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TRANSURANIUM QUARTERLY PROGRESS REPORT

FOR PERIOD ENDING NOVEMBER 30, 1962

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

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METALS AND CERAMICS DIVISION
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REACTOR CHEMISTRY DIVISION
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TRANSURANIUM QUARTERLY PROGRESS REPORT FOR PERIOD ENDING NOVEMBER 30, 1962

Compiled by
D. E. Ferguson

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OAK RIDGE NATIONAL LABORATORY
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TRANSURANIUM QUARTERLY PROGRESS REPORT FOR PERIOD ENDING NOVEMBER 30, 1962

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 are reported here.

Process Development. -- Satisfactory results have been obtained in laboratory tests of the Tramex process for separating transplutonium elements from fission products, corrosion products, and materials used in target fabrication. This process is useful for recovering all known transplutonium elements as a group from irradiated HFIR targets and also for isolating Cm^{242} plus Am^{241} from irradiated cermets of AmO_2 and aluminum. The only contaminants investigated that follow the transplutonium elements through the process are nickel, Ag^{111}, and possibly Te^{132}. Nickel, which may come from the aluminum alloy used in targets or from corrosion of process equipment, can be separated from the Tramex product by precipitating the oxalates of the heavy elements from solutions low in acid and containing ammonium ions, or by hydroxide precipitation of the heavy elements by hydrolysis of urea. Tests with rare earths as substitutes for the heavy elements gave better than 99% recovery, with nickel decontamination factors of about 300. For most processing, Ag^{111} and Te^{132} will not be a problem since they have such short half-lives.

The recommended conditions for the Tramex process were modified in three respects: (1) aluminum chloride has been added to the feed to accommodate aluminum used as target material, (2) more-concentrated hydrochloric acid has been substituted for the stripping solution to improve zirconium, niobium, and iron decontamination, and (3) a scrub of freshly prepared Alamine 336·HNO_2 has been added to improve ruthenium decontamination. Ruthenium decontamination factors in laboratory tests at these conditions were greater than 10^4. The addition of H_2O_2 to stripping

solutions was not effective in oxidizing ruthenium to an extractable form, and decontamination factors of only 10^3 were obtained. Zirconium, niobium, and rare earth decontamination factors reported last quarter were $\geqq 10^4$.

Results of testing the Tramex process in a batch countercurrent extraction at a Cm^{242} concentration equivalent to about 4 w/liter indicate no difficulties because of the high activity level. Distribution coefficients were in agreement with tracer-level studies, and phase separation was as good as or better than in the absence of activity. Since only two extraction stages and one strip stage were used, it was not possible to demonstrate complete recovery and decontamination in this test. However, all indications are that the Tramex process is satisfactory for activity levels up to at least 10 w/liter, which is the maximum level anticipated for the Transuranium Processing Plant.

Radiation stability of the solvent was confirmed in a batch test in which 0.6 $\underline{M}$ Alamine 336·HCl-DEB was in contact and intermittently mixed with 10.5 $\underline{M}$ LiCl containing Cm^{242} for a total exposure of about 300 whr/liter. The most notable effect was a 30% decrease in the curium distribution coefficient, which was still great enough for a satisfactory process, and no phase separation difficulties were encountered. Loss of acid in Tramex feed by radiolysis at high activity level poses a potentially serious problem. An acid-destruction G value of 5.6 molecules per 100 ev was measured. Unless methods of preventing acid loss by radiolysis can be found, it will be necessary to add make-up acid to prevent precipitation of hydroxides.

Stable Tramex feeds of low acidity were prepared on a 1- to 2-liter scale in laboratory glassware by simply distilling off excess acid and water. Further tests are needed when plant equipment becomes available to determine whether the configuration of the equipment is important.

Densities, boiling points, and refractive indices have been determined for solutions of 9, 10, and 11 $\underline{M}$ LiCl containing $AlCl_3$.

A 2.5-g sample of Am^{241} contaminated with lanthanum, cerium, iron, and calcium was purified by precipitating $KAmO_2CO_3$ from 3 $\underline{M}$ K_2CO_3. The precipitate contained 99.6% of the americium, free of all contaminants except cerium. This method can be used to separate americium from curium, which will remain in solution.

By using -270 mesh resin and by pressurizing the column, gassing caused by high alpha activity levels was controlled well enough to accomplish satisfactory americium-curium separation by ion exchange at activity levels equal to 0.4 to 1 mg of Cf^{252}. Elution from cation exchange resin with ammonium alpha-hydroxyisobutyrate gave a product containing 96% of the curium free of americium, and elution from anion exchange resin with 4 M $LiNO_3$ gave a product containing 99% of the curium free of americium. These methods may be useful in separating einsteinium and fermium from milligram quantities of californium.

Americium and curium sorption on anion exchange resin from 2.6 M $Al(NO_3)_3$ is adversely affected by an increase in rare earth concentration. This was tested with europium as a stand-in for curium and americium in runs at standard testing conditions. At a feed flow rate of 2 ml cm^{-2} min^{-1}, europium loading decreased from 89 to 40% as the rare earth concentration increased from 0.5 to 5.0 g/liter. This effect can be at least partially off-set by decreasing the flow rate for higher rare earth concentrations. Europium loading from feed containing 2 g/liter rare earths was increased from 62 to 89% by decreasing the flow rate from 2 to 1 ml cm^{-2} min^{-1}. It is advantageous to use acid-deficient aluminum nitrate feeds since they are more stable and since europium recovery was not affected for acid deficiencies up to 0.7 N in 2.6 M Al^{3+} solutions.

Attempts to determine the cerium loading capacity for three mesh sizes of Dowex 1-10X resin were unsuccessful since equilibrium was not attained in 96 hr at 25°C. Resin loading increased with decreasing particle size, and the approach to equilibrium was more rapid with the finer resin.

There was no indication of resin damage when 2 g of Dowex 1-10X resin was in contact for 21 days with 4 ml of 2.6 M $Al(NO_3)_3$ containing 3.14×10^8 gross alpha counts min^{-1} ml^{-1} (mostly Cm^{242}) and 1.68×10^{10} gross gamma counts min^{-1} ml^{-1} (mixed fission products).

Due to anticipated difficulty in the nitric acid dissolution of highly irradiated plutonium-aluminum alloy rods for the TRU program, a process based on the caustic dissolution of aluminum was investigated. Aluminum is readily dissolved in a solution containing 1.2 moles of NaOH and 1.55 moles of $NaNO_3$ per mole of aluminum, and the rate appears to be independent of the silicon content of the aluminum over the range 0.1 to 3.0%. By acidification with nitric acid to dissolve the plutonium, feed for plutonium recovery can be adjusted to 2.5 $\underline{M}$ $NaNO_3$, 0.8 $\underline{M}$ $Al(NO_3)_3$, and 2.6 $\underline{M}$ HNO_3. Although plutonium recovery by anion exchange is apparently satisfactory, scouting tests indicated that this amount of $NaNO_3$ will cause difficulty in the anion exchange recovery of americium and curium. First, it was not possible to adjust the feed solution by a simple distillation because $NaNO_3$ precipitated before any appreciable amount of acid was distilled off, and, second, resin loading, as tested with europium tracer, was adversely affected since $NaNO_3$ is much less effective in forming a sorbable complex. Satisfactory feed adjustment and resin loading were obtained for aluminum-to-sodium-plus-potassium mole ratios of 0.8 or greater.

The system, KNO_3-$NaNO_3$-$Al(NO_3)_3$-H_2O was investigated since a point of maximum nitrate solubility (10.4 $\underline{N}$ at 25°C) occurs at an Na:K:Al mole ratio of about 1:1:2.1.

The extraction of americium, curium, and rare earths into tertiary amine from neutral nitrate solution is an attractive alternate to the anion exchange method. In a laboratory-scale mixer-settler test in which the extractant was 0.6 $\underline{M}$ Alamine 336·HNO_3, the scrub was 8 $\underline{M}$ $LiNO_3$, and the feed was 2 $\underline{M}$ $Al(NO_3)_3$ containing 5 g/liter rare earths and 1 g/liter each of iron, nickel, and chromium, rare earth losses were 0.005%, and iron, nickel, and chromium extraction was undetectable. The aluminum decontamination factor was 300.

Curium distribution coefficients into 0.6 $\underline{M}$ Alamine 336·HNO_3-DEB from 6 $\underline{N}$ NO_3^- solutions varied from 0.051 for 6 $\underline{M}$ NH_4NO_3 to 5.4 for 6 $\underline{N}$ $Be(NO_3)_2$. This variation appears to be associated with the ability of the smaller ions to tie up larger amounts of water by hydration.

When different diluents were used for the extractant, americium distribution coefficients varied from 0.53 to 38, and americium-curium separation factors varied from 1.23 to 2.06 for the system 0.6 M Alamine 336·HNO_3 vs 3 M $LiNO_3$-1 M $Al(NO_3)_3$. Based on the viscosity of 5.4 centipoise and distribution coefficients of 8.0 for americium and 5.3 for curium, diethylbenzene is the recommended diluent.

Performance tests demonstrated the reliability of a process-line disconnect when subjected to misalignment forces, corrosive solutions, rough handling, interchange of mating parts, and other conditions simulating rough service. Socket extensions machined to limit the torque were tested with an electric impact wrench and were recommended for use in TRU cubicles to permit the use of one impact wrench for various bolt sizes.

Batch rate tests for scrubbing europium from 30% Alamine 336 in diethylbenzene by 9 or 11 M LiCl solutions show that satisfactory operation should be possible within the normal power input range for pulse columns. Tests in a glass pulse column (3/4 in. in inner diameter and 24 in. long) show average HETS of 24 in., with no marked trends for pulse conditions, column configurations, or other variables.

Calculations indicate that the irradiation of prototype HFIR targets in the MTR should be at stepped neutron fluxes between 1.8 and 4.4×10^{14} for 440 days to give 6.4×10^{21} neutrons/cm^2.

From results of exposure of protective coatings to Co^{60} gamma radiation in air and water, maximum recommended exposures are as high as 3×10^9 rads in air or water, with some tests not yet completed.

HFIR Target-Fabrication-Process Development. -- Experiments are in progress to resolve some of the difficulties encountered in preparing and encapsulating pellets. It has been demonstrated that the major obstacle to using conventional blending and pressing techniques for obtaining the desired oxide distribution through the matrix is the large difference in size between the oxide and metal powders being used.

A new end-closure design is being considered; it will utilize welding for leaktightness and either brazing or mechanical means for obtaining the necessary joint strength.

The "Magneform" equipment offers some advantages over hydraulic pressure for the collapse of the capsule around the pellets.

Cubicle 3 in the Transuranium Facility contains the primary fabrication equipment. Detail design is virtually complete on the various items for this cell. About half the equipment has been constructed and is under test.

Equipment for a plutonium target fabrication line was designed and is being purchased or is under construction.

As a result of several unanticipated problems encountered in attempting to fabricate capsules containing plutonium pellets under glove-box conditions, no irradiation tests are under way. Rescheduling these experiments awaits the solution of the difficulties encountered.

Design Studies. -- Title II design of the TRU Processing Plant is complete except for final drawing and specification review, scheduled for January 1963. Procurement by ORNL of special items such as all-Hastelloy-C material for the building contractor has begun. Building construction is scheduled to start in May 1963. Design of the building mechanical equipment such as the intercell conveyor, equipment transfer system, and cell cubicles is proceeding on schedule.

Initial drafts of the five equipment and 13 engineering flowsheets required for the TRU chemical processing equipment were completed. Conceptual-design information is being assembled in preparation for the detailed design of this equipment scheduled to begin about January 1 by a design group at K-25. Included are the dissolver and dissolver control system and all components for a solvent-extraction-cycle equipment rack, to be fabricated of Zircaloy-2 during the next six months.

The initial manpower-leveling studies, using Critical Path Scheduling techniques, were completed in September. From these studies, working schedules for the various ORNL design groups have been set up. About 46% of the total of 5600 man-days of final or Title II design is scheduled to be completed in FY-1963.

Design of the hot development equipment to be installed in cell 4 of Building 4507 is 95% complete. Installation is scheduled to be completed by March 1, 1963. Development directed towards a simple, remotely operated line disconnect for use both in the development cell and the final facility has produced a cast disconnect clamp that accepts any size of line up to 3/4-in. tubing. Disconnect sealing surfaces were successfully formed both by machining and by direct rolling and swaging of the process tubing, although galling problems associated with tantalum disconnects have not been entirely solved.

TRU Corrosion Studies. -- The sensitivity of tantalum and tantalum-plated specimens to hydrogen pickup and corrosion in a number of TRU-related environments was studied. This included solution and vapor environments. Corrosion rates were negligible, except for the tantalum-plated specimens, which picked up large amounts of hydrogen due to the complete dissolution of the stainless-steel base metal. Pickup by all-tantalum specimens amounted to not more than 3 ppm, but when tantalum was coupled with type 1100 aluminum in 6 M HCl at 105°C, the pickup rose to 64 ppm in 7 hr.

For welded and unwelded Hastelloy C, the studies of the effect of added oxygen or hydrogen peroxide to 6 M HCl were completed. The tests were made at 35°C and ran for 1000 hr, with replacement of the solution every 100 hr. Specimens were exposed to both the solution and the vapor.

After 1000 hr, corrosion rates were 0.7, 5.2, and 4.0 mils/month, respectively, for the no-added oxidant, oxygen-aeration, and peroxide addition tests. All specimens were preferentially attacked in the heat-affected zones adjacent to the welds, with the most serious attack taking place in the oxygen-aeration tests. The measured depth of penetration ranged from 28 to 32 mils. The unwelded specimens exposed to vapors corroded much the same as they did when exposed to the solutions in the no-added-oxidant and peroxide-addition tests: 1.5 and 1.7 mils/month, respectively. However, the specimen in the oxygen-aeration test had a higher rate: 3.7 mils/month.

Homalite CR-39, Homalite CR-100, and Polypenco K-51 were exposed for 1000 hr at 80°C in a number of TRU-related organic processing solutions. These tests are not completed, and good resistance was shown by the materials, with dimensional and weight changes being only minor.

Analytical Methods Development. -- A specific method was developed for the separation of americium from Cm, Bk, Cf, Pu, U, Th, Cs, Sr, Zr, Nb, Al, and the lanthanides. In essence, the method is based on the oxidation of trivalent americium to the sexavalent americyl (AmO_2^{2+}) ion, the complexing of the ion with fluoride, and the scavenging of the impurities noted above with lanthanum fluoride.

Computer methods are being investigated in order to make possible the analysis of the spectrophotometric absorption spectra of solutions that contain more than one absorbing species, any or all of which may not be at equilibrium. A detailed description of the problem was written and submitted to the Mathematics Panel to be programed for the IBM 7090 computer.

Six dissolution samples of irradiated americium were analyzed according to a method recently developed here. The results agreed well with those obtained by older methods-- ± 5%.

The fission yields of americium were studied radiochemically. The results differed slightly from the published data, with masses 89 and 91 being higher by nearly 20%, and masses 140 and 141 being lower by almost 10%.

Preliminary calculations of the fission yields of americium were made in order to facilitate the development of radiochemical analytical methods to be applied to irradiated TRU fuel rods. These results are shown in a table.

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 Facility, 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 Design Group of the Oak Ridge Gaseous Diffusion Plant.

The previous report in this series is ORNL-3375.

3. PROCESS DEVELOPMENT

3.1 Chemical Development

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3.1.1 Development of the Tramex Process

Investigations have continued on developing, testing, and improving the Tramex process for separating transplutonium elements from fission products, corrosion products, and materials used in target fabrication. This process is useful for recovering all known transplutonium elements as a group from irradiated HFIR targets and also for isolating Cm^{242} (plus the remaining Am^{241}) from irradiated cermets of americium dioxide and aluminum.

Process conditions presently recommended for maximum decontamination are shown in Fig. 3.1. These differ in three respects from the conditions previously given:[1] (1) Aluminum chloride is added to the feed, (2) More concentrated hydrochloric acid is used for stripping, and (3) The strip product is scrubbed with freshly prepared Alamine 336·HNO_2 in diethylbenzene (DEB) diluent. Therefore, aluminum chloride was added to the feed in order to simulate the aluminum chloride concentration of an actual solution. (Small amounts of aluminum chloride are beneficial since, by hydrolysis, they help replenish small losses of acid.) The increased acidity of the stripping solution and the solvent scrub are used to improve decontamination. The higher acidity is effective in keeping zirconium, niobium, and iron in the solvent phase, and the use of HNO_2 in the solvent scrub forces ruthenium to remain in the solvent. Results of tests indicate that this process is satisfactory for isolating the transplutonium elements from all probable contaminants except the nickel, which comes from the aluminum alloy used in irradiation targets and from the corrosion of process equipment. Additional demonstration at full activity levels and at plant scale are required to firmly establish the capabilities of this process.

Investigations during this report period included high-activity-level tests of the Tramex process with Cm^{242}, behavior of short-lived fission products and ruthenium, effects of alpha radiation on solvent and feed solution, evaluation of alternate solvents, methods of feed adjustment, and determination of physical properties of Tramex solutions, all discussed below.

Tramex Tests at High Activity Levels

The results of testing the Tramex process in a batch countercurrent extraction at a Cm^{242} concentration equivalent to about 4 watts/liter

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[1] D. E. Ferguson, Transuranium Quarterly Progress Report for Period Ending February 28, 1962, ORNL-3290, p 4 (June 6, 1962).

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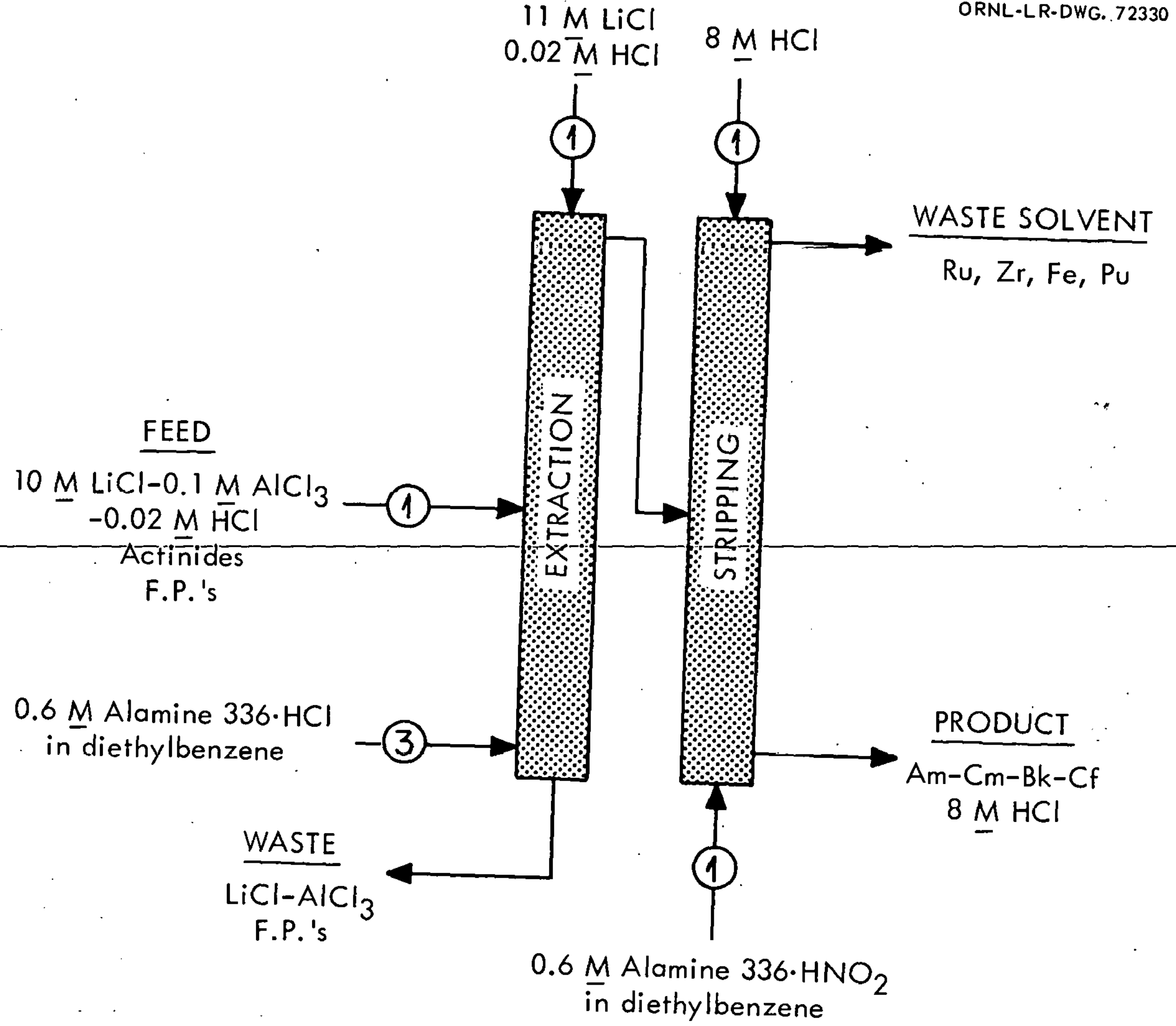

Fig. 3.1. Flowsheet for the Tramex Process.

indicate no difficulties because of the high activity level. Distribution coefficients were in agreement with tracer-level studies, and phase separation was as good as or better than in the absence of radioactivity. Because of mechanical difficulties, it was not possible to operate the scrubbing and stripping contactors as planned. Two extraction stages were used, and batch stripping was accomplished by shaking the solvent with strip solution in sample bottles. None of the operating difficulties was due in any way to the activity level of the solutions. Although it was not possible to demonstrate complete recovery and decontamination in this test, the data and operating experience indicate that the Tramex process will be satisfactory at high activity levels.

Process Description. -- The Tramex flowsheet was modified for this test as shown in Fig. 3.2. An aluminum-americium dioxide cermet target containing 40.3 mg of Am^{241} and 12.5 g of aluminum metal was prepared and then irradiated in the Oak Ridge Reactor for 39 days at a flux of 2.0×10^{14}. After cooling for two months, the aluminum was dissolved in 2.5 M NaOH. The resulting solution was decanted through a filter, and the residue containing the americium, curium, and fission products was washed with dilute sodium hydroxide and then with dilute lithium hydroxide. The residue was dissolved in warm 6 M HCl. Excess acid was distilled off, and LiCl and $AlCl_3$ were added to adjust the extraction feed to 9.9 M LiCl--0.2 M $AlCl_3$ containing about 4 w/liter of Cm^{242}. Portions (7.8 ml) of this feed were contacted with 9.0-ml volumes of 0.6 M Alamine 336·HCl--DEB in two batch-countercurrent extraction stages. Solvent containing extracted Am, Cm, Ru, and Zr was stripped with 5 to 8 M HCl by collecting each batch of solvent in a sample bottle containing the desired strip solution. These were shaken, and each phase was analyzed to determine stripping results. Some of the 5 M HCl strip solutions contained $NaNO_2$ as an oxidizer for the ruthenium, but no difference was noted from the effect of radiolytic H_2O_2.

Equipment Description. -- The equipment shown in Fig. 3.3 was described previously.[2] The essential features were six countercurrent batch-operated

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[2] D. E. Ferguson, Transuranium Quarterly Progress Report for Period Ending August 31, 1962, ORNL-3375 (Jan. 18, 1963).

