are reported to be stable in air at temperatures up to 350°C. They will, however, support combustion when ignited.

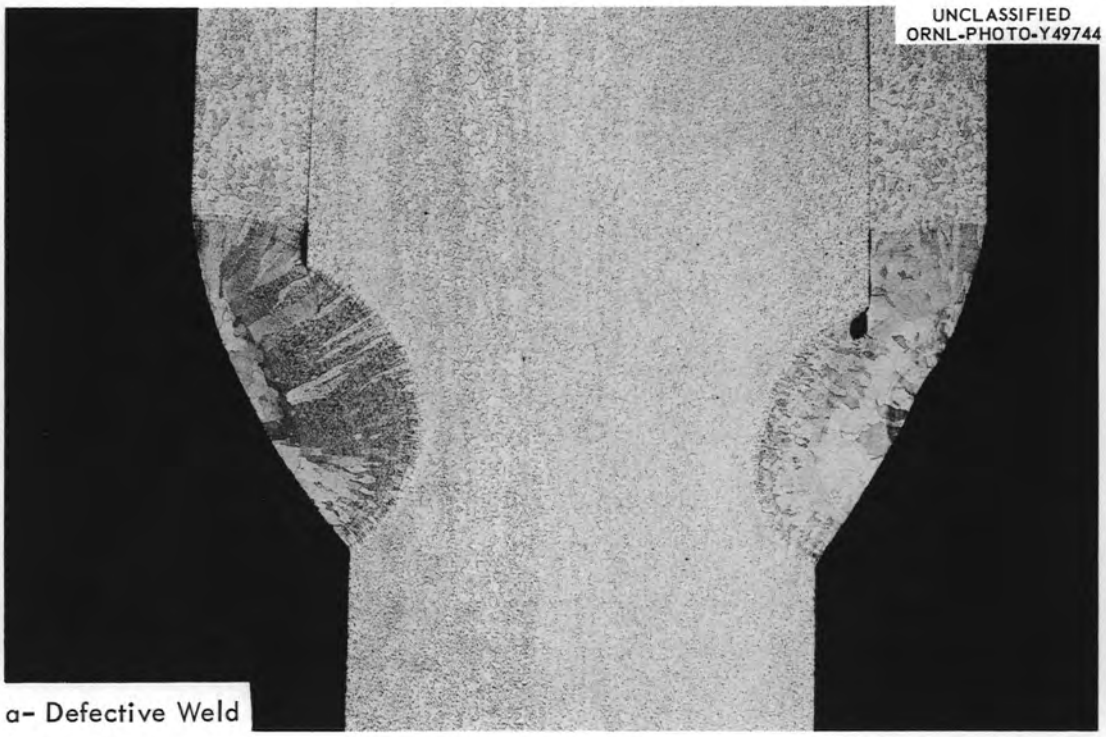
5.1.2 Welding and Brazing Studies -- (C. H. Wodtke)

The HFIR target rods consist of type X-8001 aluminum tubes, 3/8-in. in outer diameter, with a 0.060-in. wall, sealed at each end with plugs. The initial end closure can be performed in the open, but the final closure must be made in a cell. Developmental work is being concentrated on the production of a welded-and-brazed double seal. Brazing of longitudinal aluminum fins to the tubes is also being studied.

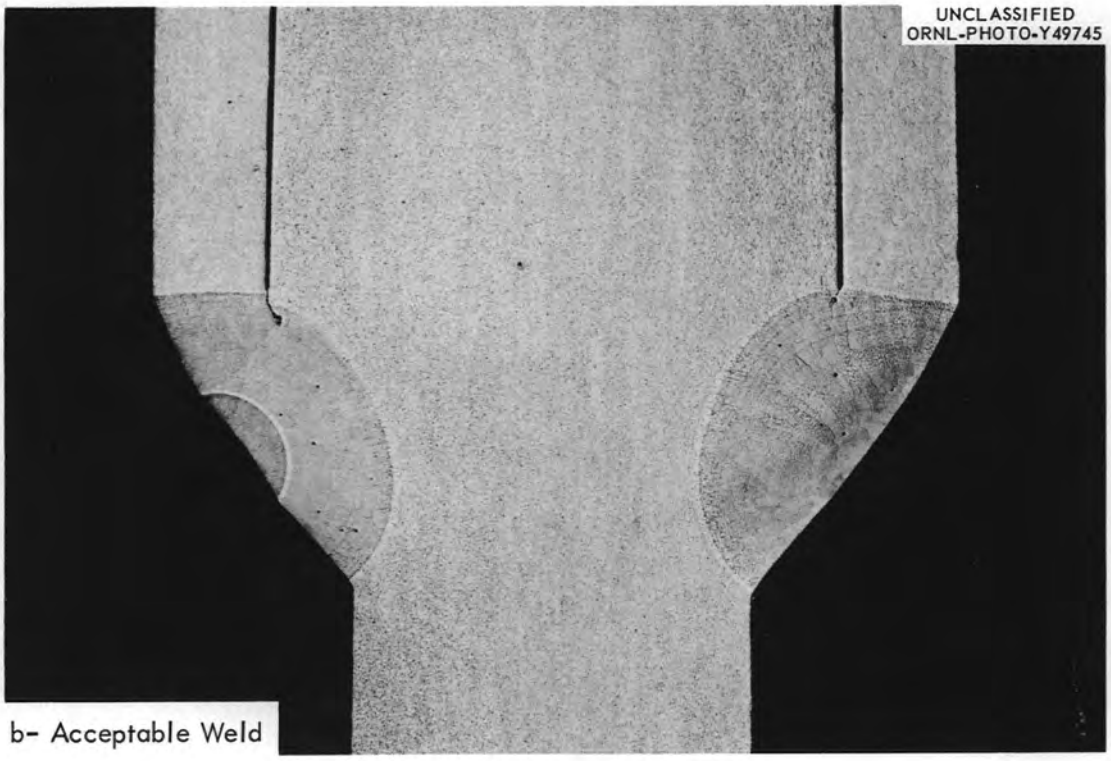
Welding of End Plugs. -- To date, welding has been done with a standard welding power supply. The results, although promising, have not proved reliable, and serious weld defects are not infrequent. A typical defective corner joint (Fig. 18a) shows incomplete fusion between tube and end plug as well as root cracking. A much better weld, however, is shown in Fig. 18b.