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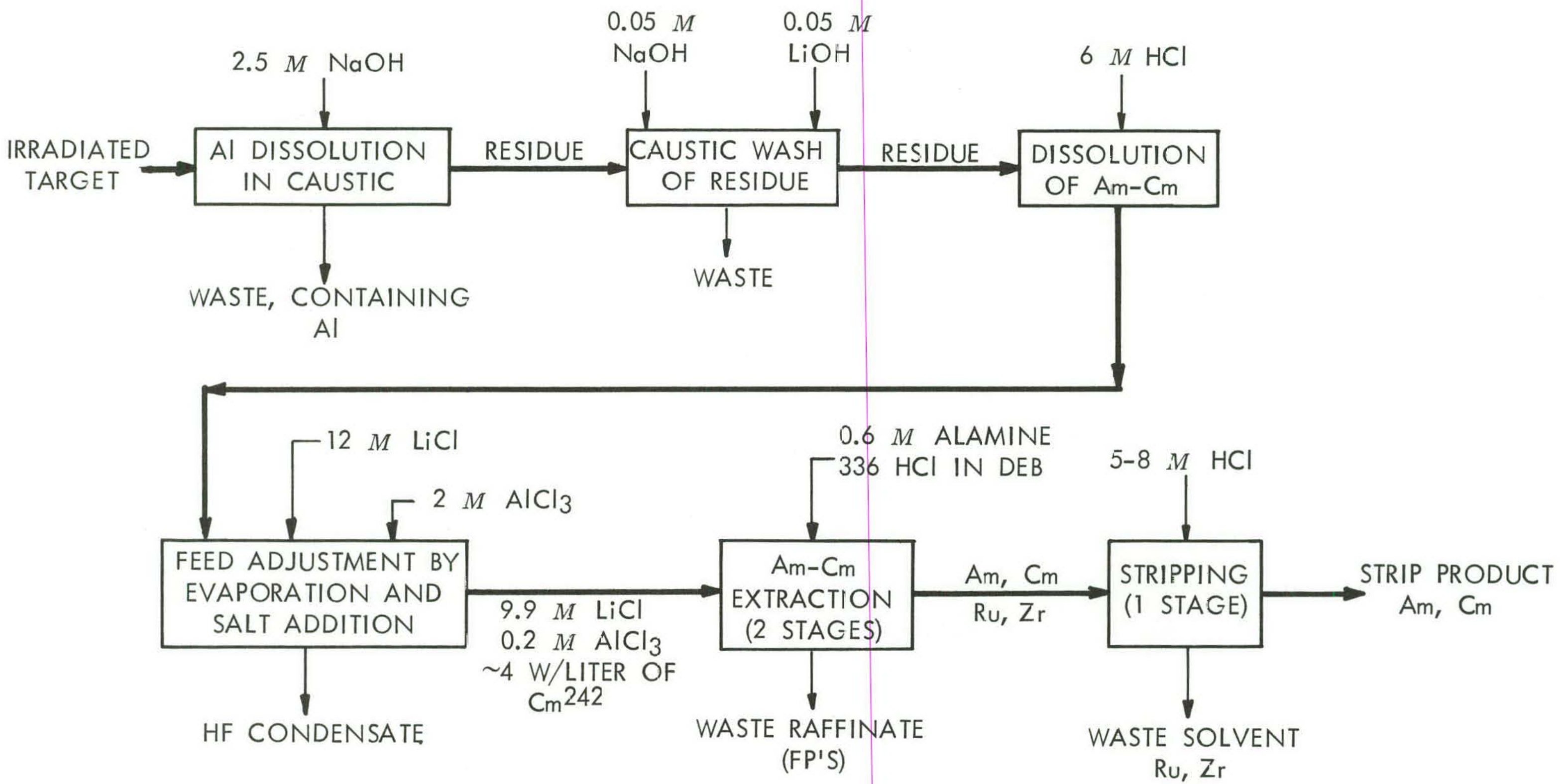

Fig. 3.2. Modified Flowsheet for High-Activity-Level Tramex Test.

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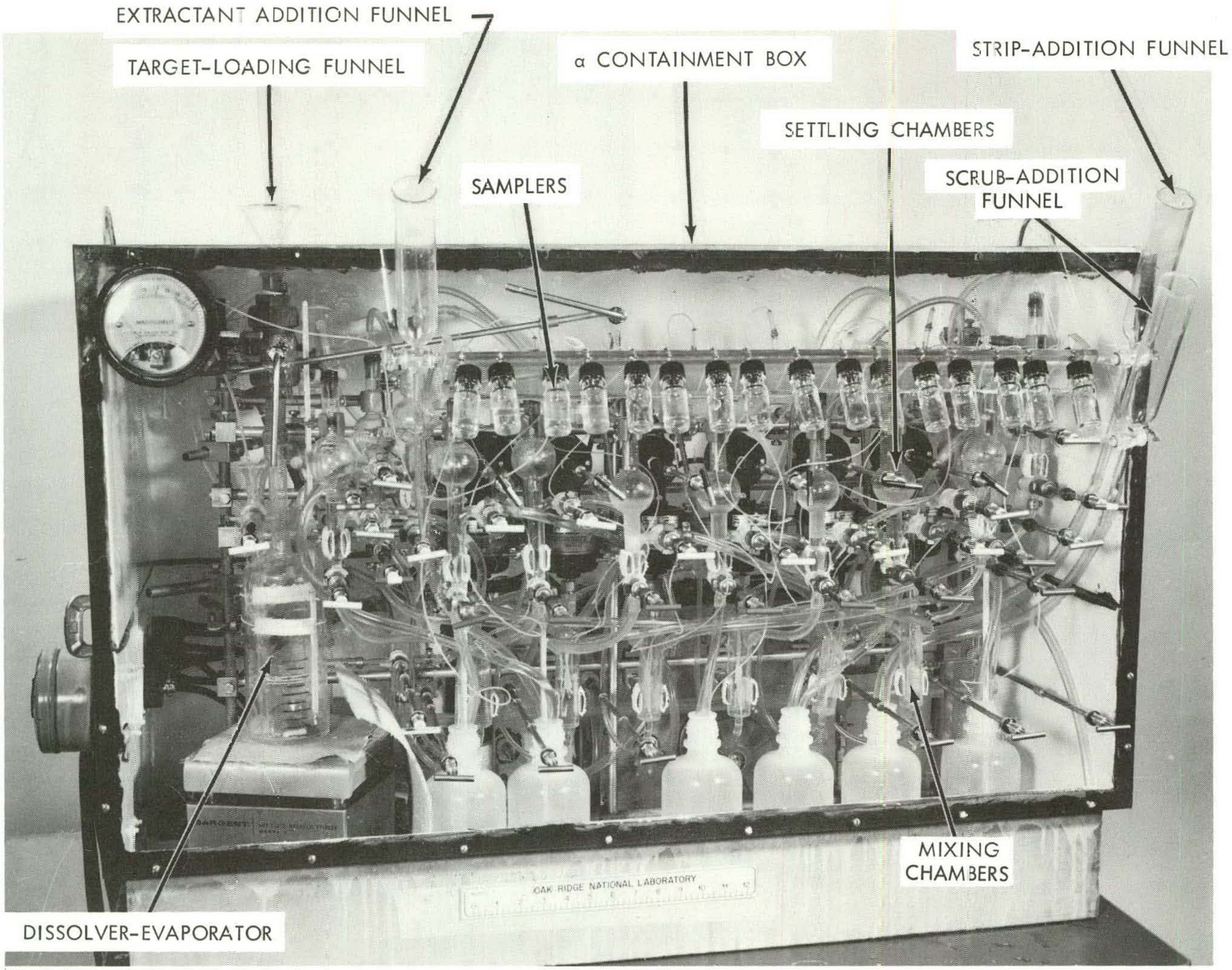

Fig. 3.3. Equipment for High-Activity-Level Tramex Test.

extraction stages, a dissolver-evaporator, and associated equipment for solution transfer, sampling, off-gas handling, and solution waste disposal. Two of the stages were planned for extraction, one for scrubbing, one for stripping, and two for washing the strip product with fresh solvent. The apparatus was mounted in an alpha-tight enclosure and installed in a cell in Building 3019 High Radiation Level Analytical Facility. Operations were performed by using master-slave manipulators.

Operational Experience and Results. -- All operating difficulties encountered were in no way caused by the radioactivity of the solutions. The primary difficulty was caused by the deleterious effect of the solvent on the Tygon tubing used to connect the vessels. Although Tygon is satisfactory when exposed to solvent for two to three weeks, the adjustment and in-cell testing required about six weeks before the high-activity-level run was started. By this time, the transfer line from stage 3 was so swollen and softened that solution could no longer be transferred, making it impossible to use the last four stages. Solution transfer and phase separations in the first two stages were comparable with operations in which there was no radioactivity.

Caustic dissolution of the aluminum jacket and matrix of the target proceeded normally. About 10% of the Cm^{242} was lost in the aluminum dissolver solution and the caustic washes. The residue was readily dissolved in warm 6 $\underline{M}$ HCl. The volume of this solution was 270 ml, and it contained 7.9 mg of Cm^{242}, 18 mg of Am^{241}, and 2.1 mg of fission products (see Table 3.1). Back calculating to the end of the irradiation period, this means that about 30% of the Am^{241} was converted to Cm^{242} and that about 6.2% of the Am^{241} fissioned during the 39-day irradiation in the ORR at a flux of 2 x 10^{14}. The dissolver solution was adjusted to about 200 ml of extraction feed containing about 3.5 mg/liter of Cm^{242}, or about 4 w/liter.

Distribution coefficients (E_a^o) for Cm, Ce, Ru, and Zr in the two extraction stages are given in Table 3.2. In general, these are comparable with values obtained at tracer levels except for Zr distribution coefficients, which are low. Stripping distribution coefficients for Ru and Zr are compared with data obtained by extracting tracers into the solvent (Fig. 3.4). It was noted in the tracer tests that ruthenium distribution coefficients obtained by stripping were about ten times higher than those obtained by extraction.

Table 3.1. Composition of the Dissolver Solution for the High-Activity-Level Tramex Test

The americium-curium was dissolved in 270 ml 6 M HCl and adjusted to 200 ml of extraction feed

Isotope	Quantity	Isotope	Quantity
Cm^{242}	7.9 mg	Ru^{103}	360 mc
Am^{241}	18 mg	Ru^{106}	140 mc
Pu^{238}	3.1 mg	Cs^{137}	1.8 mc
Pu^{242}	2.1 mg	Ba^{140}	290 mc
Fission product	2.1 mg	Ce^{141}	580 mc
Sr^{89}	110 mc	Ce^{144}	85 mc

Material balances over the second extraction stage were: Cm, 99%; Ce, 89%; Ru, 240%; and Zr, 103%. Since only two extraction stages were used and the solvent was not scrubbed, neither complete product recovery nor complete decontamination was demonstrated. The best results were obtained when an 8 M HCl strip was used. Curium recovery was 95%, and the cesium, ruthenium, and zirconium decontamination factors were 12, 17, and 5, respectively. Although these results do not offer conclusive proof of the capabilities of the Tramex process, all indications lead to the conclusion that it will be satisfactory for processing at activity levels up to 10 w/liter.

Table 3.2. Extraction Coefficients in High-Activity-Level Tramex Test

	E_a^o	
Element	Feed Stage	Second Stage
Cm	3.5	5
Ce	0.076	0.31[a]
Ru	13	1.4[b]
Zr	3.5	0.46[b]

[a] These increases may be due to decreased acidity of the feed after one contact with the solvent.

[b] These marked decreases may be due to some inextractable species of these elements.

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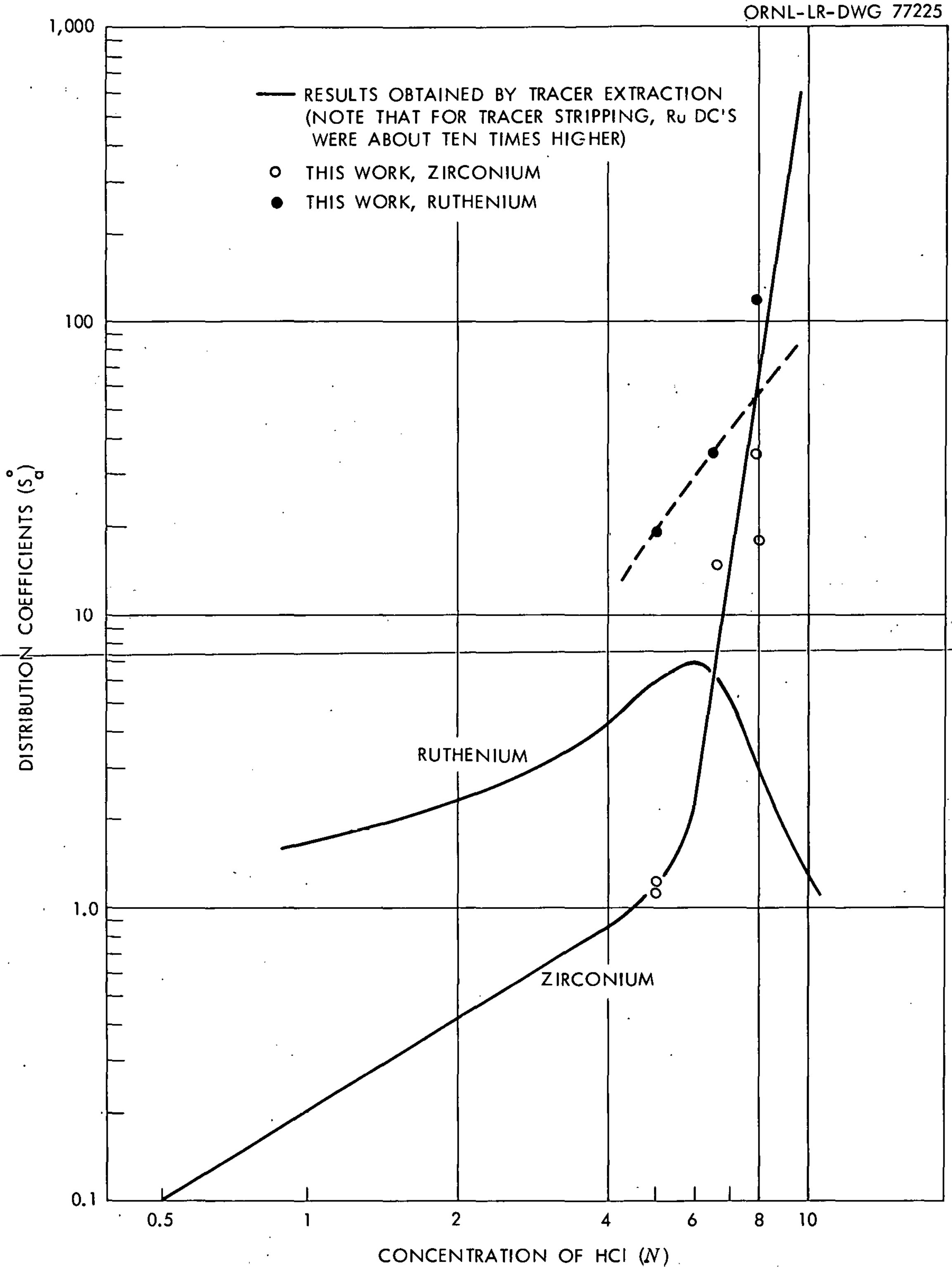

Fig. 3.4. Stripping Coefficients for Ruthenium and Zirconium in the High-Activity-Level Tramex Test.

Behavior of Short-Lived Fission Products in the Tramex Process

The 7.5-day Ag^{111} fission product quantitatively followed americium and curium through extraction and stripping in a laboratory-scale mixer-settler run made with synthetic feed spiked with irradiated Am^{241} cooled for two days. The 78-hr Te^{132} fission product was partially extracted, and a small amount was stripped with the product. Although 67-hr Mo^{99}, 8-day I^{131}, and 2.3-hr I^{132} were extracted, they remained in the solvent during stripping.

Distribution-coefficient data for Ag^{111} given in Table 3.3 confirms the behavior of silver in the mixer-settler run. Silver can be kept in the solvent if dilute acid is used to strip the transplutonium elements. However, the higher acid concentrations are required for good ruthenium and zirconium decontamination. In most processing, decay times will be long enough to eliminate Ag^{111} activity. If processing is required after only short decay times, other procedures will be needed to remove silver and possibly Te^{132}.

Table 3.3. Extractability of Silver Into 0.6 <u>M</u> Alamine 336·HCl--DEB from Chloride Solutions

Aqueous Phase	Ag^{111} Distribution Coefficient, O/A
1 <u>M</u> HCl	420
3 <u>M</u> HCl	113
5 <u>M</u> HCl	19
7 <u>M</u> HCl	2.6
9 <u>M</u> HCl	0.4
11 <u>M</u> LiCl	19
9 <u>M</u> LiCl--0.3 <u>M</u> $AlCl_3$	16

Behavior of Ruthenium in the Tramex Process

Fission product ruthenium in Tramex feed will probably exist in both the tri- and tetravalent stages. Only Ru^{4+} is extracted along with the transplutonium elements, and Ru^{3+} remains in the aqueous waste. Trivalent ruthenium is rapidly oxidized to tetravalent ruthenium by Cl_2, H_2O_2, and nitrite. Since H_2O_2 is produced by radiolysis of the feed solution, it is difficult to keep ruthenium quantitatively in the trivalent state so that it will not be extracted. Therefore, at least some if not all of the ruthenium can be expected to extract with the transplutonium elements. The tetravalent ruthenium that is extracted is slowly reduced by the solvent to the relatively inextractable Ru^{3+}, thus allowing a small fraction of the ruthenium to be stripped along with the transplutonium elements. In a mixer-settler run with H_2O_2 added to the strip solution, the overall ruthenium decontamination factor was only 10^3. In this case the H_2O_2 was not completely effective in reoxidizing ruthenium to Ru^{4+} during stripping operations because the half-life of H_2O_2 in the strip solution is only about a minute, compared with a 15- to 20-min residence time in the mixer-settler. In tests at alpha radiation levels of 3 w/liter, H_2O_2 from water radiolysis was not sufficient to keep the ruthenium quantitatively in the solvent during stripping. In mixer-settler tests, the addition of a side stream of $NaNO_2$ to the strip solution, or scrubbing the strip product with 0.6 M Alamine 336·HNO_2, was effective in oxidizing the ruthenium or in forming an extractable complex. In these tests, ruthenium decontamination factors were equal to or greater than 10^4, which is adequate for most applications.

Effect of Alpha Radiation on Tramex Solutions

Solvent. -- No difficulty in phase separation was noted when 0.6 M Alamine 336·HCl--DEB was in contact and intermittently mixed with 10.5 M LiCl containing Cm^{242} for a total solvent exposure of about 300 whr/liter. The most notable effect was a 30% decrease in the curium distribution coefficient, which was still large enough for satisfactory processing. This test indicates that the solvent is sufficiently stable to radiation for full-activity-level processing because the maximum solvent exposure anticipated in the TRU Plant is less than 100 whr/liter.

A 10-ml sample of 10.5 M LiCl containing about 25 mg/liter of Cm^{242} (equivalent to about 3 w/liter) was mixed with 10 ml of 0.6 M Alamine 336·HCl in DEB. The two phases remained in contact, with occasional remixing, over a 116-hr period. The curium distribution coefficient decreased from 3.6 to 3.2 at an exposure of approximately 60 whr/liter and to 2.4 at about 300 whr/liter. After an exposure of about 200 whr/liter, some interfacial "crud" formed but did not cause emulsion problems. Phase separation was satisfactory when the experiment was terminated at an exposure of about 300 whr/liter. Curium was readily stripped from the solvent into 9 $\underline{M}$ HCl.

Feed Solution. -- A potentially serious problem may result from high activity levels in the Tramex feed solutions. Tests showed that radiation destroys the acid and eventually precipitates those metal cations that form insoluble hydroxides. Since Tramex feed must be low in acid in order to obtain sufficiently high distribution coefficients for the transplutonium elements, the adjusted feeds will be stable for only limited times. A simulated feed (10 $\underline{M}$ LiCl and 0.33 $\underline{M}$ HCl spiked with 1.22 w/liter of Cm^{242}) became acid deficient and precipitated more than 90% of the curium in 15 days. This corresponds to a G value for acid destruction of 5.6 molecules per 100 ev. Unless methods of preventing acid loss by radiation can be found it will be necessary to add acid to compensate for the loss.

Since H_2O_2 is a product of water radiolysis, its behavior in synthetic Tramex feed was investigated. The half-life of H_2O_2 in 10 $\underline{M}$ $LiCl_3$--0.1 $\underline{M}$ HCl was 600 hr at 8°C, 110 hr at 22°C, and 3.5 hr at 65°C. The half life of H_2O_2 in 10 $\underline{M}$ LiCl--0.15 $\underline{M}$ $AlCl_3$ at 65°C was 2 hr. Hydrogen peroxide reaction with these solutions resulted in the loss of acid.

The effect of high alpha activity levels in LiCl solutions was investigated with Cm^{242} as the activity. A 10 $\underline{M}$ LiCl solution was made up to 1.22 w/liter with highly purified Cm^{242}. Even though the solution was maintained at 8°C to minimize H_2O_2 decomposition, there was no detectable (less than 0.001 $\underline{N}$) buildup of H_2O_2. Acidity of the solution decreased from 0.33 $\underline{N}$ H^+ to 0.05 $\underline{N}$ H^+ in about six days, and in 15 days more than 90% of the curium had precipitated. The calculated G value for acid depletion is 5.6 molecules per 100 ev. It appears that most of the free radicals formed by radiolysis do not react to form H_2O_2 but react directly to destroy the acid. Analysis of the resulting gas gave 6 parts H_2 to 1 part O_2, with no Cl_2 or HCl.

Evaluation of Alternative Solvents

Triisooctyl amine (TIOA) as an alternative to Alamine 336, and diisopropylbenzene (DIPB) as an alternative to diethylbenzene (DEB), were investigated since both are commercially available in sufficient quantity and both give increased curium extraction. Curium distribution coefficients (DC's) and viscosities for various combinations of 0.6 $\underline{M}$ amine in diluent are given in Table 3.4. Although it is possible to increase curium distribution coefficients from 25 to 60%, solvents giving the highest extraction also have the highest viscosities. Since phase separation between 0.6 $\underline{M}$ Alamine 336--DEB and 10 $\underline{M}$ LiCl--0.2 $\underline{M}$ $AlCl_3$ is marginal, and temperatures of 50°C are necessary for satisfactory mixer-settler operation, it does not appear to be advisable to use a solvent with a higher viscosity.