A mechanized weld cycle appears necessary to consistently reproduce the control of variables and operations, all of which must be performed in about 7 sec. To accomplish this, an arc-length-controlled welding head and automatic electric controls for the welding cycle are being set up. A programed welding power supply with more refined control over the welding cycle is also being procured. It is expected that this equipment will be capable of producing welds with greater consistency.

Brazing of End Plugs. -- For the highly reliable containment of gases, a double seal made by brazing, in addition to welding, is being explored. Induction brazing appears to be workable for attaching end plugs in a cell. A joint was designed which utilizes a small plug brazed under the



a- Defective Weld



b- Acceptable Weld

Fig. 18. HFIR Target End-Weld Closures.

subsequently welded plug. Preliminary work indicates that the method warrants further attempts to establish a consistently satisfactory induction-brazing procedure. Leak tests and metallographic examinations of preliminary brazes showed that leaktight joints can be made. Further effort will be centered on reducing the amount of porosity and flux entrapment now obtained.

A small induction heater for use in the rod assembly area has been obtained from surplus property. It will be set up in the laboratory and checked for proper operation.

Brazing of Fins. -- Tubes with integral fins cannot be obtained in time for use in the irradiation test rods to be made soon. An attempt is being made to establish a furnace brazing procedure to attach the fins. A preliminary brazing fixture has been produced and test brazes prepared. A typical braze cross section is shown in Fig. 19.

5.2 Development of Equipment for the Fabrication of Plutonium Targets

A. L. Lotts

There was little change in the status of the plutonium target fabrication equipment during this reporting period. The principal activity was in the use of some of the equipment in attempts to develop acceptable procedures for various fabrication steps. The present status is noted as follows:

5.2.1 Glove Boxes

All glove boxes were installed in a mockup area in Building 4508 and now await the installation of processing equipment.

5.2.2 Powder-Preparation Equipment

The status of the powder-preparation equipment remains the same as previously reported.

5.2.3 Pellet-Preparation Equipment

The press was used in making a substantial number of dummy pellets. Wrinkling of pellet liners, which is attributed to the halting press stroke, was the only significant problem. The addition of an accumulator

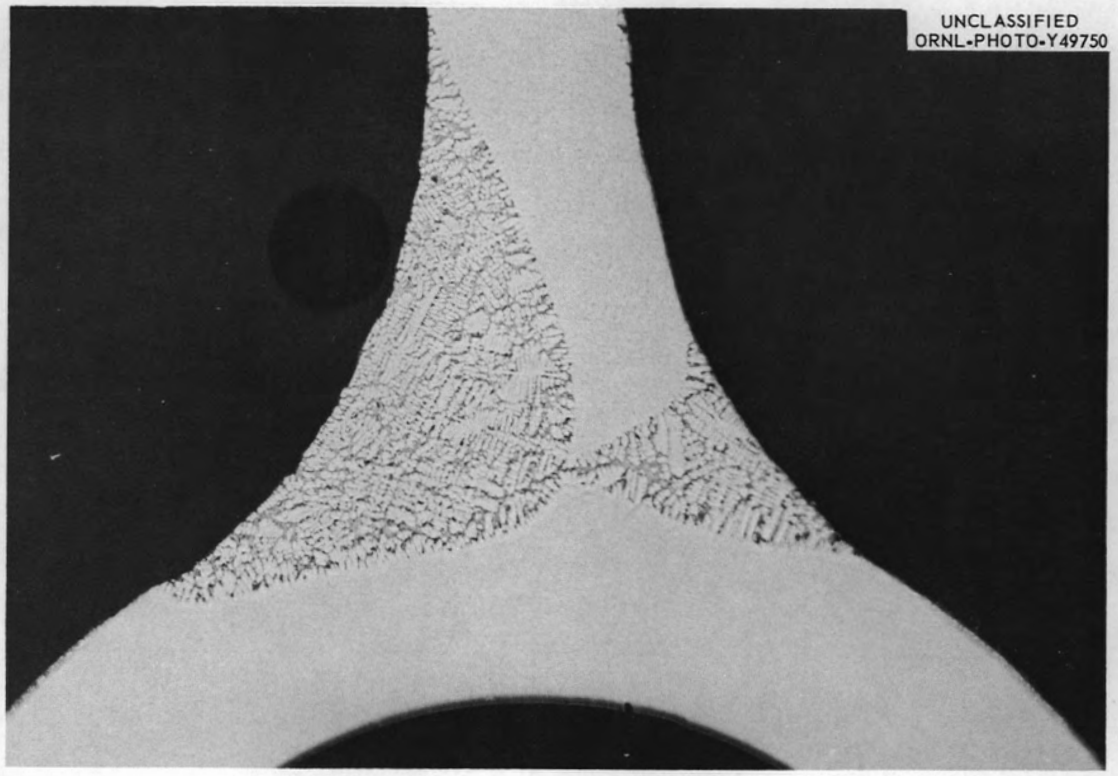


Fig. 19. Cross Section Through Fin-to-Tube Brazed Joint.

to the hydraulic system probably will provide a continuous fast press stroke and ensure the fabrication of pellets with smooth liners. A new improved press base was constructed and has performed satisfactorily.

5.2.4 Tube-Closure Equipment

The tube-closure equipment was used primarily as a development device during this reporting period. In such an application, it has become increasingly apparent that a continuously variable welding speed should be available and that the power supply should have a wider range at low welding current. Nevertheless, a number of moderately successful welds were made with this equipment. Blowout of the closure welds on long tubing using the step-type end cap is not now a severe problem, but root cracking still plagues the weld. Table 4 gives the statistical

Table 4. Summary of Target-Welding Results

Tube Length (in.)	First End Cap		Closure End Cap	
	Number Welds Attempted	Number Successful Welds	Number Welds Attempted	Number Successful Welds
20	20	18[a]	6	6
10	8	8	8	6[b]
6	30	30	30	24[c]

[a]Two welds failed because of improper chill-ring fit-up.

[b]Two welds failed by blowout.

[c]Three welds failed because of improper cap fit-up; three failed by blowout.

results of welds made with the equipment during this period. Successful welds (Table 4) are those which passed a helium leak test.

Improved heat sinks, constructed of massive aluminum and rotated with the tube, performed better than others tried previously.

5.2.5 Hydrostatic-Collapse Equipment

The hydrostatic-collapse equipment was constructed and now awaits installation.

5.2.6 Helium Leak Test Equipment

The helium leak test chamber was completed.

5.2.7 Plutonium Target Fabrication Mockup

The mockup area in Building 4508 was completed; and, as previously noted, the glove boxes were installed.

5.3 Design and Development of Target Fabrication Equipment

M. K. Preston A. L. Lotts R. I. Deaderick

The basic criteria established for the target-element fabrication process have changed little from that outlined in the first quarterly progress report.[8] The following summarizes the status of equipment development in the three cubicles of the TRU Facility that are assigned for target fabrication.

5.3.1 Cubicle 3

The process and equipment flow diagram for cubicle 3 has not changed; however, a slight change in equipment location within the cubicle is being considered. To furnish a firm mounting base for the pellet-load scale, which must be accurate to ± 5 mg, the scale will be supported from the wall between cubicles 3 and 4. This change also necessitates slight relocation of the equipment fabrication circle.

During the last quarter, conceptual design was started on two additional pieces of equipment. Final design for three items of equipment was completed and approved for construction. Detail designs for two other items were completed, but have not yet been checked and approved. Bench testing and modifications of three pieces of equipment are continuing.

Cell-Equipment-Removal Crane. -- Equipment-mounting bases are to be installed on the sloping floor to support equipment mounts. This will furnish a level mounting plane and eliminate contamination traps under equipment. Since the bases will not provide a smooth flat surface on

[8] D. E. Ferguson, TRU Quart. Progr. Rept. February 28, 1962, ORNL-3290.

which equipment may be slid to the removal position in the center of the cubicle, a concept for an in-cell crane is being developed to handle heavy and bulky equipment in the cubicles.

Actinide Calciner. -- The following criteria were established for the design of the actinide calciner:

1. The operating temperature is to be controllable to a maximum of 900°C.

2. The furnace cavity is to accommodate the blending container.

3. A fixture is to be employed for insertion, positioning, and removal of the container.

4. The furnace cavity is to be connected to the cubicle off-gas system.

Conceptual design has not started.

Batch Scale. -- Design of the batch scale is held up pending the successful operation of the pellet-load scale.

Blender-Dispenser. The detailed design of the blender-dispenser is now being processed for final approval.

Pellet-Load Scale. Machining of parts necessary for the remote operation of the pellet-load scale was completed. With this system, specified amounts of actinide-aluminum powder can be weighed in pellet die assemblies prior to pellet forming. A self-balancing system, employing Selsyn motors, a torsion balance, a differential transformer transducer with electronic indicator, and a recorder with the slide wire removed, was assembled. Only the scale and the Selsyn receiver will be located inside the cubicle. Response of the scale will be transmitted to the control cabinet outside the cubicle by a connecting cable.

Initial tests of the scale have demonstrated the required accuracy; however, tests on the entire system indicated that minor changes will be required to control oscillation. For this purpose, a magnetic damper is being installed on the motor drive shaft in the recorder.

Cap Powder Loader. -- The detailed design of the cap powder loader has been completed and is now being processed for final approval.

Die Assembly Feeding Station. -- Fabrication of the die assembly feeding station is complete, and bench tests are being conducted. Basic operation is satisfactory, but minor modification to the die magazine and the installation of a throttle valve to smooth out the action is desirable.

Pellet Press. -- Bench tests of the press are continuing, following minor redesign of some components to improve operation and reliability. To date, about 200 pellets (showing little or no wrinkling of the pellet liner) have been pressed. The press operated quite satisfactorily. Control of the pressurized water system is sufficiently sensitive that termination of ram travel at the bottom punch discharge point can be accomplished by valving rather than by using the existing mechanical stop, which introduces undue stresses in the press frame. Electrical controls for the valve were designed and will be installed.

Pellet Ultrasonic Cleaner and Pellet Dryer. -- Detail drawings are being completed on the pellet cleaning and dryer equipment. The electrical connections for the pellet dryer were designed so that they are automatically connected when the dryer is placed into the auxiliary enclosure. The electrical and piping connection for the ultrasonic cleaner will be completed after receipt of the unit from the vendor.

Transfer Arm III. -- Detail design of the transfer arm is complete and has been checked for correctness. The design is now being processed for final approval.

Miscellaneous Containers and Magazines. -- Containers and magazines used for transporting, feeding, and collecting small items to the various equipment in cubicle 3 were bench tested and appeared to be satisfactory.