Table 3.4. Curium Extraction from 10 $\underline{M}$ LiCl--0.2 $\underline{M}$ $AlCl_3$ into Various Solvents

Solvent	Curium DC, O/A	Solvent Viscosity (Centipoise)
0.6 $\underline{M}$ Alamine 336-DEB	6.6	4.95
0.6 $\underline{M}$ Alamine 336-DIPB	8.5	6.43
0.6 $\underline{M}$ TIOA-DEB	8.2	5.83
0.6 $\underline{M}$ TIOA-DIPB	10.6	7.80

Tramex Feed-Adjustment Studies

Tests made on a 1- to 2-liter scale in laboratory glassware indicate that it is possible to prepare stable Tramex feed of low acidity by simply distilling off acid and water until the desired salt concentration is obtained. Stable solutions containing 0.2 $\underline{M}$ $AlCl_3$ and from 6 to 10 $\underline{M}$ LiCl were prepared. The aluminum precipitated from a feed adjusted to 0.2 $\underline{M}$ $AlCl_3$ and 10.5 $\underline{M}$ LiCl; however, data in Fig. 3.5 indicate that the $AlCl_3$ solubility for 10.5 $\underline{M}$ LiCl is only 0.14 $\underline{M}$. These solutions were prepared by distilling dilute solutions containing 2 $\underline{M}$ HCl and $AlCl_3$ and LiCl in the proper ratio. Further work is needed to determine if aluminum

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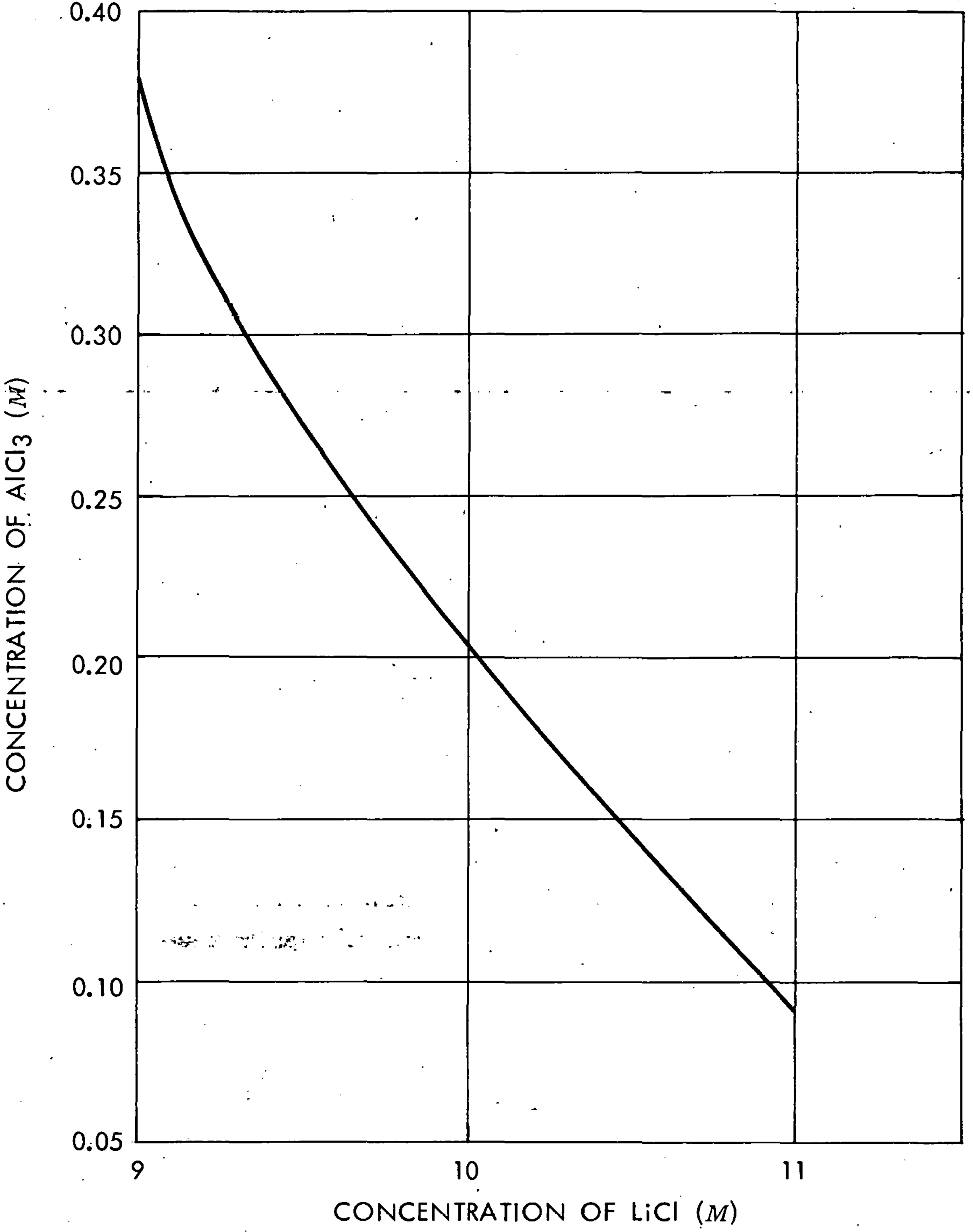

Fig. 3.5. Solubility of Aluminum Chloride in Concentrated Solutions of Lithium Chloride at 24 ± 1°C.

hydrolysis and precipitation occur under the same conditions and if the configuration of the equipment is important.

Physical Properties of Tramex Solutions

A program has been started to accumulate data on the physical properties of solutions used in the Tramex process. The solubility of $AlCl_3$ in concentrated LiCl solutions is important, for example, since it may determine the volume of feed solution required to process targets fabricated from aluminum. The solubility of $AlCl_3$ at room temperature ($24 \pm 1^{\circ}C$) decreases from 0.38 M in 9 M LiCl to 0.09 M in 11 M LiCl (see Fig. 3.5). The proposed Tramex feed is 10 M $LiCl_3$ containing 0.1 M $AlCl_3$. This appears to be a safe aluminum concentration since the solubility is 0.2 M $AlCl_3$ in 10 M $LiCl_3$.

Densities, boiling points, and refractive indices for 9, 10, and 11 M LiCl containing $AlCl_3$ are given in Fig. 3.6.

3.1.2 Separation of Americium and Curium from Each Other

Americium and curium can be separated from each other by chromatographic elution from ion exchange resin or by precipitation based on differences in solubility when americium is in a higher oxidation state. The ion exchange methods are of special interest since they may be useful for separating californium, einsteinium, and fermium. During this report period the precipitation of gram quantities of $KAmO_2CO_3$ was studied, and good separation of americium and curium by chromatographic elution from cation exchange resin with ammonium alpha-hydroxyisobutyrate and from anion exchange resin with 4 M $LiNO_3$ was demonstrated at high activity levels. Measured solubilities of rare earth cyanoargentate complexes indicate little or no possibility of using these complexes for americium-curium separation.

Precipitation of $KAmO_2CO_3$

A 2.5-g sample of Am^{241} contaminated with La, Ce, Fe, and Ca was purified by precipitating $KAmO_2CO_3$ from 3 M K_2CO_3. The precipitate contained 99.6% of the americium, and it was free of all contaminants except cerium. This method can also be used to separate americium from curium, which will remain in solution.

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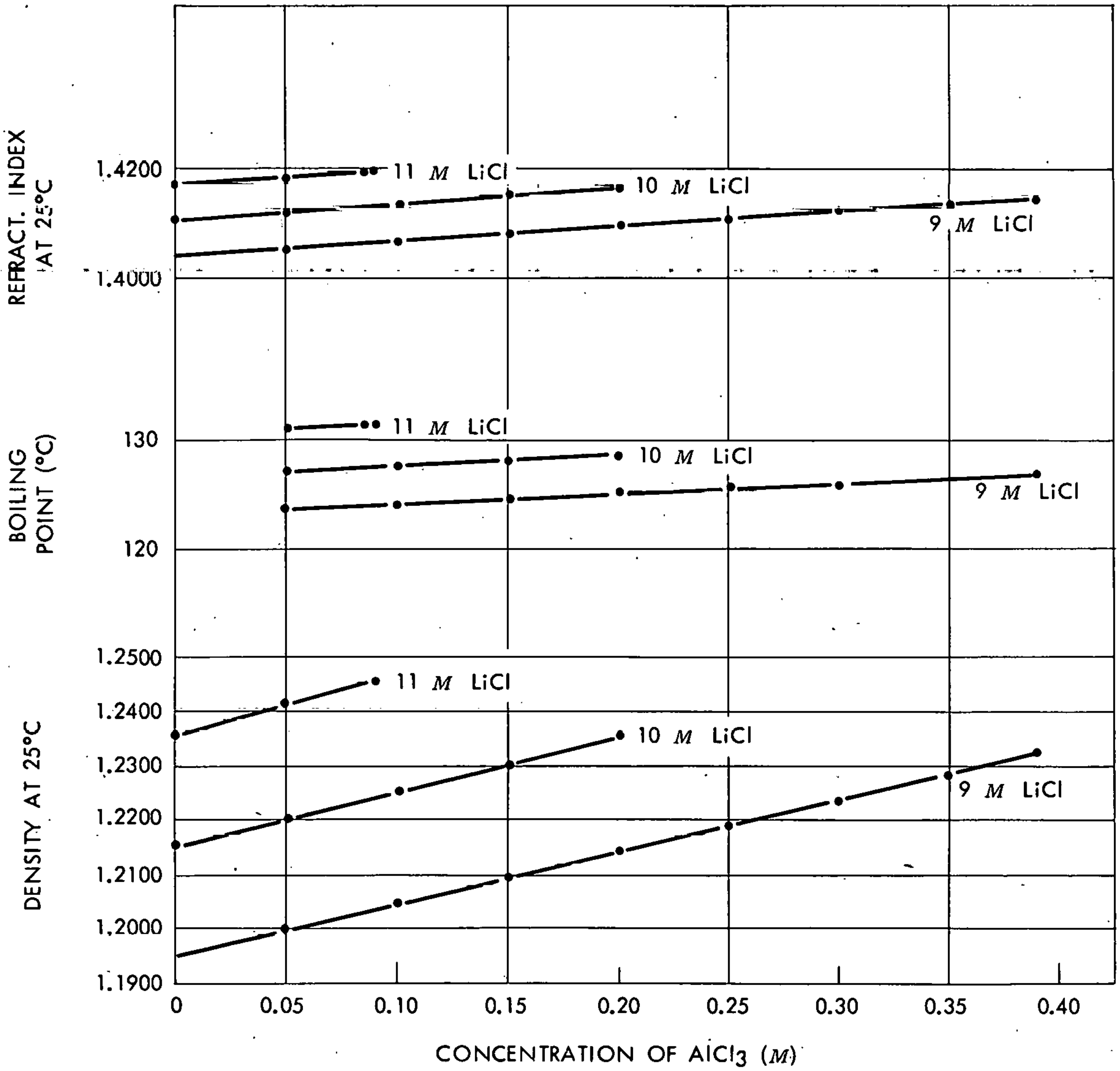

Fig. 3.6. Refractive Indices, Boiling Points, and Densities of LiCl-$AlCl_3$ Solutions.

A 2.5-g sample of Am^{241} contaminated with 7 g of lanthanum, 1 g of cerium, about 0.1 g of iron, and a trace of calcium was dissolved in dilute hydrochloric acid. A carbonate solution was prepared by the slow addition and vigorous mixing of the slightly acid sample solution into 1 liter of $3\ \underline{M}\ K_2CO_3$. The solution was adjusted to $0.1\ \underline{M}$ in NaOCl and heated at 80°C for 3 hr to oxidize the americium, which precipitated as $KAmO_2CO_3$. This precipitate contained 99.6% of the americium and most of the cerium, which was also oxidized by the NaOCl. The $KAmO_2CO_3$ was a very dense, light-tan precipitate, easily filtered, and its solubility in the filtrate was about 7 mg/liter. Cerium was the only contaminant found in the americium product.

Ion Exchange Separation of Americium and Curium at High Activity Level

By using fine-mesh resin and by pressurizing the column, gassing caused by high alpha activity levels was controlled well enough to accomplish satisfactory americium-curium separation by chromatographic elution with either ammonium alpha-hydroxyisobutyrate from cation resin or with $4\ \underline{M}\ LiNO_3$ from anion resin. Since the Cm^{242} activity levels in these tests were equivalent to 0.4 and 1 mg of Cf^{252}, indications are that californium-einsteinium-fermium separations at the milligram level may be possible.

A quantity of Cm^{242} activity equivalent to 0.4 mg of Cf^{252} was separated from Am^{241} by chromatographic elution from a 5-ml column of -270-mesh Dowex 50 resin with ammonium alpha-hydroxyisobutyrate. The volume of gas formed was reduced by pressurizing the column. This resulted in successful operation, and a product containing 96% of the curium free from americium was obtained.

Curium-242 activity equivalent to 1 mg of Cf^{252} was successfully separated from Am^{241} on a 5-ml column of Dowex 1-8X (-270-mesh) resin. The americium and curium were loaded from $8\ \underline{M}\ LiNO_3$ and eluted with $4\ \underline{M}\ LiNO_3$ adjusted to a pH of 1.7. As in the above test, the gas volume was reduced by pressurizing the column. The elution was completely satisfactory (Fig. 3.7), and the product contained 99% of the curium, almost free of americium. Plans are in progress to test this system for californium-einsteinium separation.

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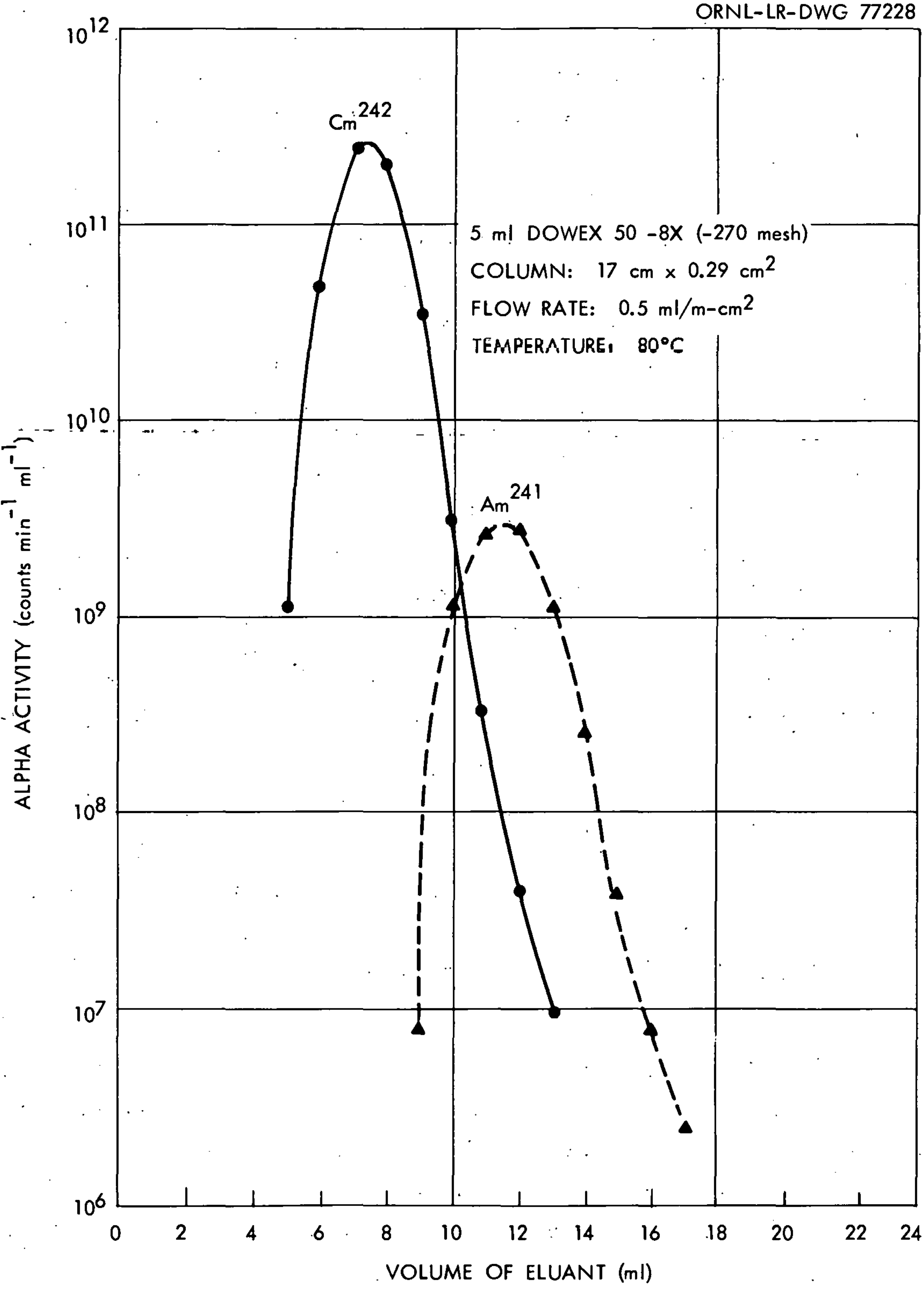

Fig. 3.7. Curves for the Elution of Americium and Curium from Dowex 1 with 4 N $LiNO_3$.

Preparation and Solubilities of Rare Earth Cyanoargentate Complexes

It was reported[3] that the solubility of the rare earth (RE) cyanoargentates increased with increasing atomic number up to europium and then sharply decreased with gadolinium. This might provide a means of separating americium and curium since americium is often homologous with europium, and curium is often homologous with gadolinium. However, our results for the solubility measurements did not agree with those reported. The following solubilities in H_2O at pH 6.0 were measured:

Rare Earth	$RE[Ag(CN)_2]_3 \cdot 3H_2O$ (g/liter)
La	11.4
Ce	9.4
Pr	8.4
Gd	11.6
Tb	16.0
Ho	27.2
Tm	58.8

The complexes were formed by precipitating from neutral nitrate solutions with $KAg(CN)_2$ at pH 6.0. The precipitates were filtered and dried.

3.1.3 Behavior of Nickel During Simulated Curium Precipitation

Nickel can be kept in solution when rare earths, which were substituted for curium, are precipitated. This method of nickel decontamination is especially important since the present Tramex flowsheet does not remove nickel. The following results were obtained for rare earth precipitation from a nitric acid solution containing 1 g/liter nickel and 1 g/liter rare earths:

Conditions	Nickel Decontamination Factor	Rare Earth Precipitation
1 $\underline{M}$ HNO_3; pptn. with 0.5 $\underline{M}$ $H_2C_2O_4$	1.4	>98%
1 $\underline{M}$ HNO_3; pptn. with 0.5 $\underline{M}$ $(NH_4)_2C_2O_4$	6.7	>98%
0.1 $\underline{M}$ HNO_3; pptn. with 0.5 $\underline{M}$ $(NH_4)_2C_2O_4$	260	>99%
Hydroxide pptn. by hydrolysis of urea	330	>99%

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[3] R. A. Chupakhina, Yu. V. Indukaev, and V. V. Serebrennikov, Russian J. Inorg. Chem., 6(12), 1371-72 (1961).

These results show that by keeping the acid concentration low, and in the presence of ammonium ion, most of the nickel remains in solution, and good nickel decontamination can be obtained.

3.1.4 Anion Exchange Recovery of Americium and Curium from Nitrate Solutions

Additional studies were made of the variables that affect the anion exchange recovery of americium and curium contained in the raffinate when plutonium is recovered from highly irradiated plutonium-aluminum alloy. This process[4] makes use of americium, curium, and rare earth sorption on anion exchange resin from concentrated aluminum nitrate solutions of low acidity. Americium and curium are recovered in dilute nitric acid solution free of aluminum, corrosion products, and most fission products except the rare earths. Additional separation by the Tramex process is necessary to purify the americium-curium.

During this report period the effects of rare earth concentration in the feed, flow rate through the column, and acid deficiency in the feed were investigated. Attempts to measure resin capacity for cerium were unsuccessful since equilibrium was not attained in the 96-hr contact time used. No evidence of radiation damage was noted when full-activity-level feed was in contact with the resin for 21 days.

Effect of Feed Variables: Rare Earth Concentration, Flow Rate, Acid Deficiency

The behavior of europium tracer, which sorbs slightly less than curium, was used to measure the effects of these variables. Tests were made with a 1-in.-diam glass column filled with resin to a depth of 45 cm. Europium losses were measured during sorption at 60°C from 15 column volumes of feed and during subsequent washing with five displacement volumes of 8 M $LiNO_3$.

Increasing the rare earth concentration above 0.5 g/liter in 2.6 M $Al(NO_3)_3$ feed adversely affected europium recovery (Fig. 3.8). With a flow rate of 2 ml cm^{-2} min^{-1}, europium recoveries were 89, 87, 77, 62,

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[4] D. E. Ferguson, Transuranium Quarterly Progress Report for Period Ending February 28, 1962, ORNL-3290, p 7.

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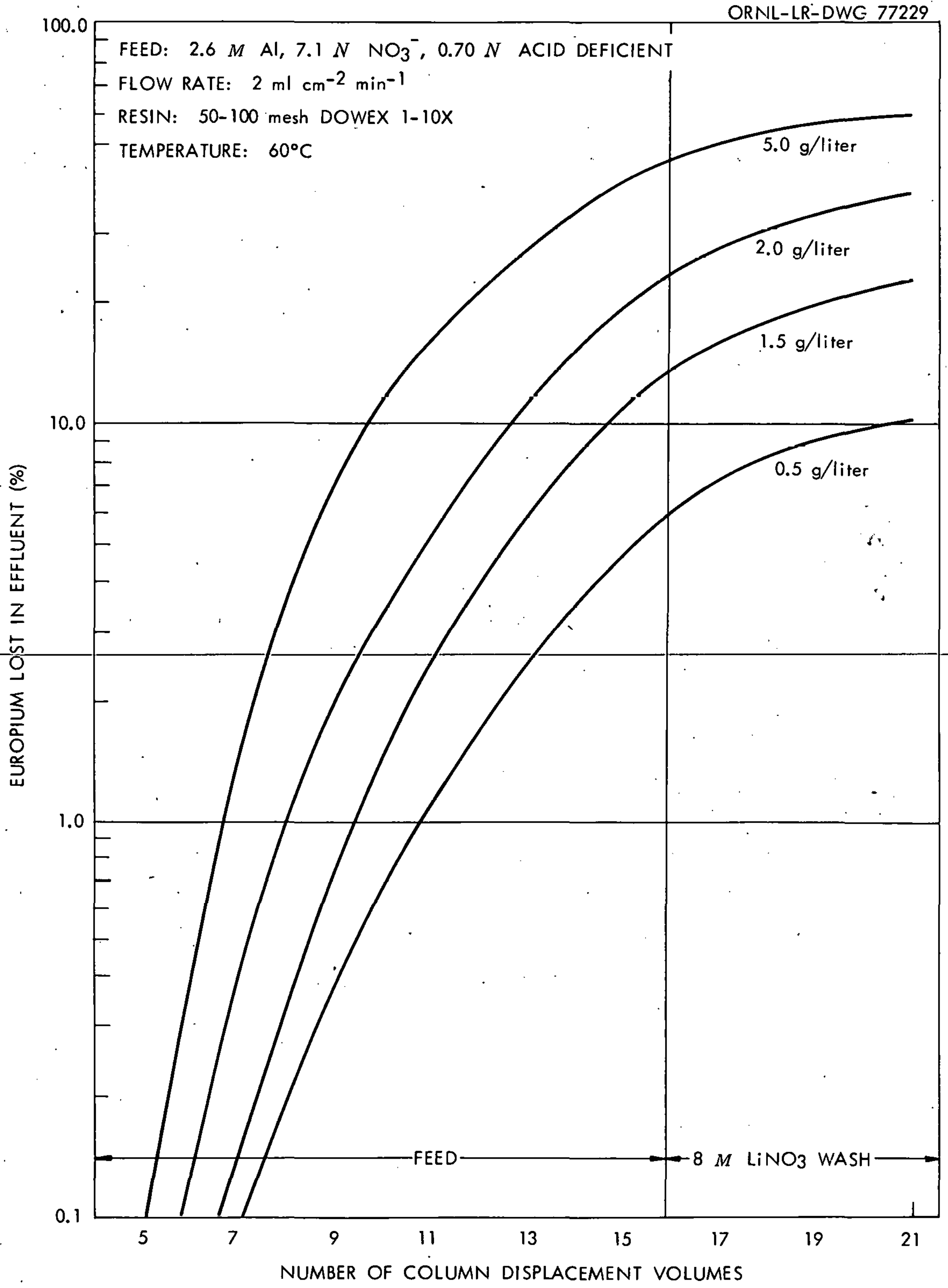

Fig. 3.8. Europium Losses During Rare Earth Loading on Anion Exchange Resin: Effect of Concentration of Rare Earth Feed Solution.

and 40% for rare earth feed concentrations of 0.5, 1.0, 1.5, 2.0, and 5.0 g/liter, respectively. This effect can be offset by decreasing the flow rate, as shown by the results plotted in Fig. 3.9. Europium recovery from feed containing 2 g/liter of rare earths was increased from 62 to 89% by decreasing the flow rate from 2 to 1 ml cm^{-2} min^{-1}. The same effect is shown in Fig. 3.10, for feed containing 5 g/liter of rare earths; however, europium losses were 40% even at a flow rate of 0.6 ml cm^{-2} min^{-1}. Since there was no appreciable europium loss during loading from the first ten column volumes of feed, complete europium recovery can be obtained by limiting the amount of feed to less than this value.

Since acid-deficient aluminum nitrate is more soluble than neutral aluminum nitrate, the effect that acid deficiency in the feed had on europium recovery was investigated. With 2.6 $\underline{M}$ Al^{3+} it was found that europium recovery was unaffected in changing from neutral feed to 0.7 $\underline{N}$ acid-deficient feed. However, in going from 0.7 $\underline{N}$ acid-deficient feed to 1.17 $\underline{N}$ acid-deficient feed, europium recovered in a standard run decreased from 90% to about 81%.

Resin Capacity

An attempt was made to determine the cerium loading capacity for three mesh sizes of Dowex 1-10X resin. However, equilibrium was not attained in 96 hr at 25°C. In these experiments 1 g of dry resin was tumbled in 10 ml of 9 $\underline{M}$ $LiNO_3$ that contained 20 g/liter of cerium as $Ce(NO_3)_3$, plus cerium tracer. The results are shown in Fig. 3.11. Resin loading increased with decreasing particle size, and equilibrium was approached more rapidly with the finer resin. In 96 hr, it appears that equilibrium was nearly reached for -400 mesh resin but not for the 100- to 200- or 50- to 100-mesh resin.

Resin Stability at Full Activity Level

Dowex 1-10X resin in contact with high-activity-level feed for 21 days showed no evidence of radiation damage. Two grams of resin were in contact for 21 days with 4 ml of 2.6 $\underline{M}$ $Al(NO_3)_3$ containing 3.14×10^8 gross alpha counts min^{-1} ml^{-1} and 1.68×10^{10} gross gamma counts min^{-1} ml^{-1}. This solution was prepared from full-activity-level plutonium raffinate.

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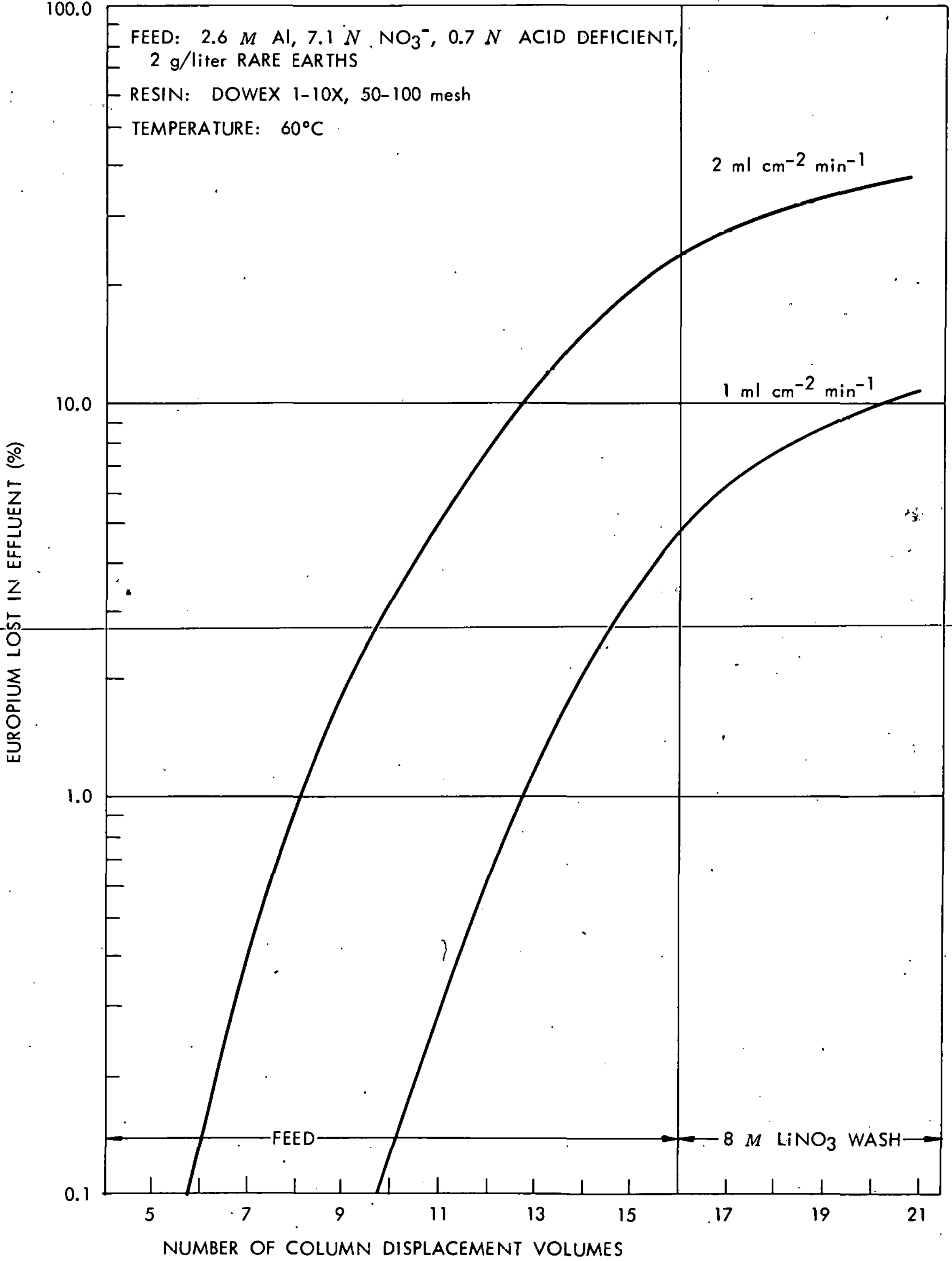

Fig. 3.9. Europium Losses During Rare Earth Loading of Anion Exchange Resin: Effect of Flow Rate for a Feed Containing 2 g/liter Rare Earths.

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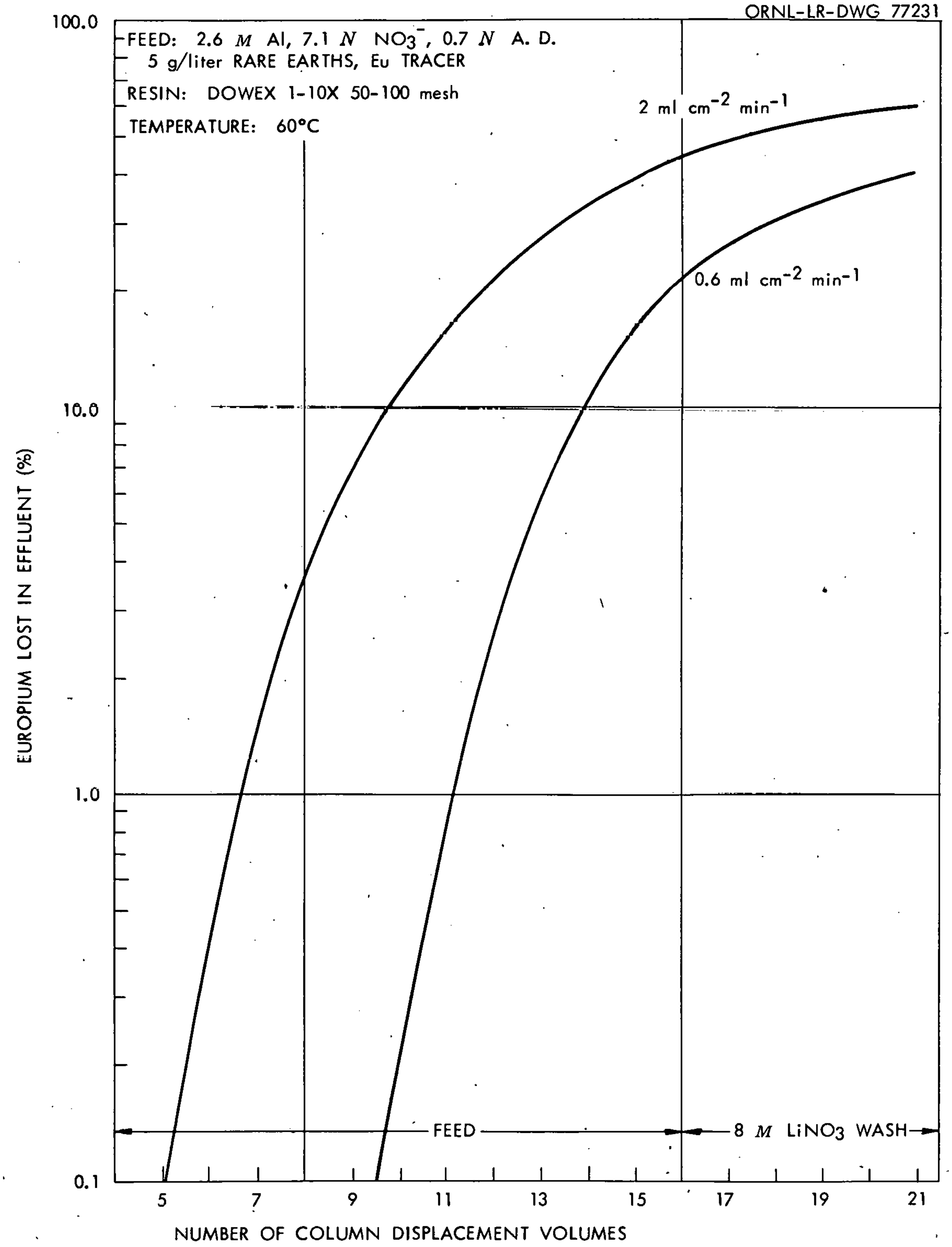

Fig. 3.10. Europium Losses During Rare Earth Loading of Anion Exchange Resin: Effect of Flow Rate for a Feed Containing 5 g/liter Rare Earths.

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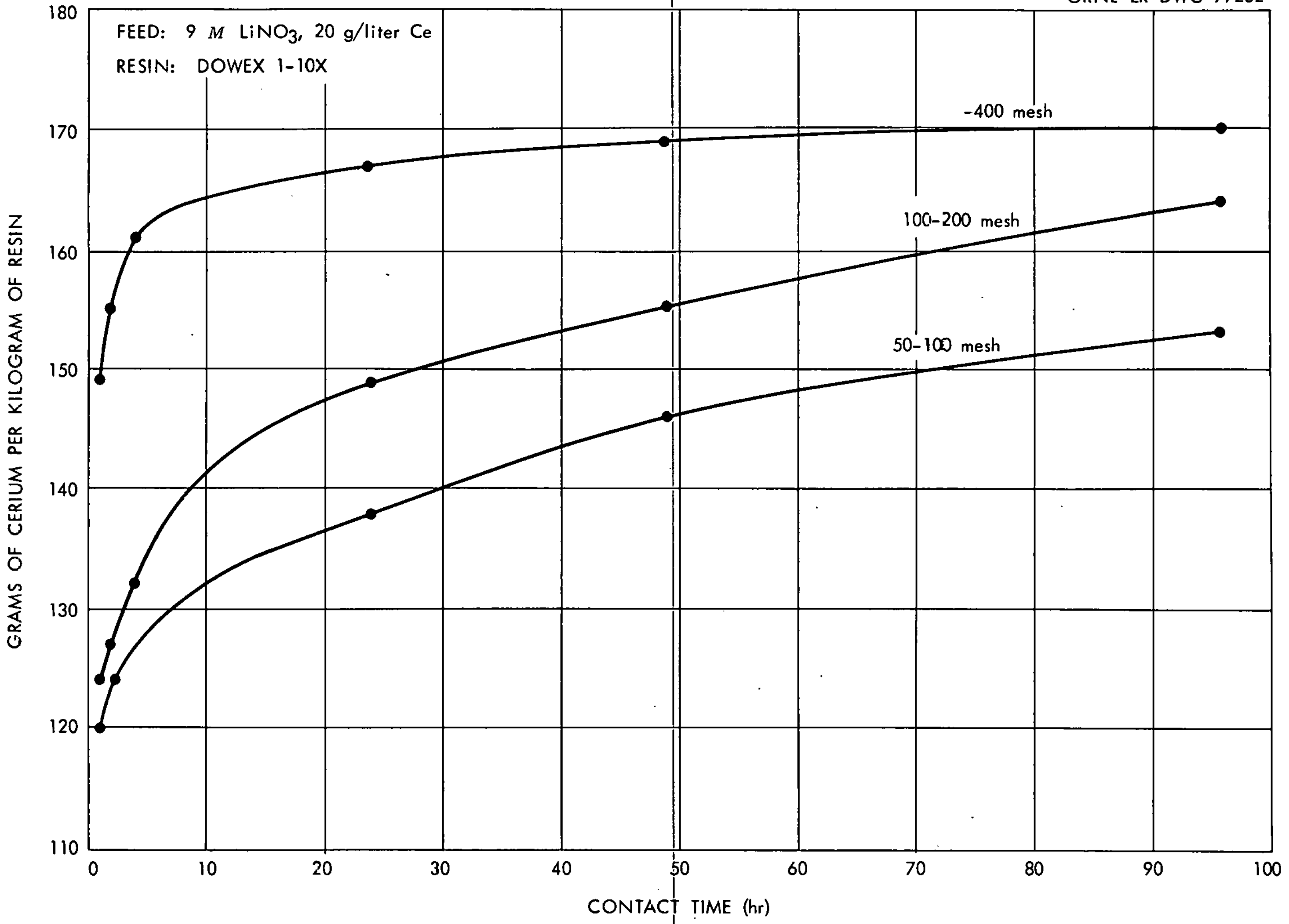

Fig. 3.11. Cerium Loading on Dowex 1-10X Resin at 25°C.

There was no visible discoloration of the resin, and the gross alpha distribution coefficient (K_d) increased slowly during the first eight days and then remained almost constant at about 50. This behavior is similar to that for cerium noted above.

3.1.5 Modification of Process for Recovery of Plutonium, Americium, and Curium from Plutonium-Aluminium Rods

Due to the anticipated difficulty in the nitric acid dissolution of aluminum present in highly irradiated plutonium-aluminium rods for the TRU program, a process based on caustic dissolution of aluminum was investigated. In this process, dissolver solution containing 1.2 moles of NaOH and 1.55 moles of $NaNO_3$ per mole of aluminum is used. Sodium nitrate is required to convert hydrogen, liberated by the dissolution reaction, to ammonia. Aluminum dissolution is followed by acidification with nitric acid to dissolve plutonium and other metals. Feed for plutonium recovery is adjusted to contain 2.5 $\underline{M}$ $NaNO_3$, 0.8 $\underline{M}$ $Al(NO_3)_3$, and 2.6 $\underline{M}$ HNO_3.

Although plutonium recovery is apparently satisfactory, scouting tests indicated that this amount of $NaNO_3$ will cause difficulty in the anion exchange recovery of americium and curium from the plutonium waste. Feed for the americium-curium recovery must be an acid-free concentrated salt solution with a nitrate concentration equivalent to at least 2.33 $\underline{M}$ $Al(NO_3)_3$ (7 $\underline{N}$ nitrate). Acid-free feed cannot be prepared by the evaporation of plutonium waste because $NaNO_3$ precipitates before any appreciable amount of acid is distilled off. Column loading is also adversely affected by feed containing a high proportion of $NaNO_3$ because $NaNO_3$ is less effective than $Al(NO_3)_3$ as a complexer for americium and curium.

It appeared that these difficulties could be overcome by increasing the $Al(NO_3)_3$:$NaNO_3$ ratio of the adjusted feed and by obtaining the highest nitrate concentration possible. Mixtures of KNO_3, $NaNO_3$, and $Al(NO_3)_3$ were considered as feed because a point of maximum nitrate solubility (10.4 $\underline{N}$ at 25°C) occurs at an Na:K:Al mole ratio of about 1:1:2.1.

In order to define variables necessary for americium-curium recovery from plutonium waste in which either a caustic dissolution or a nitric acid dissolution in conjunction with a caustic dissolution is used, the following studies were made: (1) The effect of potassium on aluminum dissolution was investigated; (2) column runs were made with feed containing various ratios of $Al(NO_3)_3$, $NaNO_3$, and KNO_3; (3) feed adjustments were investigated using various $Al(NO_3)_3$ mole ratios; and (4) nitrate solubility studies were made for $NaNO_3$-$Al(NO_3)_3$ and $NaNO_3$-KNO_3-$Al(NO_3)_3$ mixtures.

Aluminum Dissolution Studies

The caustic-dissolution procedure requires the use of 1.2 moles of NaOH and 1.55 moles of $NaNO_3$ per mole of aluminum, and substituting KNO_3 for some of the $NaNO_3$ does not interfere with the dissolution. It was found that with an overall Na:K mole ratio of 1, the aluminum dissolution rate, in the presence of excess aluminum, can be controlled by varying the NaOH concentration. Results obtained for 10- and 30-min dissolutions as a function of NaOH concentration are shown in Fig. 3.12. Also, varying the silicon content of the aluminum from 0.1 to 3.0% did not affect the dissolution rate.

The Effect of Aluminum Concentration on Americium-Curium Loading on Anion Resin

Column runs at 60°C, using europium tracer and macro quantities of rare earths, were made to determine the effect of feed containing $NaNO_3$ and KNO_3 on americium-curium loading. Using a flow rate of 1 ml cm^{-2} min^{-1} with Dowex 1-10X resin (50 to 100 mesh), europium recovery was only 63% from feed containing 1.1 $\underline{M}$ $Al(NO_3)_3$, 3.0 $\underline{M}$ $NaNO_3$, and 1.66 $\underline{M}$ KNO_3. By increasing the $Al(NO_3)_3$ concentration to 2 $\underline{M}$ and decreasing both the $NaNO_3$ and KNO_3 concentrations to 1 $\underline{M}$, europium recovery was increased to 94%, indicating that satisfactory americium-curium recoveries can be obtained from feed that contains at least 2 $\underline{M}$ $Al(NO_3)_3$.

Feed Adjustment Studies for Americium-Curium Recovery

Acid-free feeds can be successfully prepared by distillation until the acid is depleted, followed by dilution to the desired salt concentration, provided that the aluminum-to-sodium-and-potassium ratio is sufficiently

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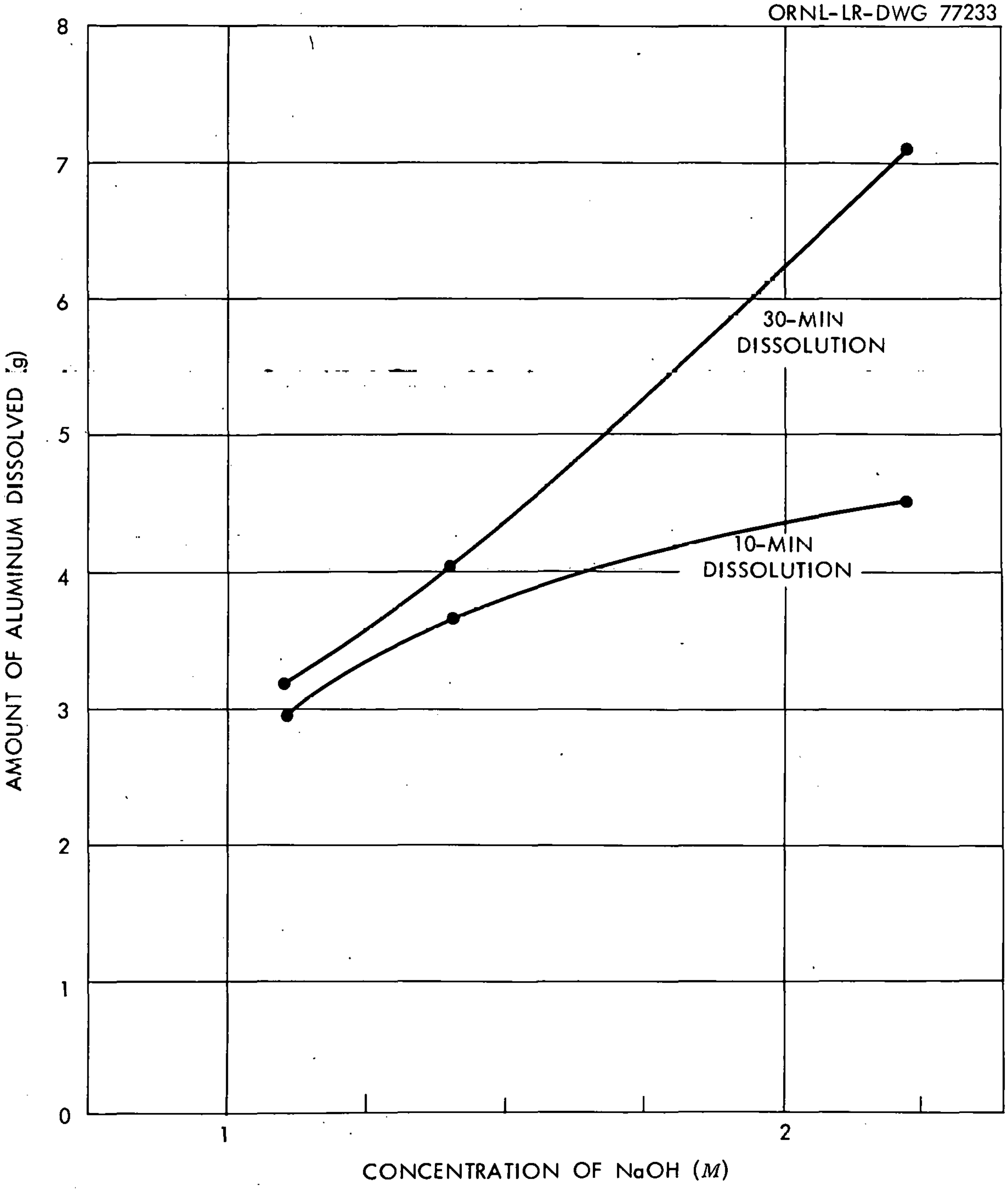

Fig. 3.12. Dissolution of Aluminum as a Function of Sodium Hydroxide Concentration.

high. By replacing half the $NaNO_3$ with KNO_3, feeds can be successfully prepared when the Al:Na + K mole ratio is as low as 0.8. With a mole ratio of 3.3, acid-free feed is obtained at 140°C, but, when the mole ratio decreases below 3.3, higher temperatures are necessary to obtain neutral feeds.

Solubilities for the System: $Al(NO_3)_3$-$NaNO_3$-KNO_3-H_2O

Nitrate solubility was determined for three systems in which the aluminum concentration is sufficient to provide satisfactory americium-curium loading characteristics. These systems include mixtures of $NaNO_3$ and $Al(NO_3)_3$, mixtures of $NaNO_3$, KNO_3, and $Al(NO_3)_3$ in which the Na:K mole ratio is 1, and mixtures of $NaNO_3$, KNO_3, and $Al(NO_3)_3$ in which the Na:K mole ratio is 2. In each case the solubility range studied varied from pure $Al(NO_3)_3$ to a point of maximum nitrate solubility. Solubility data are given in Fig. 3.13, where nitrate concentration is plotted as a function of the percentage of nitrate that is associated with aluminum. Curves indicating the $Al(NO_3)_3$ component of the mixtures are shown, as well as total nitrate curves.