5.3.2 Cubicle 2

The process flowsheet and equipment layout for cubicle 2 have not changed. Conceptual designs on two items of equipment, which had been approved, were revised and reissued. Detail design of four pieces of equipment has started. Fabrication was completed on one item of equipment; construction of one major equipment group is about 90% complete.

Pellet Inspection and Loading Equipment.--Machining of the various parts of the pellet inspection and loading equipment is complete, and assembly is under way.

End-Cap Welding Machine. -- Conceptual design of the end-cap welding equipment is being deferred until a reliable welding procedure can be developed.

Helium Leak Test System. -- Equipment for test and evaluation of the simplified helium leak test system, as outlined in the previous report, was assembled. The use of a larger spring on the leak-test-chamber sealing cap, and thinning of the "O-ring" holding disk to give more O-ring compression, decreased the leak rate of the chamber from 1.5×10^{-9} std cc/sec to 1.5 to 2.0×10^{-10} std cc/sec. Additional design is under way on the disconnects that will be used for removal and replacement of the two 4-in.-diam absolute filters required in the system. Interconnecting cables were assembled for the remote operation of the leak detector at distances up to 50 ft.

Hydrostatic-Collapse Equipment. -- Fabrication of the hydrostatic pressure chamber is still being deferred until more data are obtained on the pressure requirements. By the use of magnetic-pulse forming methods (Magneforming) the radial gap between pellet and liner was reduced to a range of 0.0002 to 0.0005 in. This gap varied from 0.0005 to 0.001 in. for hydrostatic-pressure collapse. Tubing and pellets are being fabricated to conduct additional tests with the Magneform machine.

Target Ultrasonic Cleaner. -- Detail design of the target ultrasonic cleaner is about 30% complete. Vendors are being consulted for comments on the proposed design to determine power requirements and the optimum container configuration.

Target Transfer Arm II. -- Conceptual design of the target-transfer arm has been delayed pending additional design information on the process equipment in the cells and a firm design of the target-rod configuration.

Target Transfer to Cubicle 1. -- Studies showed that target rods cannot be successfully collapsed with the hexagonal cans in place but that the hexagonal can should be attached remotely following collapse.

The detailed design of the target-transfer equipment was curtailed until a new concept could be established for transferring an intermediate finned tube. The original concept was revised and approved.

5.3.3 Cubicle 1

The process flowsheet and equipment layout for cubicle 1 have not changed. Conceptual designs and design studies were started on the target surface-smear fixture and the dimensional inspection fixture. Detail design of the target receiver which receives the target rods from the above transfer device was started. Other designs for cubicle 1 will be expanded when the target-rod design has been firmly established.

6. DESIGN STUDIES

W. E. Unger

6.1 Design of "Hot" Development Facility, Cells 3 and 4, Building 4507

F. L. Peishel P. L. Robertson
E. M. Shuford W. R. Whitson

The design of all equipment and accessories for cell 4, Building 4507, is complete except for adaptation of a UCRL remote sampling device to the equipment in the cell and various minor items. The plan and elevation of the final layout are shown in Figs. 20 and 21.

Modifications to the bearing design of the mixer-settlers is required because of the failure of the graphite-ceramic combination during cold testing. The modified units will use ball bearings on the impeller shafts until further work can be done to improve the more corrosion-resistant system.

The installation of cell piping and the building of equipment racks is proceeding at an accelerated rate to enable an early "cold" startup (target date: May 10, 1963). Except for the sample-removal cubicle, the major components of the makeup area, operating area, and the roof area are installed or fabricated and awaiting installation.

All equipment to be tantalum plated was prepared for transfer to Parma, Ohio. Tests conducted at ORNL and Parma indicated the cause of the failure of tantalum to bond to Hastelloy C. It is attributable to