Scouting tests indicate that nitrate solubilities of 13 N are possible for similar sodium, potassium, and aluminum systems in which the $Al(NO_3)_3$ is 2 N acid deficient.

3.1.6 Amine Extraction of Americium and Curium from Nitrate Solutions

Tertiary amine extraction is an attractive alternative to the amine exchange procedure for the recovery of americium and curium contained in raffinate after the plutonium has been recovered from highly irradiated plutonium-aluminum alloy. This process makes[5] use of the extraction of americium, curium, and rare earth into a tertiary amine solvent from neutral nitrate salt solution. Americium and curium are recovered in either dilute nitric or hydrochloric acid solution free of aluminum, corrosion products, and most fission products except the rare earths. Additional separation by the Tramex process is necessary to purify the americium and curium.

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[5] D. E. Ferguson, Transuranium Quarterly Progress Report for Period Ending August 31, 1962, ORNL-3375.

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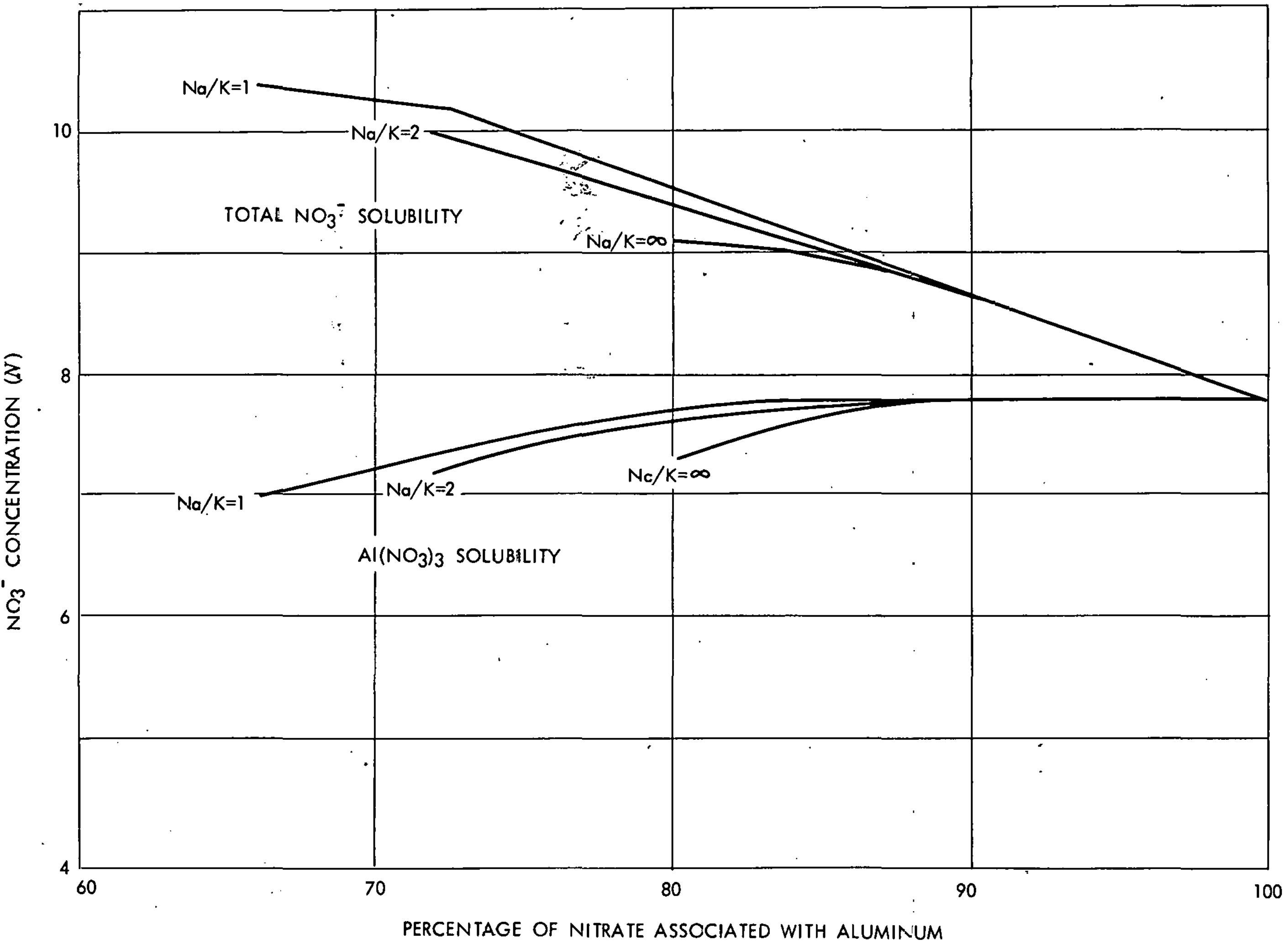

Fig. 3.13. Total-Nitrate Solubility and Aluminum Nitrate Solubility at 25°C for the System: $Al(NO_3)_3$-$NaNO_3$-KNO_3-H_2O.

During this report period, excellent recovery and decontamination from probable corrosion products was demonstrated in a laboratory-scale mixer-settler run with rare earths as stand-ins for americium and curium. Distribution coefficients determined for a number of possible contaminants also confirm the possibility of good decontamination. The most effective salts for maximum curium extraction are those nitrates that form higher hydrates. Americium and curium distribution coefficients varied by over a factor of 40 for different diluents. The solvent, 0.6 M Alamine 336·HNO_3 in DEB is a satisfactory extractant, with an americium distribution coefficient from 3 M $LiNO_3$--1 M $Al(NO_3)_3$ of 8.0.

Mixer-Settler Demonstration and Contaminant Extractability

A laboratory-scale mixer-settler test demonstrated almost complete recovery of rare earths decontaminated from corrosion products. The 2 M $Al(NO_3)_3$ simulated feed contained 5 g/liter of rare earths as stand-ins for americium and curium, and 1 g/liter each of [redacted] iron, [redacted] nickel, and chromium. The scrub was 8 M $LiNO_3$, and the extractant was 0.6 M Alamine 336·HNO_3 in DEB. With 8 extraction and 8 scrub stages and with a feed-to-scrub-to-extractant ratio of 1:1:2, rare earth loss in the aqueous waste was only 0.005%. The extraction of [redacted] iron, [redacted] nickel, and chromium was so low that they could not be detected in the extractant. Aluminum content in the extractant was 0.09 g/liter. This represents an aluminum decontamination factor of about 300.

Most contaminants, except the rare earth fission products, are so inextractable that they will remain in the aqueous waste. Distribution coefficients between 0.6 M Alamine 336·HNO_3--DEB and 1 M $Al(NO_3)_3$--3 M $LiNO_3$ were less than 0.001 for Mn, Co, Ni, Cr, Ba, and Sr, and were less than 0.1 for Fe, Cu, Zn, Ag, Sn, and Zr. Distribution coefficients for Ru, Cd, Y, and Pd were 0.3, 0.6, 2.4, and 120, respectively.

Effect of Metal Cation on Curium Extractability

Curium extraction into tertiary amine depends strongly on the type of cation used in the nitrate salting solution. Distribution coefficients between 0.6 M Alamine 336·HNO_3-DEB and 6.0 N NO_3^- solutions made from a number of different nitrate salts are given in Table 3.5. The large variation in distribution coefficients from 0.051 for 6 N NH_4NO_3 to 5.4 for 6 N $Be(NO_3)_2$ appears to be associated with the ability of the smaller ions to tie up larger amounts of water by hydration, thereby increasing the nitrate activity of the solution.

Table 3.5. Effect of Various Metal Nitrate Salting Solutions on Curium Extraction Into 0.6 M Alamine 336·HNO_3-DEB

Cation	NO_3^- (N)	Cm D.C.'s O/A	Ionic Radii[6] (A)
NH_4	6.0	0.051	1.48
Na	6.0	0.10	0.95
Ca	6.0	0.24	0.99
Mg	6.0	4.07	0.65
Li	6.0	4.04	0.60
Be	6.0	5.4	0.31

Effect of Diluent on Americium and Curium Extractability

When different diluents were used for the extractant, americium distribution coefficients (DC's) varied from 0.53 to 38, curium distribution coefficients varied from 0.43 to 20, and americium-curium separation factors varied from 1.23 to 2.06 for the system 0.6 M Alamine 336·HNO_3 vs 3 M $LiNO_3$--1 M $Al(NO_3)_3$. These data, along with viscosity of the solvent and water content of the solvent after equilibration, are given in Table 3.6. Based on the viscosity of 5.4 centipoise and distribution coefficients of 8.0 for americium and 5.3 for curium, diethylbenzene is a satisfactory diluent, and it is recommended because it is the diluent also used for the Tramex process.

Water content of the solvent was nearly constant for a wide variety of diluents and appears to be associated with the amine at the mole ratio of 4 amines to 1 water. Water content did increase slightly when 2-ethylhexanol was added to prevent the formation of a second organic phase.

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[6] Linus Pauling, Nature of the Chemical Bond, p 350, Cornell University Press (1948).

Table 3.6. Effect of Various Diluents on Americium-Curium Extractability Into 0.6 M Alamine 336·HNO_3 from 3 M $LiNO_3$--1 M $Al(NO_3^-)_3$

Diluent	Viscosity (centipoises)	Water Content (g/liter)	Am DC	Cm DC	α Am/Cm
1,2,dichlorobenzene	1.97	3.90	0.53	0.43	1.23
Decane + 10% alcohol	8.15	4.29	2.95	1.52	1.94
Benzene	3.14	2.57	8.04	3.93	2.04
Amsco + 5% alcohol	13.01	3.91	8.45	4.10	2.06
Diethylbenzene	5.39	2.70	7.98	5.27	1.49
CCl_4	5.21	2.08	9.00	5.35	1.68
Heptane + 5% alcohol	6.08	3.75	12.3	7.53	1.63
Toluene	3.06	2.43	13.02	8.48	1.53
Triethylbenzene	8.67	3.18	15.1	9.79	1.54
Xylene	3.44	2.39	20.7	12.96	1.59
Diisopropylbenzene	9.09	3.02	28.8	16.7	1.72
Cyclohexane	9.33	2.93	38.2	20.5	1.86

3.2 Experimental Engineering Studies

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

3.2.1 Development of Disconnects -- (T. S. Mackey)

The problem of maintaining the Transuranium processing equipment and ensuring a high degree of flexibility in equipment arrangement will be solved by removal of the entire equipment rack or the defective component. Quick-acting disconnects will be placed in strategic locations to permit the remote breaking of the various process lines necessary for the substitution of the new equipment component.

The design criteria for the disconnect specifies metal-to-metal seats for sealing action, without gaskets or other loose parts that would interfere or make remote handling with manipulators difficult. Two designs satisfying the above requirements were developed, and test models of each performed satisfactorily.[7]

The first design provides a large sealing-surface for contact. This is accomplished by forcing the male cone by bolt loading into the female cone. Initial contact is made at the tip of the male cone, and contact along the entire length is accomplished by proper bolt loading. Repeated sealing action depends on the slight remaining flexibility of the male cone, which absorbs the wear and will be made replaceable in case of leakage.

An additional design was tested and proved satisfactory after several make and break cycles and also several thermal cycles. This design incorporates spring tension for providing the proper unit stress at the sealing surfaces without undue deformation. The spring action was believed beneficial in providing uniform sealing action and also in avoiding excessive stress buildup during thermal cycling.

The present work consisted in subjecting the two designs to several severe tests in an attempt to isolate the strong and weak points of each and thereby help to select the most suitable design for the Transuranium Process service.

Experimental Procedure

Test disconnects were machined from type 304 stainless steel or Hastelloy-C, with 0.281 to 0.331 in. of inner diameter. This provided a range of flexibility in the male cones and, for a particular loading force, would provide a range of contact area. A helium leak detector was used for leak testing. The leak detector was calibrated with a standard helium leak and gave at 0.01 μ an overall sensitivity of 1×10^{-8} std cc of helium per second. The leak detection procedure was as follows: The disconnect was placed in the clasping device and then connected to the leak detector. The disconnect was then pumped down to 0.01 μ by the leak detector vacuum system. The disconnect assembly was then placed in

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[7] P. P. Holz, Development of a Six-Station Manifold Cone-Seat Disconnect, ORNL CF-61-5-117 (May 1961).

a plastic bag to which helium was admitted continuously to provide an almost pure helium atmosphere. Leakage in excess of 1×10^{-8} std cc of helium per second was immediately registered on the leak detector. If no leakage was indicated, the closure was considered perfect. No attempt was made to measure the magnitude of the disconnect leakage. However, this would have been necessary in only a few cases as most tests gave perfect seals.

Results

Only conclusions are presented here since detailed results are being published separately.[8]

Tests of repeated closures and rough handling demonstrated that, under most conditions, a surface type of seal is superior to the line type, which is accomplished with light spring tension. The force required for sealing the disconnects with punch marks to simulate rough handling far surpasses that which would be available with the 130-lb springs. If the spring force is increased to give closure with roughened cone surfaces, then it is no longer a line-contact seal but becomes an area seal. Thus, in reality, one is a variation of the other, and the selection becomes one of closing force.

In accelerated corrosion tests with aqua regia, the corrosion pattern of the disconnect is such that the inside of the disconnect is dissolved away before the sealing surface of the disconnect is attacked (Fig. 3.14). Therefore, if the disconnect cone base is thicker than the connecting process tubing one could expect the tubing to fail first.

The disconnect parts were completely interchangeable, and it will not be necessary to remove the disconnects from the defective equipment component to use on the substitute. This is a distinct advantage since it would be difficult to decontaminate the used disconnects to a point that would permit reinstallation. Even though misalignment and misaligning forces exist in the process lines a leak-tight closure can still be

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[8] T. S. Mackey, Development of a Line Disconnect for the Transuranium Processing Facility, ORNL-3389 (in press).

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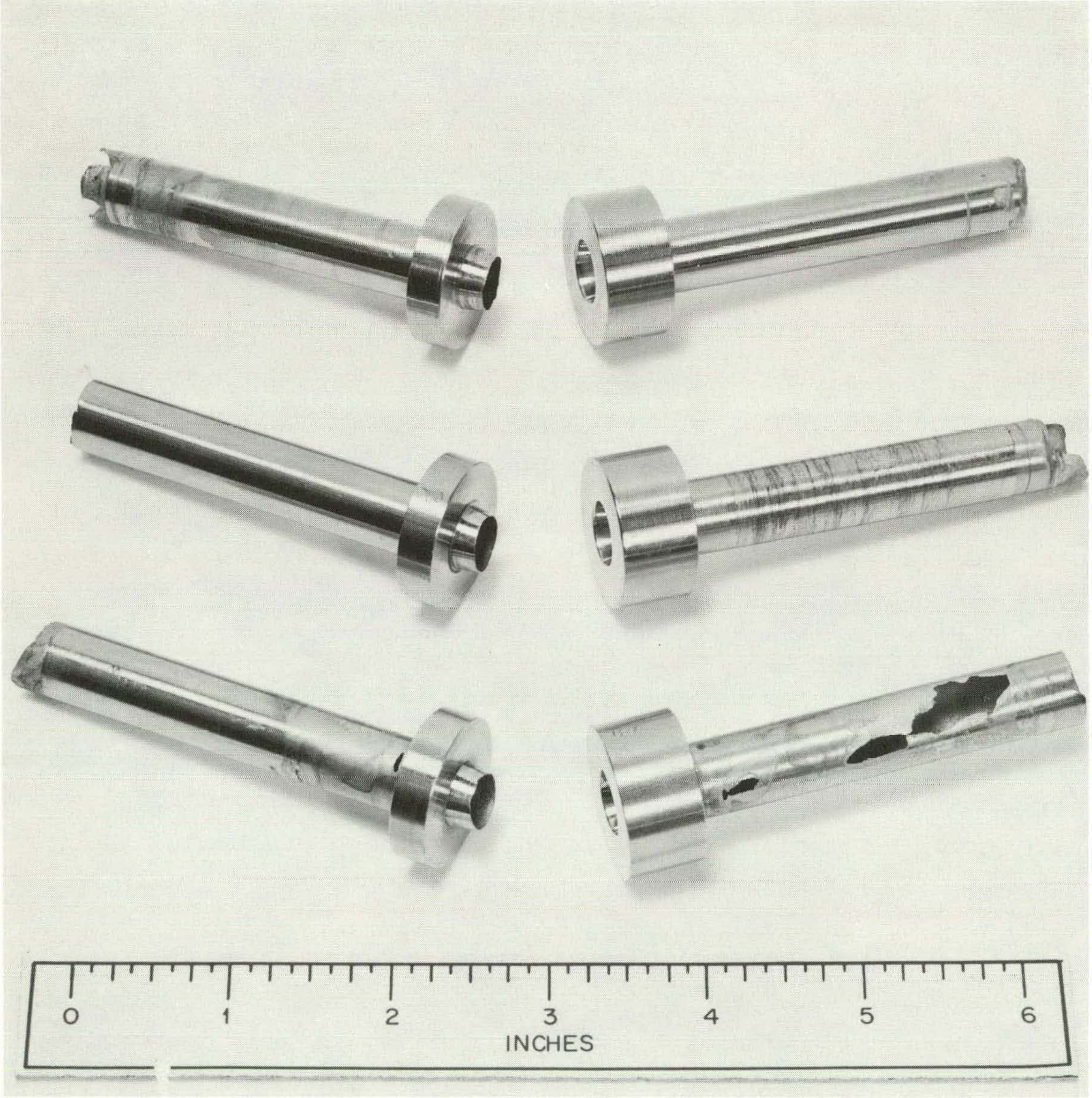

Fig. 3.14. Corrosion Pattern of Disconnects: Results of Accelerated Corrosion in Aqua Regia.

expected. The disconnects can be fabricated with standard machine tolerances and still function reliably.

The depths of the area seal vs seating forces were measured. Since it will not be possible to translate this force into torque measurements because of the large number of variables, it will be necessary to make torque requirement measurements when the prototype disconnect and clasping devices are available from the manufacturer.

Heat treatment of the Hastelloy-C disconnects in order to eliminate increased corrosion at welds was found to destroy the sealing qualities. Mechanical connection by swaging to join the disconnects to process lines was shown to be feasible.

3.2.2 Impact-Wrench Tests -- (T. S. Mackey)

Two electric impact wrenches manufactured by the Ingersol Rand Company were tested along with torque-limiting extensions made by machining standard socket wrench extensions to give thinner cross sections. The 1/2-in. socket drive extensions with a length of 5 in. were ground to give a flat, thin, spring-like section 2-5/8 in. long. The 3/8-in. extensions with a length of 6 in. were ground to give a thin section 4 in. long.

Torque requirements to remove a 1/2-in. nut (carbon steel with fine threads) were measured after tightening with the impact wrenches with and without the machined extensions. The results are as follows:

For size 4USD model B impact wrench:	Torque Required (lb-ft)
With socket directly attached	60
With 5 in. extension	40
With 5 in. extension ground to 3/8 in. thickness	34
With 5 in. extension ground to 1/4 in. thickness	22
With 5 in. extension ground to 3/16 in. thickness	18
With 5 in. extension ground to 9/64 in. thickness	12

For size 2U model A impact wrench	Torque Required (lb-ft)
With socket directly attached	35
With 6 in. extension ground to 1/4 in. thickness	25
With 6 in. extension ground to 3/16 in. thickness	20
With 6 in. extension ground to 1/8 in. thickness	15

The 1/2-in. drive 4USD model B wrench has sufficient torque output to damage 1/2-in. bolt threads if the socket is attached directly to the wrench. By using graduated extensions as indicated alone, it should be possible to safely use the impact wrench on much smaller bolts. After the various bolt sizes are determined, it would probably be wise to determine the maximum torque capacity of the various bolts and then to permanently fasten corresponding sockets to calibrated extensions with the proper torque output. This would guard against thread damage and bolt shearing. The size and weight of both models is almost the same. Since a full range of torque is available with the larger wrench, it is not planned at present to convert the smaller wrench to manipulater use.

3.2.3 Development of Solvent Extraction Contactor -- (F. L. Daley)

Experimental studies were continued to evaluate and improve the contacting equipment used in the Transuranic Element Processing Program. The flowsheet studied consisted in the scrubbing of europium in tracer amounts (a representative of the rare earth fission products) from 30% Alamine 336 in diethylbenzene (DEB) by 11 M LiCl--0.02 M HCl or 9 M LiCl--1 N $AlCl_3$. Batch rate tests were made in order to establish the kinetics of the scrubbing cycle. A small-scale (0.75-in.-ID) pulse column was used as the contacting equipment for these continuous tests. Table 3.7 lists physical properties of the solutions tested.

Batch Rate Tests

Batch rate tests were made to determine the rate constant (ka) of the scrubbing cycle. The test equipment consisted of a 6-in.-diam x

12-in.-high baffled-tank mixer equipped with a 3-in.-diam six-blade turbine. Samples were taken at timed intervals during the tests.

Table 3.7 Physical Properties

Solution	Specific Gravity (g/cc at 50°C)	Viscosity (centipoise at 50°C)
30% Alamine 336 in DEB	0.848	2.67
11 M LiCl--0.02 M HCl	1.233	3.34
9 M LiCl--1 N $AlCl_3$	1.220	3.45

The data in Table 3.8 were plotted, and the batch scrub rates (ka) calculated.[9] Increasing the solution temperature at a constant power input (about 2.5 hp per 1000 gal of aqueous-continuous operation) from 25 to 50 to 70°C gave rate constants of 0.100, 0.460, and 0.766 min^{-1}, respectively.

Organic-continuous operation (at 50°C, 2.5 hp per 1000 gal) resulted in a slightly lower ka of 0.230 min^{-1}. The plot (Fig. 3.15) of log ka vs the reciprocal of the temperature is a straight line. An increase in power input from 2.5 to 220 hp per 1000 gal (at 50°C, aqueous-continuous operation) increased ka from 0.460 to 13.8 min^{-1}. Satisfactory operation should be achieved at elevated temperature and within the power input range normal for pulse column operation.

Continuous Runs in a Pulsed Column

Europium scrubbing test runs were made using a glass pulse column (0.74 in. in inner diameter and 24 in. long) as the contactor. The column configurations tested were: 1/4 in. procelain Berl-saddle packing, stainless steel sieve plates (1/32 in. holes, 5% free area, spaced 1/4 in. apart), and tantalum sieve plates (1/16 in. holes, 5% free area, spaced 1/4 in. or 1/8-1/8-1/2 in. apart). The column was pulsed with a micro-bellows pump to give either a truncated pulse wave or a sinusoidal pulse wave. The

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[9] A. D. Ryon, F. L. Daley, and R. S. Lowrie, Design and Scaleup of Mixer Settler for the Dapex Solvent Extraction Process, ORNL-2951 (Sept. 15, 1960).

Table 3.8. Batch Rate Data for Europium Scrubbing Cycle

Aqueous Feed: 9 M LiCl - 1 N $AlCl_3$
Organic Feed: 30% Alamine 336 in DEB Containing Eu Tracer
Phase Ratio A/O: 1/1
Continuous Phase: Aqueous

Run No.	Solution Temp. (°C)	Power Input (hp/1000 gal)	Eu Concentration in Organic Phase, Counts min^{-1} ml^{-1}											Ka min^{-1}
			0 sec	5	10	20	30	45	60	120	240	480	960	
39	25	2.5	$8.8\text{x}10^4$	$(7.6\text{x}10^4)^a$	$(7.5\text{x}10^4)^a$	$(7.2\text{x}10^4)^a$	$(6.9\text{x}10^4)^a$	$(6.6\text{x}10^4)^a$	$(6.3\text{x}10^4)^a$	$(5.4\text{x}10^4)^a$	$(4.3\text{x}10^4)^a$	$(3.1\text{x}10^4)^a$	$(2.1\text{x}10^4)^a$	0.100
52	50	2.5	$1.4\text{x}10^5$	$6.7\text{x}10^4$	$6.3\text{x}10^4$	$5.2\text{x}10^4$	$4.6\text{x}10^4$	$3.9\text{x}10^4$	$3.0\text{x}10^4$	$1.4\text{x}10^4$	$5.5\text{x}10^3$	$3.5\text{x}10^3$	$2.6\text{x}10^3$	0.462
53	75	2.5	$1.1\text{x}10^5$	$4.7\text{x}10^4$	$4.6\text{x}10^4$	$3.5\text{x}10^4$	$2.6\text{x}10^4$	$1.9\text{x}10^4$	$1.3\text{x}10^4$	$3.7\text{x}10^3$	$1.8\text{x}10^3$	$1.3\text{x}10^3$	$1.1\text{x}10^3$	0.766
40[b]	50	2.5	$6.6\text{x}10^4$	$(5.3\text{x}10^4)^a$	$(5.1\text{x}10^4)^a$	$(4.7\text{x}10^4)^a$	$(4.4\text{x}10^4)^a$	$(3.9\text{x}10^4)^a$	$(3.6\text{x}10^4)^a$	$(2.8\text{x}10^4)^a$	$(1.2\text{x}10^4)^a$	$(7.8\text{x}10^3)^a$	$(5.7\text{x}10^3)^a$	0.230
50	50	220	$3.6\text{x}10^5$	$7.4\text{x}10^3$	$4.0\text{x}10^3$		$4.1\text{x}10^3$	$3.7\text{x}10^3$	$3.2\text{x}10^3$	$3.2\text{x}10^3$	$2.7\text{x}10^3$	$2.7\text{x}10^3$	$2.3\text{x}10^3$	13.8

[a] Numbers in parentheses were calculated by use of material balance from analysis of aqueous phase.

[b] Organic continuous phase instead of aqueous continuous.

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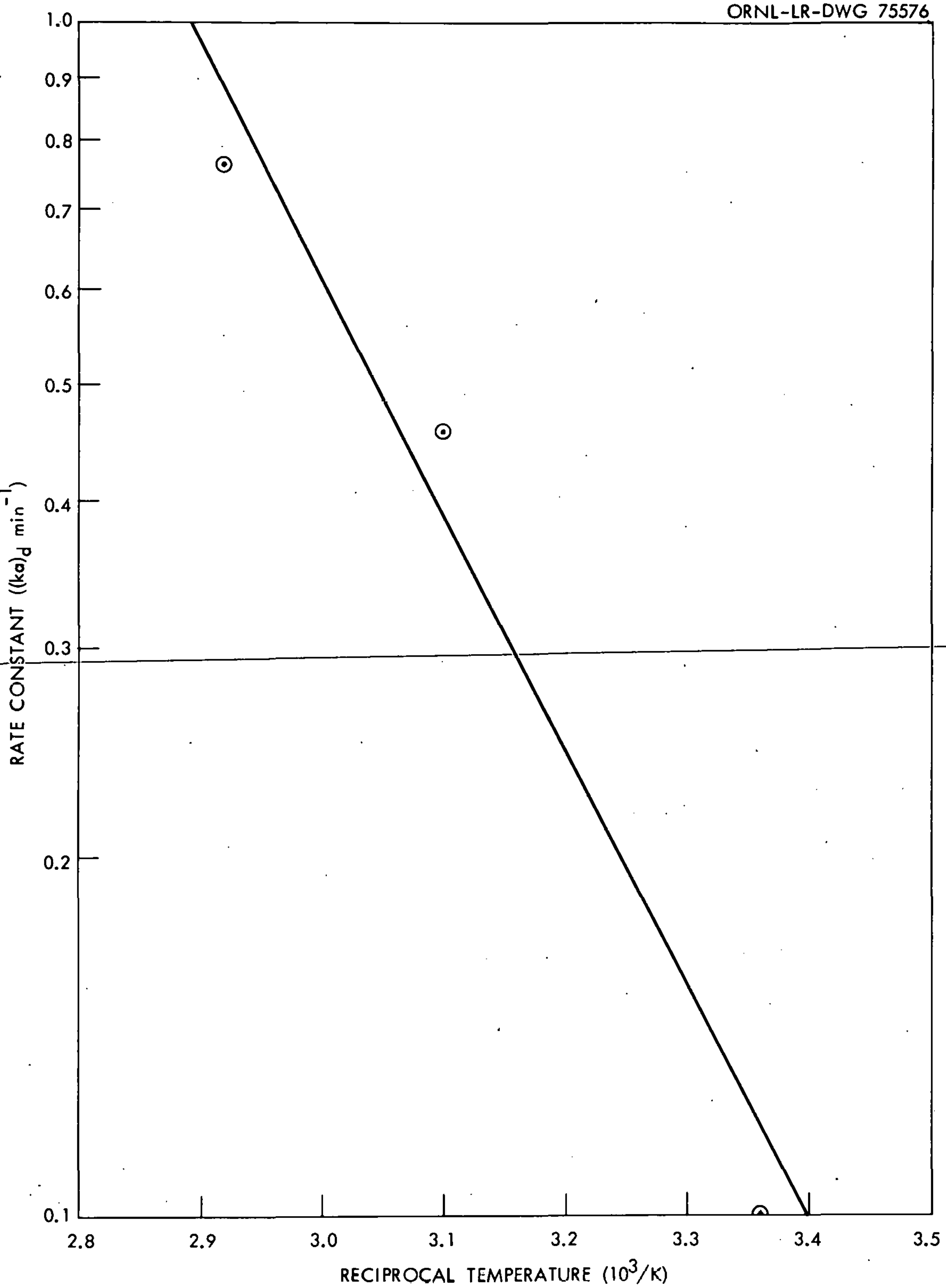

Fig. 3.15. Effect of Temperature on Batch Scrubbing Rate.

aqueous-organic interface was controlled at the top of the column by use of an air-bubbler-controlled pressure pot in the aqueous discharge stream.

Several unsuccessful runs were made in the pulse column packed with Berl saddles or stainless steel sieve plates using 11 M LiCl--0.02 M HCl as the scrubbing agent. Inability to control the acid concentration of the scrub solution during column operation resulted in a variable distribution coefficient, and no significant data were obtained. Tantalum sieve plates were installed in the column, all stainless steel in the system was replaced with plastic and a series of runs made using 9 M LiCl--1 N $AlCl_3$ as the scrubbing agent. Table 3.9 contains the data from these runs. The run conditions were set to give operation near flooding in the column. Operational variables tested were: pulse frequency, 50 to 240 cycles per minute; pulse amplitude, 0.20 to 0.40 in.; total flow (A/O = 1/3), 12 to 32 ml/min; percentage of organic holdup in column, 16 to 35%; truncated or sinusoidal pulse wave; plate spacing; and column temperature, 50 to 70°C. Column HETS' were calculated by use of a McCabe-Thiele diagram for each of the runs (Table 3.9). No marked trend in HETS values due to any of the operational variables was found, the average HETS was 24 in.

3.2.4 Target Irradiation -- (S. D. Clinton)

Calculations for irradiating three prototype HFIR targets in the MTR were completed and sent to Idaho for approval. A peak unperturbed flux of 6.4×10^{21} neutrons/cm^2 was requested for the three rods. This time-integrated flux can be obtained after 441 days by moving the targets to higher fluxes three times during the irradiation (desired peak neutron flux will vary between 1.8 and 4.4×10^{14}). The average burnup of the targets should be about 760 kw-days per initial gram of Pu^{239}. Assuming 100% release of fission product gases, the maximum pressure inside the capsule should be 750 psia, which will create a stress of 2900 psi at the end-plug welds. Other properties of the proposed HFIR target irradiations in the MTR are as follows:

Table 3.9. Europium Scrub Data for the Pulsed Column

Organic:	30% Alamine 336 in DEB; europium tracer
Aqueous:	9 M LiCl--1 N $AlCl_3$
A/O:	1/3
Sieve Plate:	Spacing 1/4 in.; 1/6 in. diam holes; 5% free area
Column Temperature:	50°C
Pulse Amplitude:	0.20 inch

				Eu Concentration Counts min^{-1} ml^{-1}							
				Continuous-Run Data			Equilibrium Data				
Pulse Frequency (CPM)	Type of Pulse Wave	Total Throughput (ml/min)	% Organic Holdup	Organic Feed	Organic Effluent	Aqueous Effluent	Aqueous	Organic	D_a^o	HETS (in.)	Run No.
45	Truncated	17	28	$2.5x10^5$	$1.7x10^4$	$5.5x10^5$	$7.0x10^5$ $2.6x10^5$	$8.4x10^3$ $1.7x10^3$	0.012 0.006	25	54
50	Truncated	20	28	$3.2x10^4$	$4.5x10^3$	$3.0x10^4$	$9.2x10^4$ $3.0x10^4$	$1.7x10^3$ $1.0x10^3$	0.018 0.030	27	36
60	Truncated	12	16	$1.1x10^5$	$5.5x10^3$	$2.5x10^5$	$3.1x10^5$ $9.9x10^4$	$3.0x10^3$ 626	0.010 0.006	25	44
70	Truncated	16	20	$1.3x10^5$	$1.5x10^4$	$2.7x10^5$	$3.6x10^5$ $1.2x10^5$	$3.5x10^3$ $6.6x10^3$	0.010 0.055	26	37
90	Truncated	12	17	$2.3x10^5$	$3.1x10^3$	$7.2x10^5$	$7.7x10^5$ $2.6x10^5$	$7.1x10^3$ $2.3x10^3$	0.009 0.009	17	49
90	Truncated	12	20	$3.5x10^5$	$2.1x10^4$	$8.7x10^5$	$1.0x10^6$ $3.5x10^5$	$7.2x10^4$ $2.9x10^3$	0.007 0.008	25	55
100	Sinusiodal	12	20	$1.3x10^5$	$3.5x10^3$	$4.0x10^5$	$3.8x10^5$ $1.3x10^5$	$3.3x10^2$ $2.2x10^2$	0.0008 0.002	24	48
160	Sinusiodal	16	22	$1.6x10^5$	$1.5x10^4$	$3.8x10^5$	$4.8x10^5$ $1.6x10^5$	$3.0x10^3$ $1.1x10^3$	0.006 0.007	26	45
200	Truncated	12	24	$1.2x10^5$	$5.0x10^3$	$2.9x10^5$	$3.4x10^5$ $1.1x10^4$	$3.9x10^3$ $1.2x10^3$	0.012 0.011	24	35
200	Truncated	24	20	$8.6x10^4$	$2.8x10^4$	$1.9x10^5$	$2.3x10^5$ $8.2x10^4$	$9.2x10^2$ $4.0x10^2$	0.003 0.005	33	41
220	Sinusoidal	13	16	$1.5x10^5$	$1.4x10^4$	$3.4x10^5$	$4.2x10^5$ $1.5x10^5$	$1.5x10^4$ $3.3x10^4$	0.036 0.023	25	56
240	Sinusoidal	20	35	$2.3x10^5$	$6.8x10^3$	$7.0x10^5$	$6.5x10^5$ $2.2x10^5$	$9.1x10^3$ $2.4x10^3$	0.014 0.011	16	46
240	Sinusoidal	32	20	$3.4x10^5$	$1.2x10^4$	$8.8x10^5$	$9.5x10^5$ $3.7x10^5$	$1.4x10^4$ $2.8x10^3$	0.014 0.008	22	47

Condition	Surface Heat Flux (Btu hr^{-1} ft^{-2})	Surface Temp. (°F)	Central Temp. (°F)
Average	593,000	206	1066
Peak	652,000	221	1167
Hot spot	950,000	267[a]	--

[a] The saturated steam temp. at the bottom edge of the fueled section (coolant flow is downward) is 270°F.

3.2.5 Evaluation of Protective Coatings -- (G. A. West)

In radiation-damage tests, the upper tolerable limit of several protective coatings in air and deionized water toward Co^{60} gamma radiation, at about 1 x 10^6 r/hr and 40 to 50°C, was determined as shown in decreasing order of resistance. Tests are continuing on five coatings in water and 34 coatings in air. Results to date are shown in Table 3.10.

Table 3.10. Results of Radiation-Damage Tests of Protective Coatings

Coating	Recommended Maximum Exposure (r)	Failure Occurred (r)
Fiberglas Reinforced Coatings in Deionized Water		
Amercoat 76-66, epoxy	~3 x 10^9	(test not complete)
P.P.G.Co., UC9647, epoxy	~1 x 10^9	2.3 x 10^9
P.P.G.Co., UC12168, epoxy	~1 x 10^9	2.0 x 10^9
Ceilcote, 505.2, epoxy	(test not complete)	
Amercoat 74 Surfacer, 74 seal, epoxy	~1 x 10^8	2.5 x 10^8
P.P.G.Co., UC 9647-UL8134, epoxy-vinyl	~1 x 10^8	2.4 x 10^8
Plain Coatings in Deionized Water		
Amercoat 66, epoxy	3 x 10^9	(test not complete)
Sika, Colma protective coating, epoxy	2 x 10^9	(test not complete)
Wisconsin Prot. Ct. Co., Plasite 7155, phenolic	2 x 10^9	3.8 x 10^9
Carboline, Phenoline 368, phenolic	1.5 x 10^9	2.7 x 10^9

Coating	Recommended Maximum Exposure (r)	Failure Occurred (r)
Wisconsin Prot. Ct. Co., Plasite 7122, phenolic	1.5×10^9	2.7×10^9
Carboline, Phenoline 300, phenolic	1×10^9	2.1×10^9
Carboline, Phenoline 305, phenolic	1×10^9	2.1×10^9
Sika, Colma Surface Kote, epoxy	1×10^9	2×10^9
Wisconsin Prot. Ct. Co., Plasite 7100, phenolic	8×10^8	1.2×10^9
Wisconsin Prot. Ct. Co., Plasite 9009, epoxy	8×10^8	1.2×10^9
Wilbur and Williams, Tile Cote, epoxy	6×10^8	1×10^9
Amercoat 74 Surfacer, 74 seal, epoxy	5×10^8	9×10^8
Amercoat 1680, inorganic	$>3 \times 10^9$	(test not complete)
Coatings in Air		
10 coatings have resistance	$>4.5 \times 10^9$	(test not complete)
16 coatings have resistance	$>3 \times 10^9$	(test not complete)
14 coatings have resistance from	5×10^8	test complete
to	2.8×10^9	

4. DEVELOPMENT OF FABRICATION PROCESS FOR HFIR TARGET ELEMENTS

D. A. Douglas

4.1 Discussion of Problem Areas

D. A. Douglas

During the past quarter, the unsuccessful attempt to fabricate irradiation capsules using glove box methods confirmed the desirability of re-examining the fabrication procedures that were providing the criteria for equipment development. There appears to be three main problem areas. One concerns the production of pellets with the required thermal conductivity. A second is the need for end closures that provide both a leak seal and adequate strength. The third is the development of a collapsing procedure capable of producing a separation between pellet and clad of less than 0.5 mil. It is possible that the solution of one or more of these problems will require a new approach rather than further development of the present approach.

4.1.1 Pellet Preparation

Equipment capable of remote operation has been successfully developed for calcining, blending, and metering the powders. The pressing procedures produce pellets of the desired density. Still unresolved is the problem of achieving a continuous aluminum matrix to provide the necessary thermal conductivity. Also lacking is a method for determining the thermal conductivity nondestructively in a glove box.

Obtaining the continuous metal matrix is made difficult because the average oxide particle is 6 μ in size, compared with 19 μ for the aluminum powder. Also, the aluminum powders tend to be very fluffy and difficult to handle. Several new approaches are under study, which may result in a better quality and a more reproducible pellet structure.

It would be highly desirable to be able to evaluate the thermal conductivity of each pellet as it is produced. Several methods, some direct and some indirect, for achieving this have been conceived. Work is in progress to explore autoradiographic, eddy current, electrical resistance, and heat flow as possible methods. No results are presently available.

4.1.2 End Closures for Capsules

The high cost of the production plant and the value of the product make it imperative that the target capsule not fail. It must sustain hydraulic loads, thermal stresses, and fission-gas pressure. The regions of greatest concern in this regard are the two end closures. To date, fusion welding has been tried in order to provide the necessary integrity and strength. By using short-arc techniques and water-cooled chill blocks, porosity-free welds have been obtained. The fact that only 50% of the wall thickness was fused during this operation renders such joints unacceptable. When greater pentrations are attempted, blowout or gross porosity results. It has been decided that consistent reliable welds can not be made according to the present concepts. The end-closure problem is being studied to determine which of several other methods is the most attractive. The use of filler metal and combinations of mechanical attachment, brazing, and welding are being considered. A mechanical test has been devised to test the efficiencies of the joints being made by various procedures.

4.1.3 Tube-Collapse Studies

The general problem of extracting heat from the pellets at a rate that will prevent melting of the aluminum requires not only high thermal conductivity in the pellets but also intimate contact between the pellet and the capsule wall. Some calculations have indicated that a separation no greater than 0.1 mil is required. The validity of this calculation can be questioned because of the unknowns, for which assumptions had to be made. If the calculations are correct, however, then some procedures superior to the present one are needed. Metallographic examination of sections of tubes collapsed under hydraulic pressures of 12,000 to 30,000 lb revealed separations of 0.5 to 1.0 mil. Some preliminary experiments using "Magneform" equipment at the Oak Ridge Gaseous Diffusion Plant indicate that more uniform collapse and less separation is obtained Some experiments to determine the benefits to be derived from warm collapse are under way.

4.2 Development of Equipment for Fabricating Target Elements for TRU Facility

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

The basic criteria established for fabricating the target elements has changed little from that outlined in the first quarterly progress report.[10] The following gives the status of equipment development in the three cubicles of the TRU facility that are assigned for target fabrication.

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[10] D. E. Ferguson, Transuranium Quarterly Progress Report for Period Ending February 28, 1962, ORNL-3290.

4.2.1 Cubicle 3

The process flowsheet and equipment layout for cubicle 3 have not changed. Conceptual designs on three pieces of equipment for the cubicle were issued for comment and have been approved. Detail design was started or completed on eight items of equipment for cubicle 3, and final design approval was given on four of them. Fabrication is underway on one item of equipment, and three units are complete and are being tested and evaluated.

The status of the equipment for cubicle 3 follows:

Actinide Calciner. -- Although criteria have been established, the actinide calciner has not been conceptually designed. Design will start next quarter.

Batch Scale. -- Initiation of design of the batch scale is awaiting the successful operation of the pellet-load scale discussed below.

Blender-Dispenser. -- The conceptual design of the blender-dispenser, shown in Fig. 4.1, was approved, and final design details were completed and are verified. A mockup, which proved the concept to be feasible, was assembled. The mockup indicated that a feed rate of 0.5 g/min, which is easily obtained, will be slow enough to enable accurate dispensing of the powder mixture to the pellet-load scale.

Die Scale. -- Preliminary design and machine work incidental to the testing and evaluation of the torsion balance for weighing the pellet loading is 85% complete. In addition to the above investigation, a commercial source has been invited to present a proposal for equipment that will weigh the pellet loading and control the powder-dispensing rate.

Cap-Powder Loader. -- The final design of the loader is complete, but the drawings have not yet been approved for construction.

Die Assembly Feeding Station. -- Fabrication is complete, and preliminary tests indicate design to be satisfactory. Complete bench test and evaluation of the design were started.

Pellet Press. -- Fabrication of the pellet press (see Fig. 4.2) and installation of piping, pumps, and instrumentation is complete. Bench

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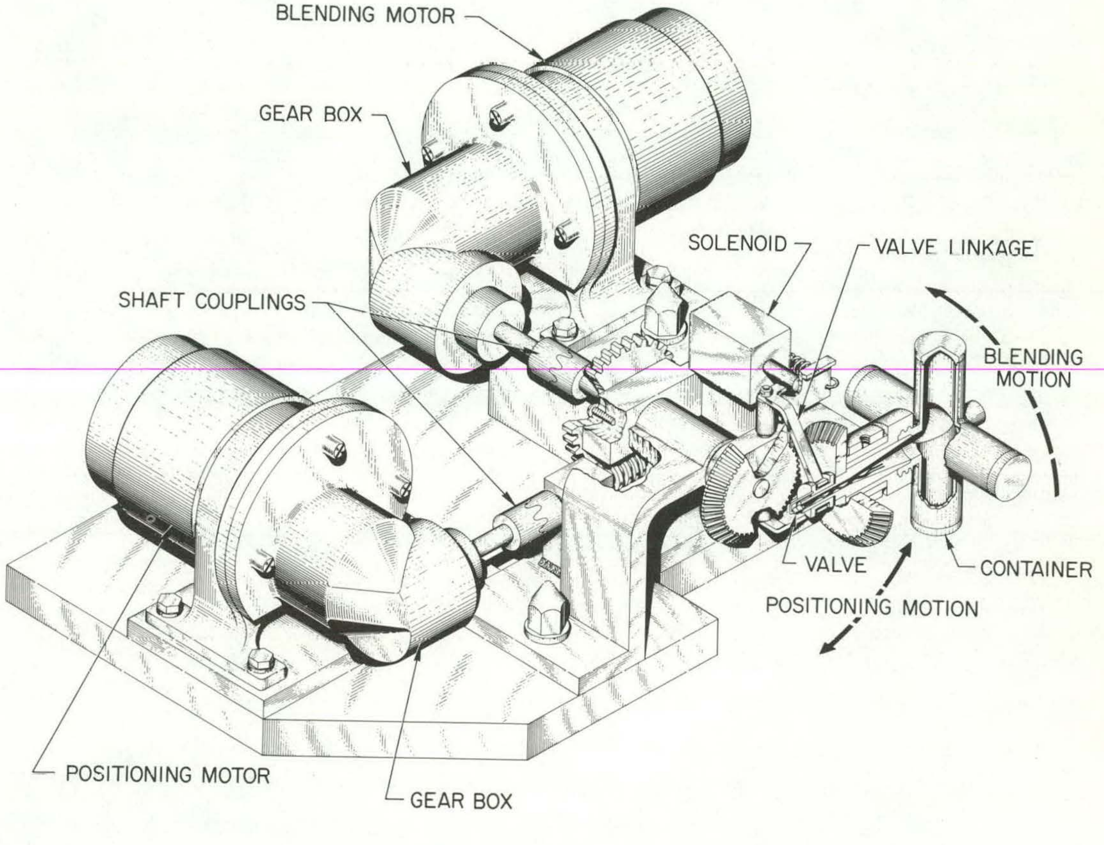

Fig. 4.1. Blender Dispenser.