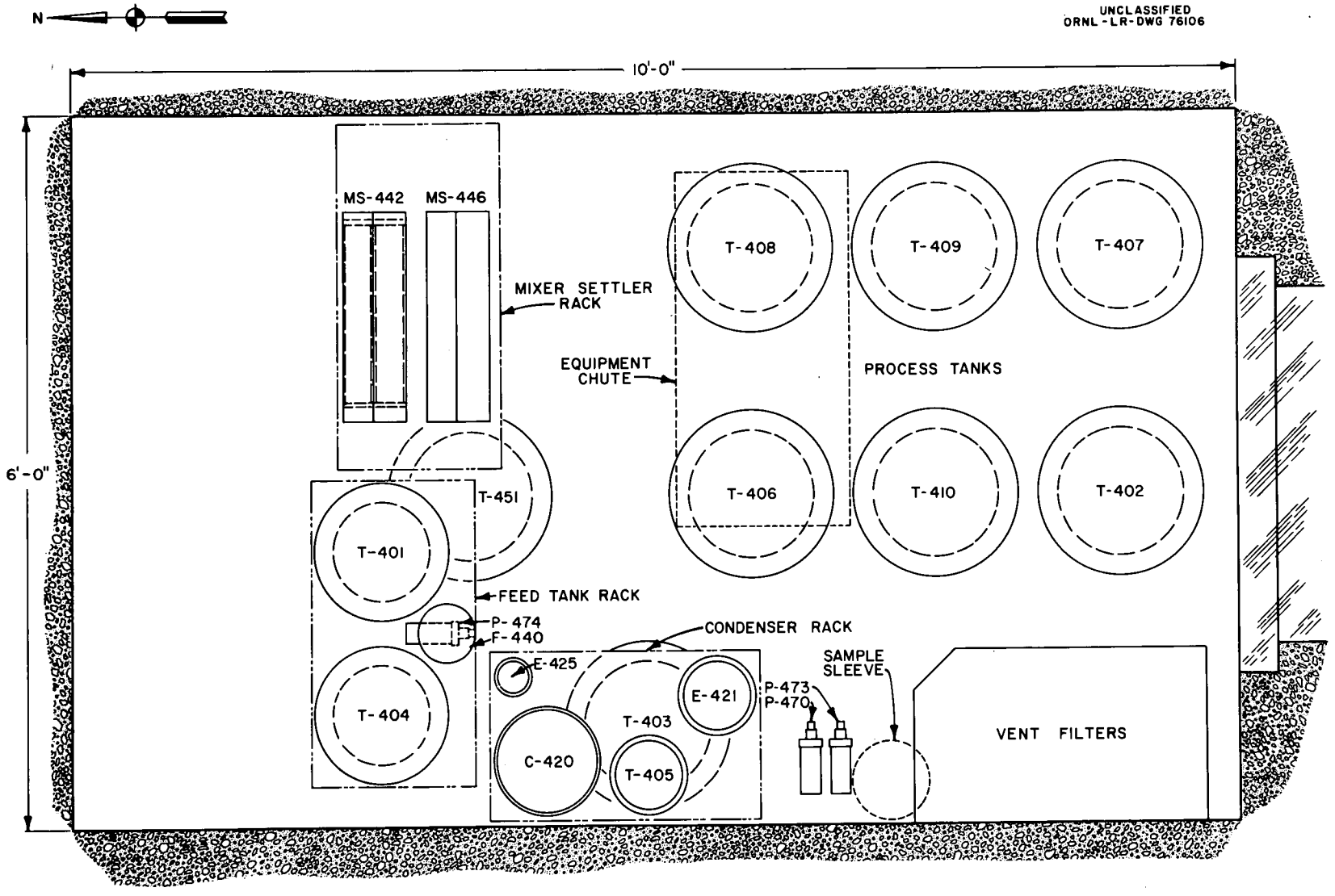


Fig. 20. Cell Plan.

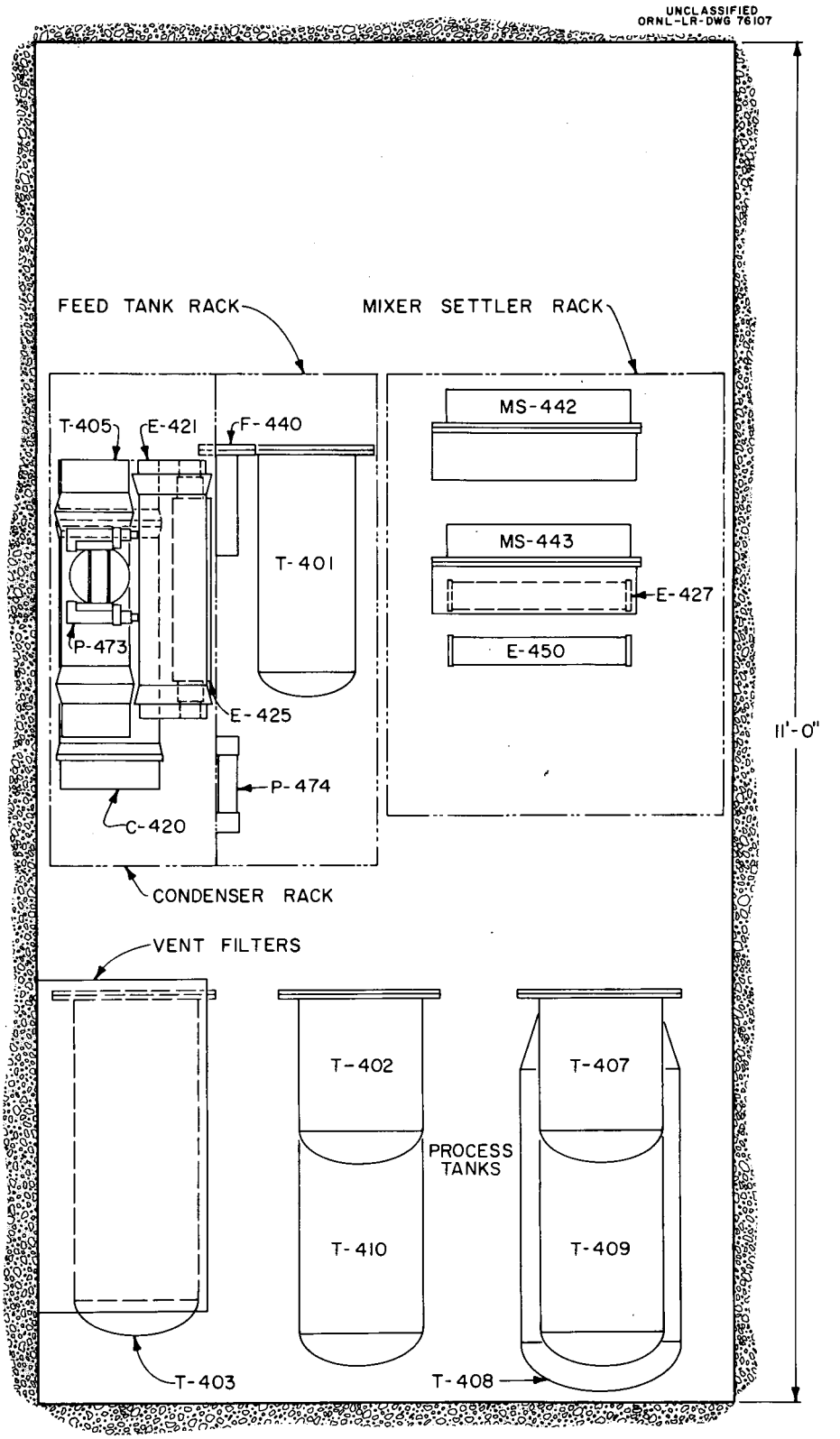


Fig. 21. Cell Elevation.

the brittleness of the intermetallics of tantalum and nickel formed at the interface. Parma Laboratory proposes the electrodeposition of 0.0002 to 0.0005 in. of copper to the Hastelloy C before tantalum plating, thereby providing a diffusion barrier. Preliminary corrosion information at ORNL indicates that the copper does not accelerate corrosion rates of the base material. Parma Laboratory was instructed to proceed with application of the copper flash.

Tests are being conducted with tantalum disconnects to further improve their reliability. Burnishing the sealing surfaces produced a satisfactory joint which remains leaktight after several makes and breaks.

The americium-curium rare earth solution, remaining from processing the burnup sample TRU rods in cell 1 of Building 4507, is now stored in tank T-417 in cell 4.

In early February 1963, a critical-path analysis of the remaining work to be done in Building 4507 (cell 4) indicated that a start date for cold operations of May 10, 1963, might be achieved.

A subsequent updating based on firmer shop estimates showed that unless the in-cell piping and the mixer-settler-rack piping activity could be accelerated, the cold startup date would be extended. Efforts are under way to reduce these times.

6.2 Design of Chemical Processing Equipment

W. D. Burch O. O. Yarbro

6.2.1 Flowsheets

The engineering flowsheets were completed for check-print distribution. Excessive crowding on two of the flowsheets made it necessary for these to be redrawn and split into four; a total of 15 engineering flowsheets thus result. Refinements incorporated into these flowsheets must be reflected by rather significant revisions to the previously issued equipment flowsheets.

6.2.2 Detailed Design at ORGDP

Progress on the design of the chemical processing equipment has accelerated with the start of detailed design at ORGDP in January. The initial group of four engineers and two draftsmen will be augmented by four or five additional draftsmen as soon as they can be efficiently utilized. Work is in progress in the following areas:

1. Equipment Rack Mockup Tests: Alternate designs for the service-line manifold connections on the side of the equipment rack and the process jumper lines to the hot disconnect well are completed. Mockups will be fabricated and tested in order to specify the best design.

2. In-Cell Sampler: Design of a 12-station in-cell sampler is 75% complete. A mockup will be constructed for testing critical items such as sample-bottle elevating mechanism, needle block replacement and other maintenance problems.

3. Hot-Disconnect-Well Track Arrangement: The concept for replacing the hot disconnect well by lowering it onto tracks installed below the cubicle and removing it into the tank pit[9] has been detailed. Clearances are small but appear to be adequate. A mockup will be built to test the removal procedure.

6.2.3 Ion Exchange Column Mockup

An ion exchange column mockup was designed and built (Fig. 22) of glass with stainless steel internals to test (1) resin loading and unloading and (2) resin retention during upflow loading. Resin is loaded as a dilute slurry, the resin being retained between 100-mesh screens as the excess water flows to waste. Upflow loading is desirable to minimize gassing problems, so provisions were made to retain the bed through use of a movable top screen. The bed is held down by applying hydraulic force to a small cylinder attached to the screen. It is hoped that by making clearances small (less than 0.010 in.) between the column and upper screen support, no flexible gasket or wiper will be required to prevent resin from being washed by the screen. The O-ring seals on

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[9] D. E. Ferguson, TRU Quart. Progr. Rept. Feb. 28, 1962, ORNL-3290, p 58.