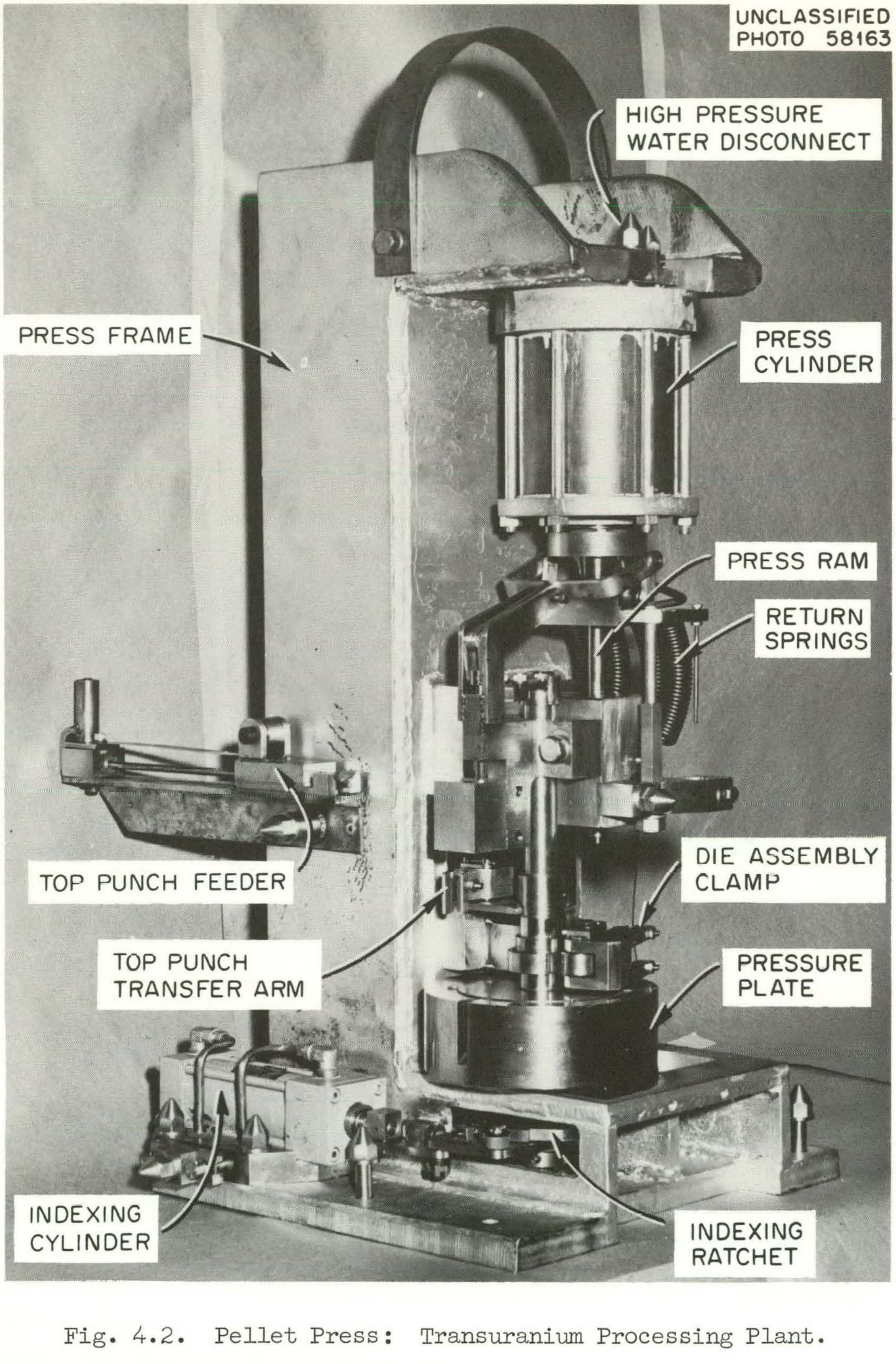

Fig. 4.2. Pellet Press: Transuranium Processing Plant.

tests of the equipment have started, and some minor modifications were made to ensure its satisfactory operation. To date, about 75 pellets have been pressed at pressures varying from 30 to 50 tons. The press operation is now reliable enough that pellet-pressing data can be taken to establish the best operating parameters.

Pellet Ultrasonic Cleaner and Pellet Dryer. -- Final design details of both the pellet ultrasonic cleaner (Fig. 4.3) and the pellet dryer are complete but have not been approved for fabrication. The dryer is very similar in design to the ultrasonic cleaner. Tests of the cleaner and pellet transfer device, which are being procured from outside vendors, are to be conducted to determine if the concept is workable. At this writing, the pellet transfer track has been received, and preliminary tests were satisfactory. As soon as both units are tested together satisfactorily, the complete design will be released for fabrication.

Transfer Arm III. -- Detail design of the cubicle 3 transfer arm is complete, and the drawings are being checked for correctness before being issued for approval.

Miscellaneous Containers. -- Containers for transporting punch and die assemblies, top punches, and their feed magazines have been made. These containers will also be used to collect and dispose of used equipment. The pellet transfer container, which is used to collect the fabricated pellets at the pellet press and transport them by manipulator to the ultrasonic cleaner, has been made. Test and evaluation of this equipment have just started.

4.2.2 Cubicle 2

The process flowsheet and equipment layout for cubicle 2 has not changed. Conceptual design on two additional pieces of equipment was approved and detail design has started. Fabrication was completed on one item for cubicle 2 and started on another major equipment group.

The equipment status for the cubicle follows:

Pellet Inspection and Loading Equipment. -- The pellet inspection and loading equipment (Fig. 4.4), which consists of a pellet diameter

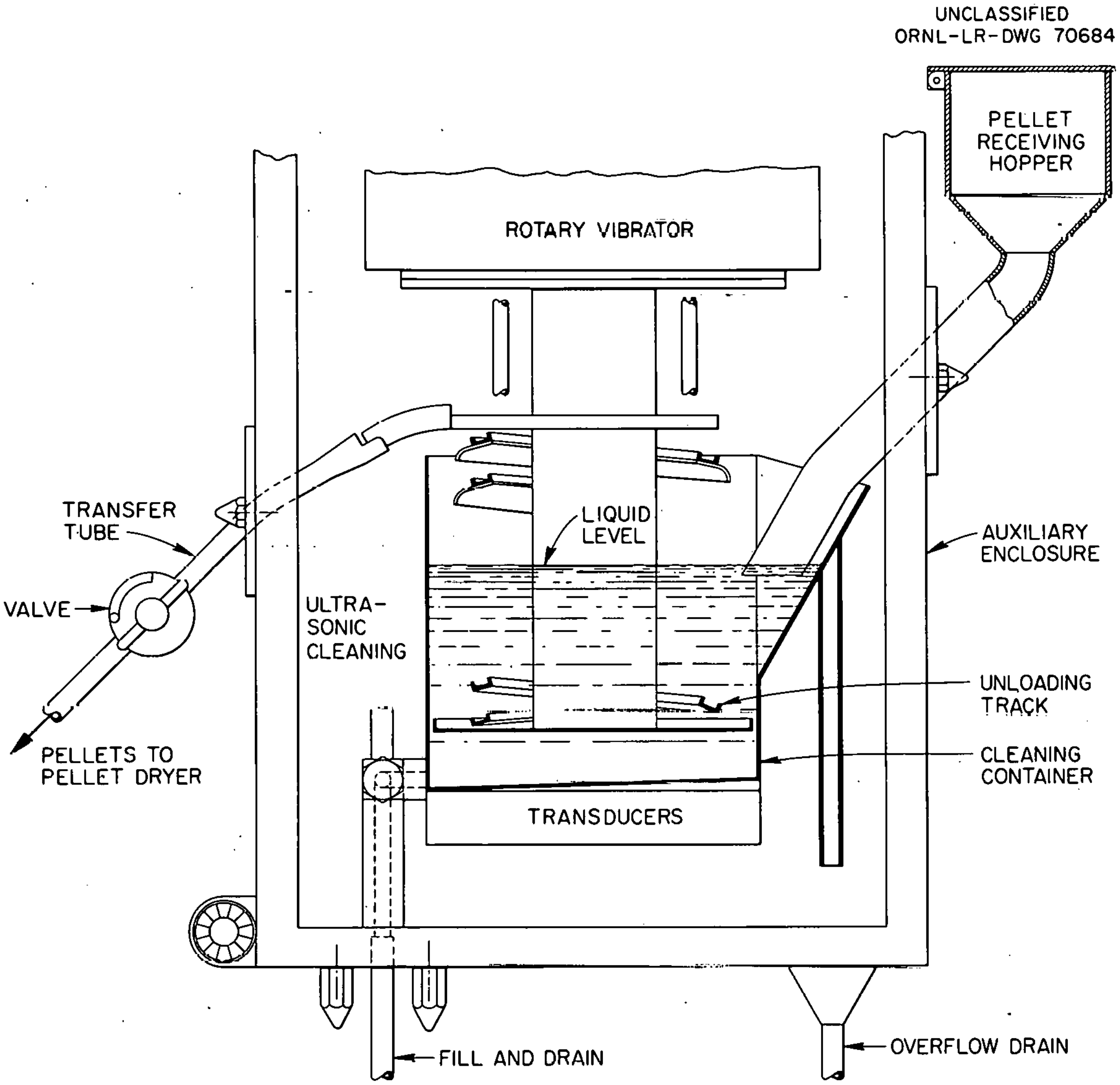

Fig. 4.3. Ultrasonic Cleaner for Pellets.

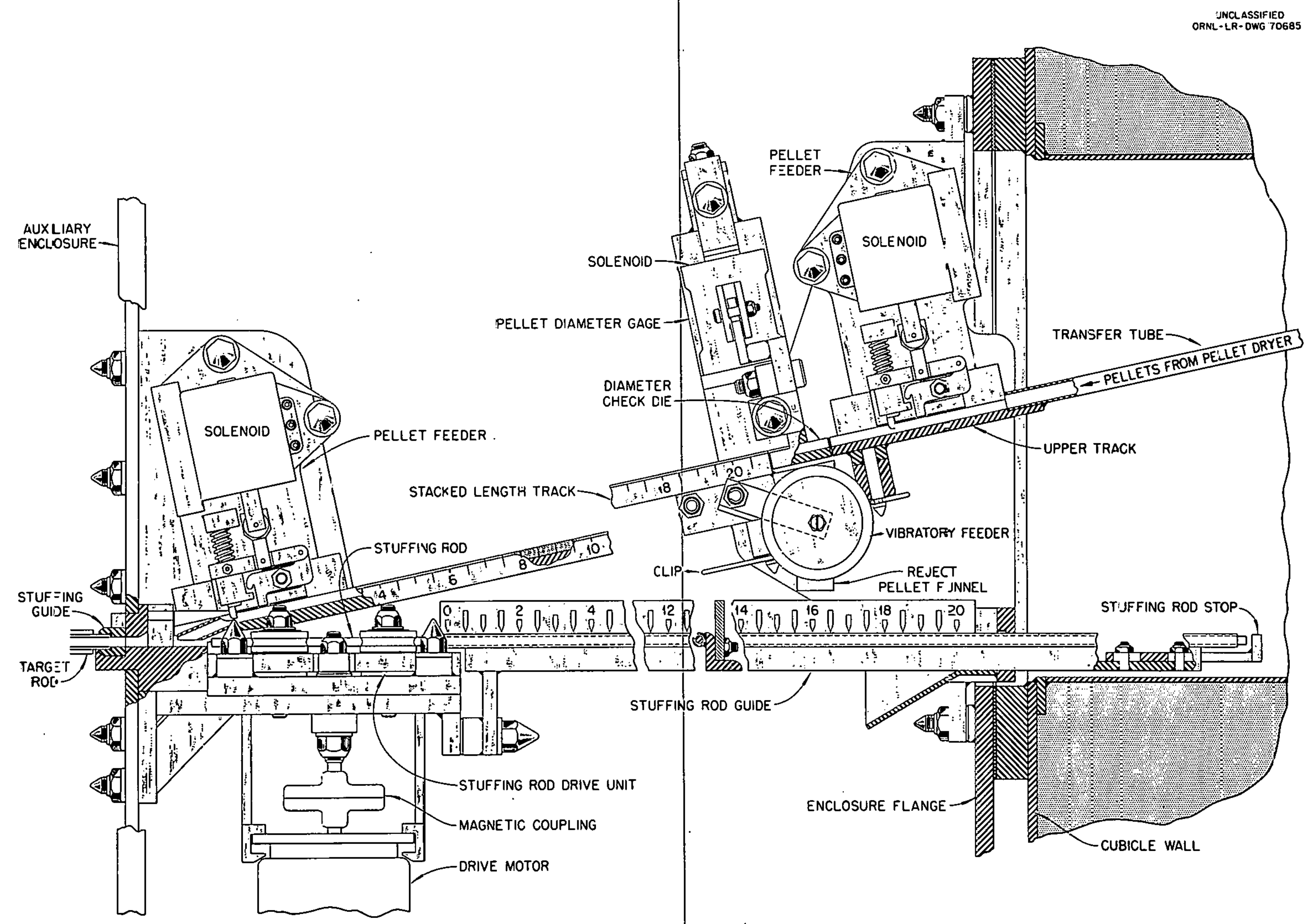

Fig. 4.4. Pellet Inspection and Loading Equipment.

gate, stacked-pellet length gage, two pellet feeders, target-tube loader, and the auxiliary enclosure for the above equipment, was approved and fabrication has started.

End-Cap Welding Machine. -- Conceptual design of the end-cap welding machine is being delayed until a suitable and reliable welding procedure can be established.

Helium-Leak-Test System. -- Procurement, fabrication, and assembly of equipment for test and evaluation of the proposed helium-leak-test system is almost complete, and some tests have been made to substantiate the proposed system.

In the proposed system, the target-rod leak-test chamber, located in cubicle 2, will have two evacuation lines (equipped with absolute filters) penetrating the cell walls to the chemical makeup area where they terminate at the evacuation manifold. The helium mass-spectrometer type of leak detector and test manifold are to be locally operated and controlled outside the area of intense radiation. This eliminates the need for the remotely operated leak detector originally proposed. The sensitivity requirement of the leak detector system is 1.0×10^{-8} std cc/sec. This acceptable size leak in HFIR targets is based on the calculated amount of fission-product-gas escape from the target during the irradiation and charging operations as well as the possible leakage of water into the target during the two-year operation in the reactor.

Components of the system have been undergoing tests, and the data show no evidence of helium holdup in an 8-in., round, absolute filter made by Flanders Filters, Inc. A smaller 4-in.-diam cylindrical absolute filter recently designed is being fabricated. The time response of the system will be improved through the use of the smaller-volume filters.

A prototype evacuation and leak-test manifold was designed and built. Performance tests based on the leak-test sensitivity criteria were completed on this prototype manifold, using an interconnecting sample line to the target-rod test chamber. The sampling line (5/8-in.-OD tubing) was 50 ft long. Signal-stabilization times of 15 to 20 min were obtained for leak rates ranging from 4×10^{-8} to 1.5×10^{-6} std cc/sec of helium.

The air-operated, electrically controlled vacuum valves in the prototype manifold performed satisfactorily with seat and stem leak rates of less than 1.0×10^{-10} std cc/sec (helium). As a result of these tests, the prototype manifold design can be simplified, eliminating eight of the valves and replacing the expansion type sampling method with a dynamic flow system. This reduces the number of operator manipulations necessary for the operation of the leak-test manifold. The simplified flow schematic of the revised system is shown in Fig. 4.5.

Design criteria demands vacuum-tight molecular seals at the sealing cap of the target-rod leak-test chamber and at the absolute filters so that replacement can be easily made by a remotely operated manipulator. These seals must be consistently leaktight to better than 1.0×10^{-9} std cc/sec. All-metal seals are preferred, but certain elastomers with a low rate of radiation deterioration may be acceptable. A dual-seal design with a nitrogen-gas buffer zone to eliminate possible helium inleakage from normal atmospheric air will be tested first.

A recently designed and completed tubing-disconnect fitting (utilizing the above concept) with two concentric butyl O-rings was tested. Preliminary data yielded leak rates from 1.0×10^{-10} to 1.8×10^{-9} std cc/sec. A single-bolt U-frame clamp seals the mating gasket surface. Such a fitting may be used for tubing terminations of the absolute filter so that the assembly can be easily replaced.

Hydrostatic Collapse Equipment. -- Fabrication of the hydrostatic pressure chamber is being deferred until more data are obtained on the pressures required to give an effective collapse of the tube to the pellets. Investigation has started on the use of magnetic-pulse forming (Magneforming) as a possible solution to the problems encountered when hydrostatic pressure is used to effect the collapse. To date, the problem has been the incomplete collapse over the pellet area unless extremely high pressures (25,000 to 30,000 psi) are utilized. Pressures of these magnitudes collapse the sleeve that supports the end gas plenum. Type 7075-T6 aluminum, which has a yield strength of 75,000 psi, has been used as support-sleeve material. Magnetic-pulse forming has the principal advantage over the use of hydrostatic pressures in that the area to be

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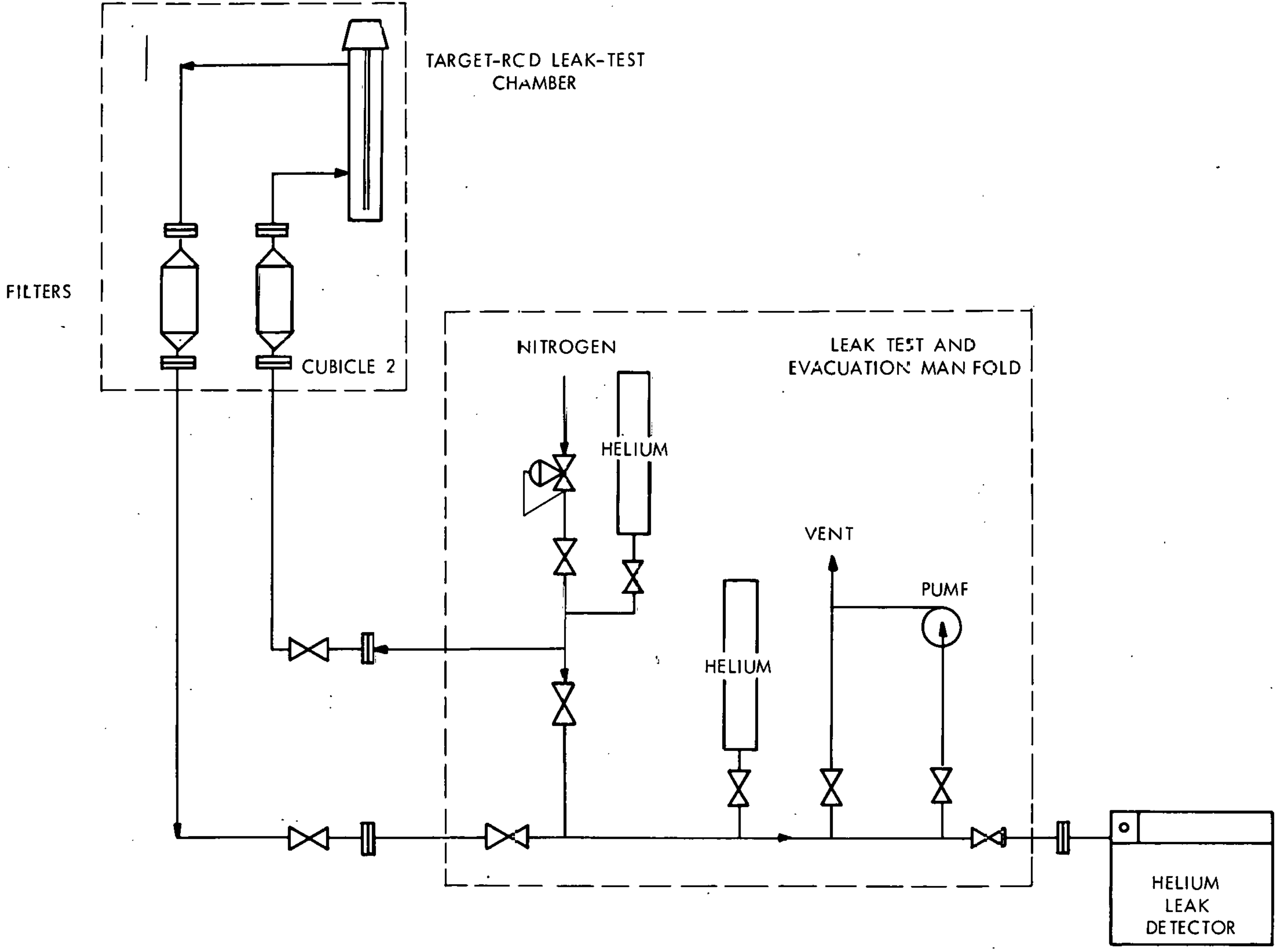

Fig. 4.5. Target-Rod Leak-Test System.

collapsed can be selected. The gas-plenum support liner, therefore, could be eliminated if this method is selected.

Target Ultrasonic Cleaner. -- The concept for the ultrasonic cleaner has been changed since the previous report was issued. That report stated that the unit, including transfer devices, was to be enclosed in a large auxiliary enclosure. The idea now is to place the target rod into a small cylindrical container that will act as an auxiliary enclosure. This unit containing the rod will be placed into the ultrasonic cleaner and flooded with solution for cleaning the rod. The new approach has been approved, and detail design has started.

Target Transfer Arm II. -- Conceptual design of the target-transfer arm awaits the establishment of more information on the process equipment in the cell and a firm decision on the configuration of the target rod itself. The transfer arm, however, will have rotary, horizontal, and vertical motions, which will be powered by electric motors and positioned by positive stops for the various positions.

Target Transfer to Cubicle 1. -- The concept for transferring the target rod through the wall to cubicle 1 has been changed to eliminate a large auxiliary enclosure and a complicated mechanism for effecting the transfer. Now the idea is to place the target rod into a cylindrical container having a dust-tight top and bottom and internal guide rollers. This container, as previously discussed, is placed into the ultrasonic cleaner, the target rod is cleaned, than the rod is transported to a rotating fixture where the container is rotated 110°. The top lid is opened and the target rod is allowed to roll through the wall to a receiving fixture in cubicle 1, where it is rotated back to the vertical position for transporting to other operations. This concept was approved, and detail design is proceeding.

4.2.3 Cubicle 1

The process flowsheet and equipment layout for cubicle 1 has not changed. Conceptual designs for the equipment have not been further expanded because the target-rod design has not been firmly established.

4.3 Development of Equipment for Fabricating Plutonium Targets

A. L. Lotts

The plans for equipment and procedures for the fabrication of plutonium-bearing HFIR target elements have remained essentially unchanged from those previously reported. The schedule has changed so that the required operational date is now November 1963. Some equipment has been procured and tested; other items are being constructed. A summary of the present status of equipment is given below.

Glove Boxes. -- Seven commercial glove boxes were ordered. Two have been received, and the others are expected to be delivered shortly.

Powder-Preparation Equipment. -- The status of the powder-preparation equipment remains essentially the same; that is, most of the items have been obtained, and the only outstanding item is the blender. It appears that if a blender is used it must be specially constructed because no commercial source has yet been found.

Pellet-Preparation Equipment. -- The 10-ton press, consisting of a frame and an air-driven pumping system, was assembled, and it performed satisfactorily in tests in which dummy pellets were made. In addition, a pressing base, which allows convenient and safe pressing and ejection of pellets, was constructed and tested. The base was used in pressing pellets for the HFIR target prototypes discussed in Sec 4.4. Since this experience showed that a number of improvements are desirable, a new improved base was designed and is now being built.

Tube-Closure Equipment. -- Assembly of the tube-closure equipment was completed. The equipment consists of components for evacuation of the target tube, back-filling of the tube with helium, placement of the tube end cap, and tungsten-inert-gas welding of the tube-cap joint. A lathe bed holds the assembled element in the horizontal position, and the element is rotated by a motor-driven chuck.

The control circuit provided allows either automatic or manual welding. In the automatic mode, the following is the sequence of operation:

1. The current to be used is preset on the direct-current arc welder.
2. The tube is inserted into the chuck and the tailstock, and evacuation, helium backfilling, and end-cap placement are accomplished manually.
3. The welding torch is set in place.
4. The welding sequence is started by one control switch. Upon starting, all operations proceed automatically. When the tube begins its rotation, the arc is struck, and the current is feathered to the maximum in about an eighth of a revolution. Before striking the arc, argon flow is started, and upon striking the arc, the gas is switched to helium. Welding at maximum current proceeds until one revolution is made, and the current is then feathered to zero through about an eighth of a revolution. The rotation of the rod is then reversed, and the circuit is reset for the next welding sequence.