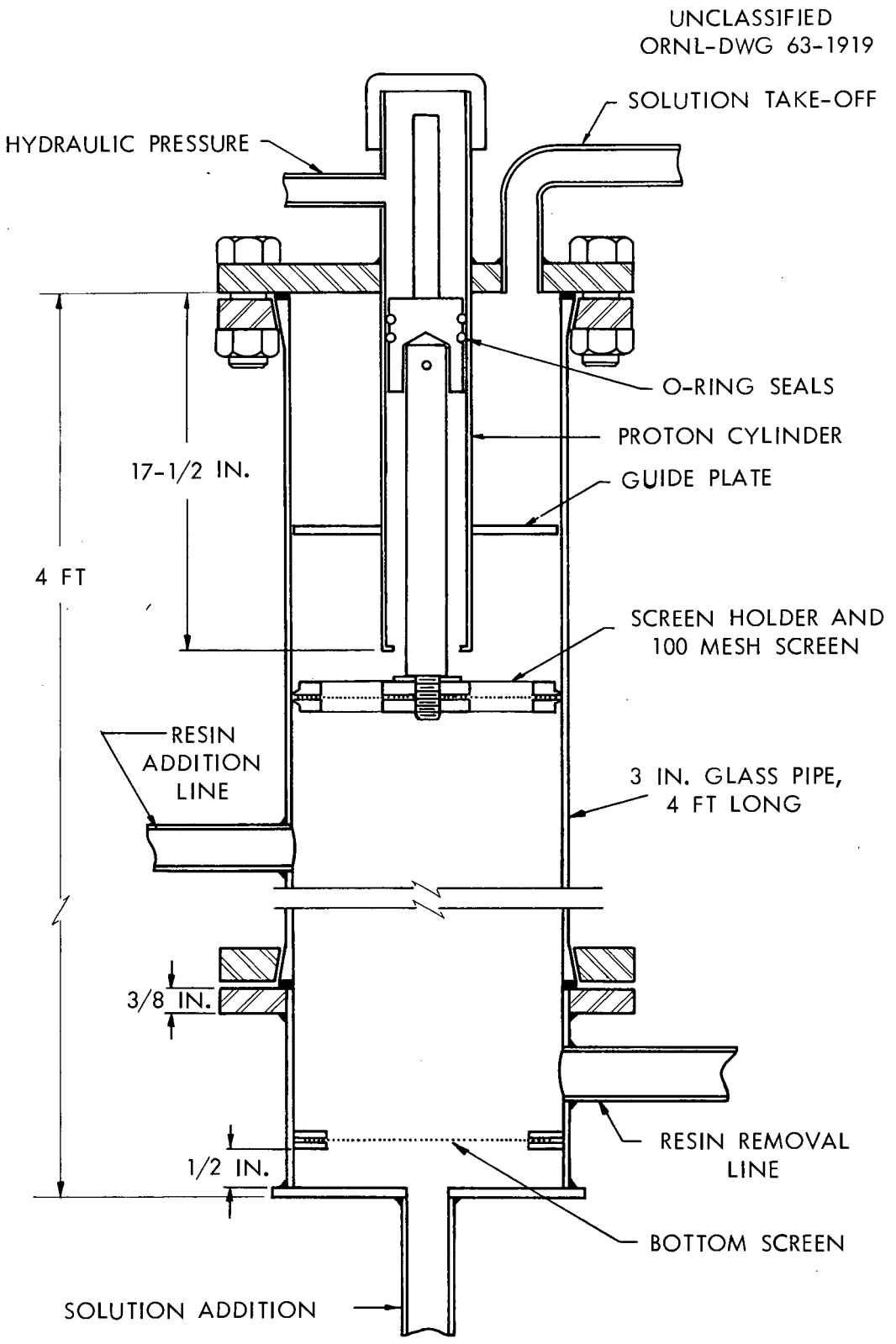

Fig. 22. Mockup of Ion Exchange Column.

the hydraulic piston will be designed to be replaced remotely if the other features appear workable. Removal of the resin will be accomplished by slurrying the resin with water from outside the cell. Water will be introduced both at the top and bottom of the column.

6.2.4 Material Selection for Process Tubing

Information obtained recently on the corrosion of Zircaloy-2 and on the relative cost of Hastelloy C and Zircaloy-2 show that a more corrosion-resistant piping system can be installed, probably at a cheaper total cost, if the piping system is built entirely of Zircaloy-2, rather than a combined Zircaloy-2--Hastelloy C system. In general, low-temperature corrosion rates on Zircaloy-2 are 10 times lower than corrosion rates for Hastelloy C. Tubing raw material costs based on recent firm-price quotations have been estimated at $48,000 for the combined system and $55,000 for the all-Zircaloy-2 system, but substantial savings in installation costs will result through the elimination of one special material from the job. The figures quoted do not include costs for tantalum and stainless steel, which will add $10,000 to $15,000 to the total. Based on this analysis, the requisition for $28,000 worth of Hastelloy C tubing was cancelled prior to placing a firm purchase order.

6.3 TRU Processing Plant Building Design

G. B. Berry B. F. Bottenfield E. J. Breeding
F. L. Hannon W. L. Morgan

6.3.1 Building Design

The complete set of drawings and specifications for the lump-sum contract bid package for the TRU plant building was reviewed and approved by ORNL and the AEC-ORO, and the package was delivered to Oak Ridge for reproduction and issue for bids, estimated to take place about mid-March. The final package contained 69 contract specification sections and 391 contract drawings, in addition to 3 specifications and 32 drawings for information only.

Purchase requisitions were issued for all items to be procured by ORNL for the lump-sum contractor. The present estimates of deliveries

indicate that all materials will be on hand for the contractor as required to maintain his construction schedule.

The procurement of additional items of equipment, such as the intercell conveyor, the waste-tank-pit processing equipment and waste header, and the vessel off-gas scrubber and header will be initiated during the next quarter.

6.3.2 Mechanical Equipment Design

Intercell Conveyor. -- Design of all intercell conveyor components except the valve panel, canister, and control station was completed, and the drawings were issued for approval.

Drawings of the conveyor housing were revised to incorporate broader tolerances where possible in order to simplify fabrication and installation.

Equipment Transfer Case. -- Detailed design of all components of the equipment transfer case was completed except for control components, which have been issued for approval.

Fabrication of top and bottom assembly components are about 90% complete.

Cell Cubicle. -- Drawings of the floor pans were revised to incorporate broader tolerances where possible to simplify fabrication, and the horizontal overall dimensions were reduced to facilitate installation.

Drawings of the back walls were revised to provide cooling system penetrations in all cubicles and removable panels for access to cubicles in cells 8 and 9.

Carrier Charging Mechanism. -- Preliminary design was recently initiated on components to be installed beneath the transfer-area cubicle floor pan to accomplish cubicle-to-carrier transfer.

6.4 Critical Path Schedule Analysis

B. N. Robards

New manpower leveling for ORNL design was completed in January 1963. The results of this leveling were incorporated into the TRU Master Schedule

to prevent unrealistic start times from being imposed on the design and subsequent activities and to obtain a more accurate picture of the available float.

An ORNL Title II design "S" curve, Fig. 23, depicting the projected percentage completion of Title II design as a function of time in project days was obtained from the leveled master schedule. This curve is being used in a monthly evaluation of ORNL Title II design progress.

The first monthly review of the TRU project was held February 8. These meetings are intended to present to project management and other interested personnel the status of ORNL participation on the project. Each phase of the project was reviewed, with emphasis on particular trouble areas and immediate needs.

The building construction start date is now June 10, 1963, indicating a building completion date of February 14, 1965. The project critical path, however, is still governed by ORNL procurement of the cell-shielding-window sleeves. The required delivery date of the window sleeves is January 20, 1964. This leaves a period of 10-1/2 months as of March 1, 1963, in which ORNL must select the vendor, approve the design, validate the mockup tests, and deliver the window sleeves to the building contractor.

7. ANALYTICAL DEVELOPMENT

L. T. Corbin J. C. White

7.1 Analytical Radiochemical Methods Development

F. L. Moore

During the period just ended, nine methods were written for the TRU section of the ORNL Master Analytical Manual to be used in various analytical laboratories. These are:

Gross alpha, direct method

Gross alpha, LaF_3 method

Gross alpha, amine extraction method

Plutonium alpha, LaF_3 method

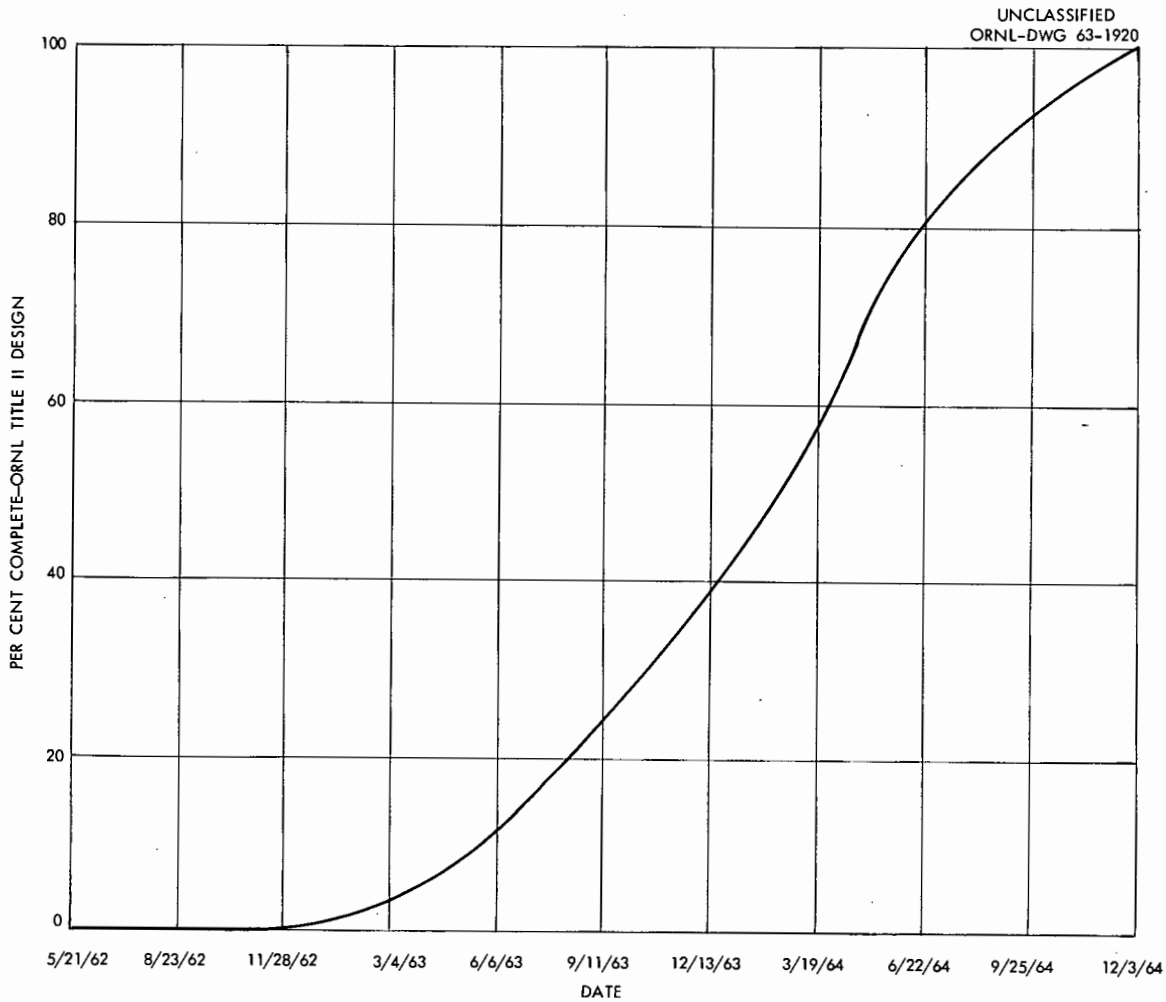

Fig. 23. Title II Design Progress Report.

Plutonium alpha, TTA method

Transplutonium alpha, LaF_3 method

Transplutonium alpha, amine extraction method

Alpha spectrometry, silicon diode detector

Americium, oxidation method

Several approaches are under investigation for the direct instrumental determination of californium isotopes involving alpha spectrometry and fission counting.

Some time has been spent in the recovery and purification of Bk^{249} and Cf^{252} isotopes from analytical wastes in order to obtain tracers for further studies.

Work is near completion on a new method for Cs^{137}, one of the major fission products to be determined in the TRU program. The method results from recent developments for inhibiting the alkaline hydrolysis of TTA. Highly-selective, essentially quantitative extraction of Cs^{137} is possible with TTA dissolved in solvents containing the nitro group. Small amounts of lithium markedly enhance the extraction. This method is potentially useful for the preliminary remote isolation of Cs^{137} from TRU solutions, prior to bench work or radioactivity measurements. Details will appear in a forthcoming ORNL-CF memo.

7.2 Absorption Spectrophotometric Studies

D. A. Costanzo

A computer program was written which makes possible the rapid analysis of spectrophotometric data for multicomponent systems that are not at equilibrium. The spectrophotometric technique was employed to follow the course of the reactions for the simultaneous disproportionation and polymerization of ionic Pu^{4+}. The program was utilized to determine the exact concentrations of ionic Pu^{3+}, Pu^{4+} and Pu^{6+}, as well as the polymerized Pu^{4+} in the reacting mixture. The results obtained for three- and four-component systems of known kinetics indicate an error of the method of within $\pm 1\%$.

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