A number of welds, including those on the HFIR prototypes discussed in Sec 4.4 were made with the machine. The control circuitry and drive components performed satisfactorily; however, several auxiliary components were unsatisfactory--principally the welding-torch holder and heat sink. A stage, which rigidly holds the torch and allows adjustment in two directions, must be built. In addition, a more efficient heat sink must be developed.

The heat sink is required for the removal of heat from the tube during welding so as to minimize the expansion of gas in the tube and, consequently, preclude the blowout of the weld. The heat sink used in cold tests of the welding unit was water cooled, made of brass, and stationary. The tube rotated in a hole provided in the heat sink, and welding was done through a small hole in the top of the heat sink. Although the water-cooled heat sink performed more satisfactorily than simple copper chill rings, it has disadvantages in that visual observation during welding is poor, and alignment to prevent scoring of the tube is difficult to maintain. Recently, other heat sinks were constructed of aluminum, but they have not yet been tried. Since they are massive and rotate with the tube, they are supported from the lathe bed by rollers.

Hydrostatic-Collapse Equipment. -- The hydrostatic collapse equipment is now being built. It consists of a chamber and pump which are designed to operate at a maximum pressure of 20,000 psi.

Helium-Leak-Test Equipment. -- The helium-leak-test chamber, which will be attached to one of the glove boxes, is now being built. In addition, another chamber with an absolute filter was assembled for conducting leak tests on the HFIR prototypes. This assembly and the leak detector performed satsifactorily.

Mockup of Equipment for Fabrication of Plutonium Target. -- During this reporting period, the design work required for the preparation of a portion of the storage area of Building 4508 for the mockup of the equipment for plutonium-target fabrication was accomplished. Because of the nonavailability of an alpha laboratory sufficiently far in advance of the required date for the commencement of hot operations, it is planned to install and test a complete plutonium-target fabrication line (using stand-in oxides for PuO_2) in this space, under simulated conditions. In this manner, the equipment can be optimized prior to its installation in the alpha laboratory, thus minimizing the time required in the alpha laboratory before starting hot operations.

4.4 Fabrication of HFIR Target Irradiation Prototype

A. L. Lotts S. A. Rabin

The fabrication of four prototype HFIR target elements containing Pu^{239} in lieu of Pu^{242} or the higher actinide elements was attempted. Fabrication methods proposed for the HFIR target elements were used wherever possible. Because of a multitude of difficulties and some unexpected problems, none of the prototype elements was successfully fabricated. The design of the prototypes was given in the previous quarterly report.[11] Three of the prototypes were to have been irradiated in the Materials Testing Reactor (MTR), while one was to have been retained as a control for the experiment.

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[11] D. E. Ferguson, Transuranium Quarterly Progress Report for Period Ending August 31, 1962, ORNL-3375.

All work involving uncontained plutonium was done in an alpha laboratory in Building 3508. Existing equipment in that building was used for all steps in making the pellets, and equipment being developed for the plutonium target fabrication was moved to Building 3508 for the welding and leak-checking operations. Collapse of the prototype elements was done at the Y-12 Plant.

The first major difficulty encountered was that of pressing the pellets. To prevent wrinkling of the pellets during pressing, it was necessary to press-fit each aluminum pellet liner into the die. When the first pellets made (using the reference pressure of 30 tsi) have low compacted density, it was thought that the cause was increased friction between the pellet liner and the die. In an attempt to compensate for what was believed to be increased friction, the pressing pressure was increased to 50 tsi. Even at this maximum allowable level, the proper pellet density could not be achieved. It later was found that the press gage was in error and that, in fact, 30 tsi was probably never reached ~~even though the gage registered 50 tsi.~~

The second major difficulty was with welding. The first end-cap weld to each finned tube was made by using a simple plug-type end-cap-joint design and fusion welding. The welds were leaktight, but only 50% penetration was obtained. The final end closures were made by using fusion welding and a step-type joint design, which in comparative tests with the plug joint indicated some advantages in preventing blowout and obtaining good penetration. A series of final closures on dummy tubes were successfully made by using the step-type joint, manual torch positioning, and a water-cooled chill block. Because of the interference of the hex can on the prototype elements with the water-cooled chill block, the chill block could not be used, and, therefore, the only means available for heat removal was a more inefficient copper chill ring. Only two leak-tight welds were made. The other two welds were, however, deemed sufficiently tight for proceeding with collapse of the elements. These welds were to have been repaired after collapse.

The assembled elements were individually sealed in "Unichrome" rubber bags, filled with mineral oil, and hydrostatically collapsed at 14,000 psi.

Three of the elements collapsed catastrophically at the support liners, which are specified as 7075-T6 aluminum and are supposed to prevent collapse of the tubing at the ends where the voids are located. It has not been determined whether the support sleeves were overaged during welding or whether they were not properly heat treated initially. After collapse, it was discovered that one element contained some mineral oil. In addition during collapse, a number of the tack welds, which are used to attach the hex can to the fins, were ruptured.

The experience in attempting to make these prototypes indicates that a number of refinements and changes must be made in the procedures before another attempt is made. It is notable, however, that no difficulties were encountered in making elements free of transferable contamination.

5. TRANSURANIUM FACILITY DESIGN

W. E. Unger

5.1 Hot Development Facility, Cells 3 and 4, Building 4507

F. L. Peishel W. R. Whitson
W. D. Burch L. N. Howell

The design of the equipment to be installed in cell 4 of Building 4507 is 95% complete. All major components are scheduled for fabrication in ORNL shops and three vendor shops. The present schedule, barring any major delays of equipment components, indicates a cell completion date for the end of February 1963.

The storage tanks beneath the 10-in. lead floor of cell 4 are in operation, and, one tank, T-416, now contains 70 liters of americium-curium--rare earth solution from the cross section program plutonium rods. The second batch of solution from the burnup sample TRU rods is now being processed in cell 1 of Building 4507 and will be transferred to tank T-417 by January 1963.

The presently planned usage for cell 4, consisting of Transuranium process development and various processing commitments, will require that the cell be operated continuously through 1965. With this in mind, all equipment has been designed to be physically durable or remotely maintainable if the former criteria could not be met. Spare piping will be installed to replace any unforeseen failures in this component.

Cell 3, as now planned, will be used as a supporting cell for cell 4. Various services will be provided to be used in conjunction with chemical glassware mounted on a table or prefabricated on an equipment rack for installation through the roof hatch of cell 3.

Cell 3 will be re-entered in 1964 to install a large dissolver and supporting components and possibly another cycle of solvent extraction equipment. The unreplaceable items such as roof plugs will be designed to allow the installation of these components at that time.

5.2 Design of Chemical Processing Equipment

W. D. Burch B. J. Crump O. O. Yarbro
T. S. Mackey E. J. Breeding T. L. Trent

Design work was continued towards refining the equipment and engineering flowsheets and preparing concept packages which define the work to be done in the final detailed equipment design. Start of the detailed design at K-25 has been delayed until all design work on the hot development facility in cell 4 of Building 4507 has been completed there.

5.2.1 Flowsheets

After review of the latest chemical developments with the laboratory development group, minor revisions to the equipment flowsheets were made and the complete set of five was issued as final drawings. Equipment for the separation of californium, einsteinium, and fermium and for other product purification steps will be added later when chemical development is complete.

Drafts of all 13 engineering flowsheets are complete, but detailed checking against the equipment flowsheets and line numbering must be completed before they are issued.

5.2.2 Dissolver and Control System

Targets will be dissolved in hydrochloric acid at a rate of about 1 g of aluminum per minute. This rate is set by heat removal and off-gas limitations. A partial dejacking will be carried out to remove the fins and the resulting solution will be discarded in order to reduce the aluminum content of the feed. The aluminum dissolution will then be completed by adding fresh acid. Following the aluminum dissolution, the hydrochloric acid concentration will be increased to about 6 M and the dissolver held near boiling for 1 or 2 hr to dissolve the actinides.

The control system for maintaining the dissolver at the desired temperature is shown in Fig. 5.1. A closed recirculating loop was used to provide controlled heat addition and removal without overloading the recirculating cooling water system of the building.

The dissolution rate is a function of the hydrochloric acid concentration and solution temperature as well as the composition of the aluminum in the target. The procedure for initiating the dissolution and bringing the dissolution rate to the required level is summarized below:

1. Charge the hydrochloric acid at about 70°F and at the correct concentration to give a dissolution rate of 1.5 g/min at 150°F.
2. Turn on the heater and set the dissolver controller at 150°F. Observe the dissolution rate as indicated by a heat balance around the dissolver as the system heats up, and adjust the control point, if required, to give a dissolution rate of 1.5 g/min. (At the design flow rate of 1/2 gpm, the temperature rise across the dissolver at the design dissolution rate is 4°F, sufficient to give a semi-quantitative measure of the dissolution rate.)
3. When the temperature of the dissolver approaches the control point, the heater is turned off, and the temperature controller adjusts the water flow to the cooler to maintain the dissolver temperature at the control point. The dissolving aluminum provides sufficient heat to maintain the dissolver at temperature.

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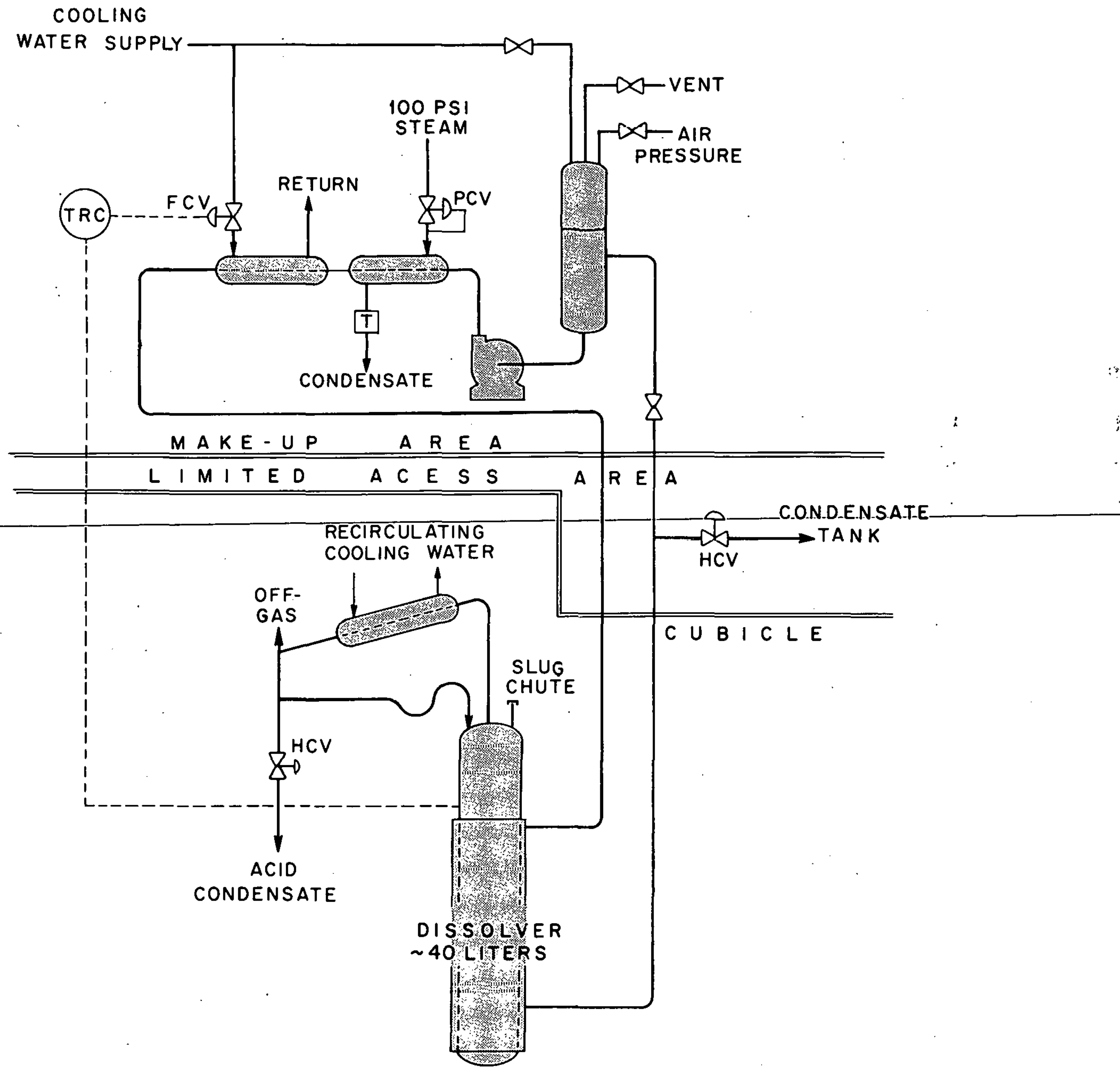

Fig. 5.1. Control System for the Dissolver.

The circulating water system has the capacity to boil the dissolver at a rate of approximately 10 liters/hr. Boiling is suppressed in the circulating water loop by pressurizing the surge pot at the pump suction to 50 psig. A condenser on the dissolver off-gas lines provides for boil-off or reflux as desired.

A code for the IBM 7090 computer was written in order to simulate the dissolution and dissolver control system. The program will be used to size the heat transfer areas, determine the stability of the system, and investigate the effects of various perturbations and simulated accidents on the control system.

5.2.3 Design of the Process Pump

In conjunction with the design of the hot development facility in Building 4507, the basic diaphragm pump mounting arrangement was redesigned for easier replacement. This mounting method, shown in Fig. 5.2, permits a replacement pump to be assembled, complete with check valves, outside the cell, transported in the conveyor to the cubicle, and then installed by making up two bolts on the equipment rack. Check valves or diaphragms may also be replaced within the cell, although the techniques and fixtures that will be required to permit this operation with the manipulators have not been worked out.

Wetted parts of the pump will be of the same material as the adjoining lines, usually Hastelloy C or Zircaloy-2.

5.2.4 Layout of the Pulse Column

The basic layout of the pulse-column solvent-extraction cycle on a 6-ft equipment rack shows that a 5-ft active length is feasible. Such a layout for the extraction column only is pictured in Fig. 5.3. Two additional columns for scrubbing and stripping will be mounted on the same rack. Temperature control will be achieved by circulating hot water through a coil encircling the column and insulated (probably with aluminum foil) with the column. The top-interface level will be measured with bubbler probes on 3-in. centers, and control will be achieved with the typical jackleg

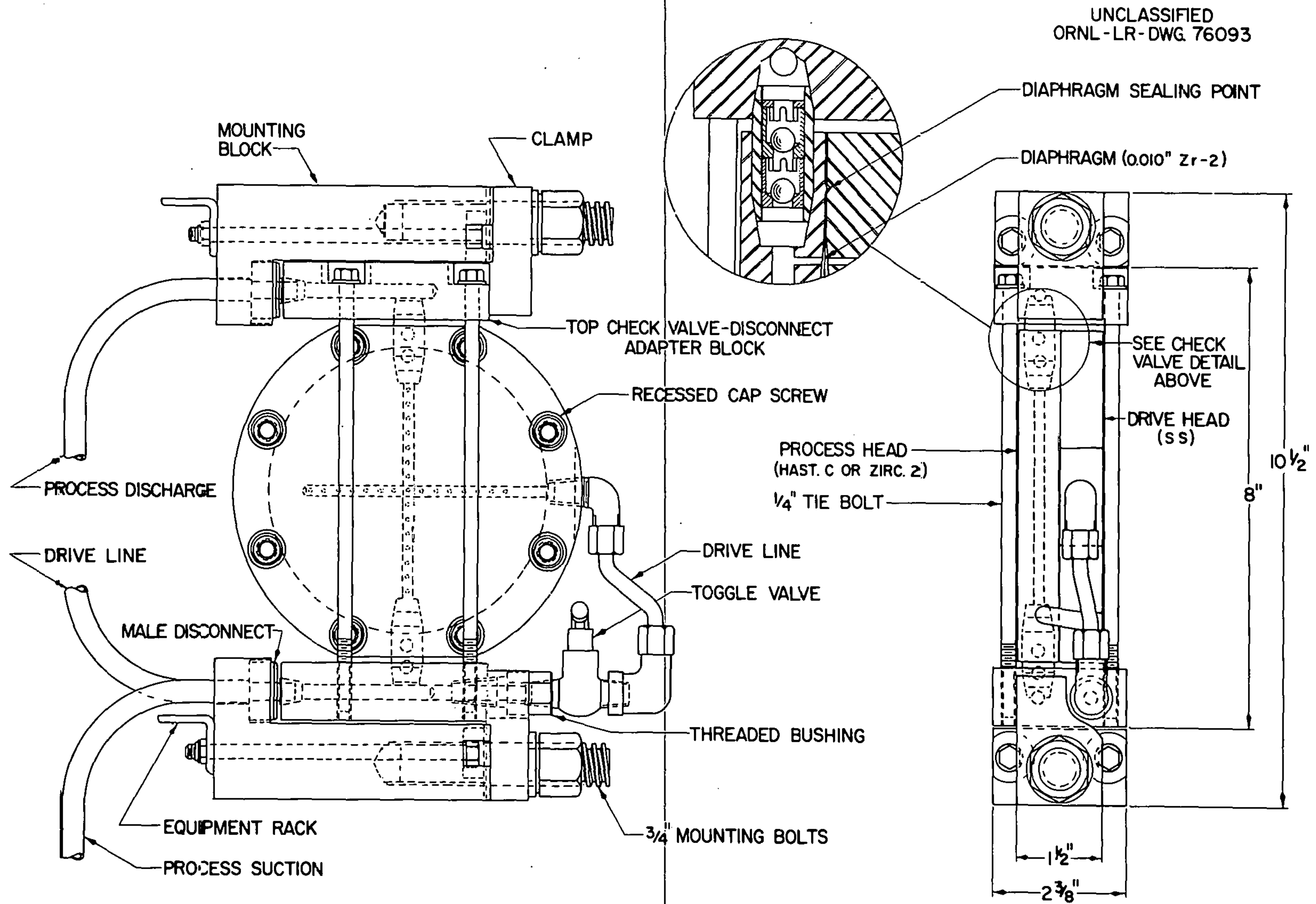

Fig. 5.2. TRU Process Pump.

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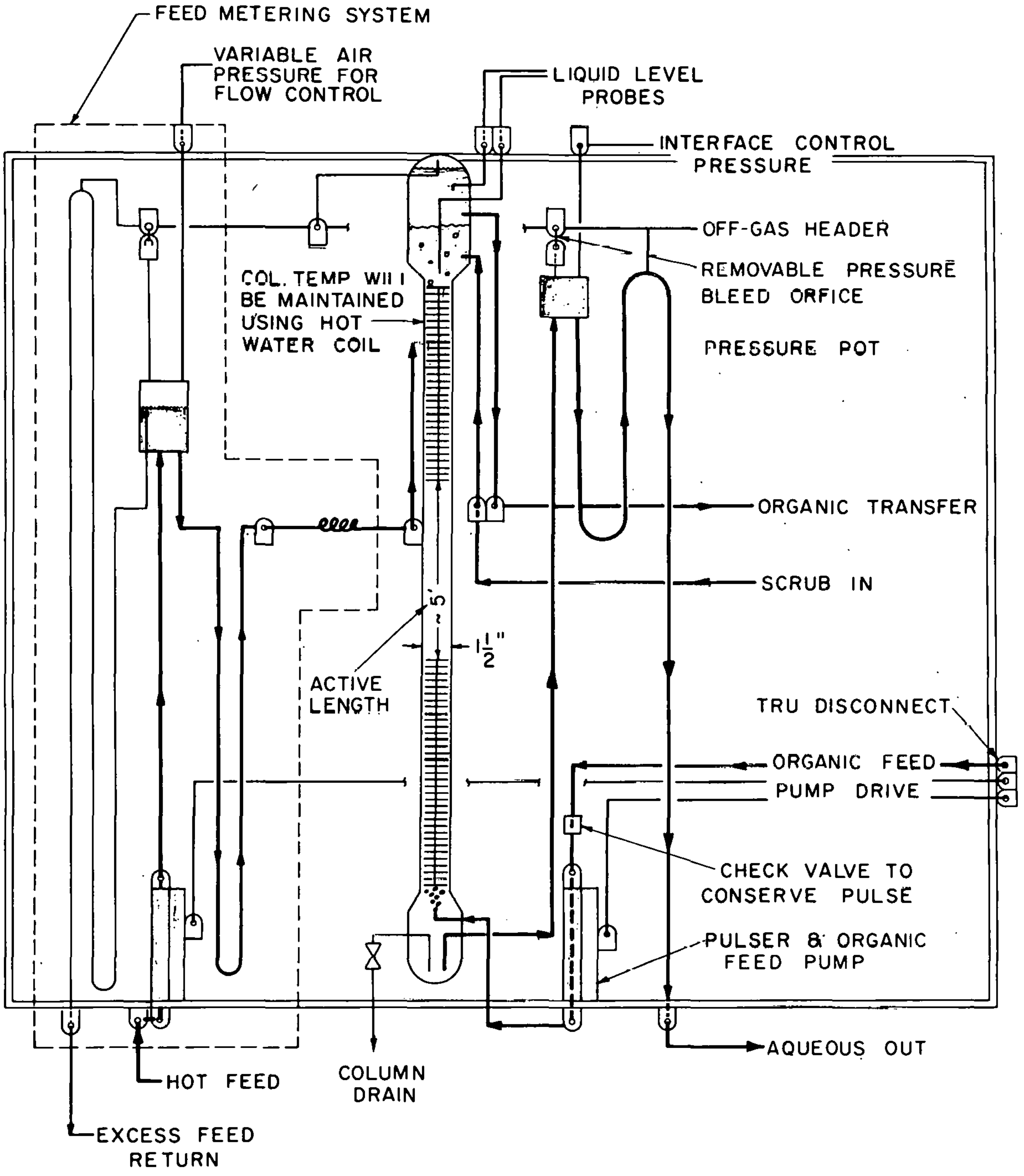

Fig. 5.3. Typical TRU Extraction Column.

pressure-pot arrangement. Pulsers will be similar to the pump pictured in Fig. 5.2, with the check valves removed and an inverted check valve added in the organic feed line which passes through the pulser. This arrangement is required to provide some pumping of organic streams between columns and as a means of ensuring that air does not get trapped in the pump. This check valve feature must be tested to ensure that proper venting of air is accomplished. Nominal feed flow capacity is in the range of 1-1/2 liters/hr.

Initial steps have been taken to obtain a set of pulse columns as described here for testing in the mockup. The primary purpose of the set is to permit hydraulic studies, using all-metal components in the geometry dictated by the TRU plant design. A second function is to test the pulsers, pumps, valves, and other components in the chemical environment to be seen in the plant. To accomplish this, the equipment rack containing the first-cycle solvent extraction system will be assembled to as near a final design as possible, using Zircaloy-2 for most of the components. It is hoped that this equipment can be used in the final facility with only minor modifications. Target date for completion is July 1, 1963.

5.2.5 Feed-Metering System

A system shown in Fig. 5.4 was designed to provide a constant, controlled, hot-feed rate to the pulse columns. Solution from the feed tank is pumped through a flow splitter that diverts a predetermined quantity to the column and the excess back to the tank. Thus the system does not depend on precise feed pump settings. Feed flow rate is varied as desired by setting air pressure to the flow splitter from an air pressure regulator--air relay system outside the cell. With equipment located in elevation as shown, the following may be accomplished:

1. Pressure drop across the capillary may be varied from 0 to 24 in. of H_2O. The capillary, consisting of about 1 ft of 1/16-in.-ID tubing, will be sized to give 1.5 liters/hr flow at 24 in. ΔP for the particular fluid being metered.

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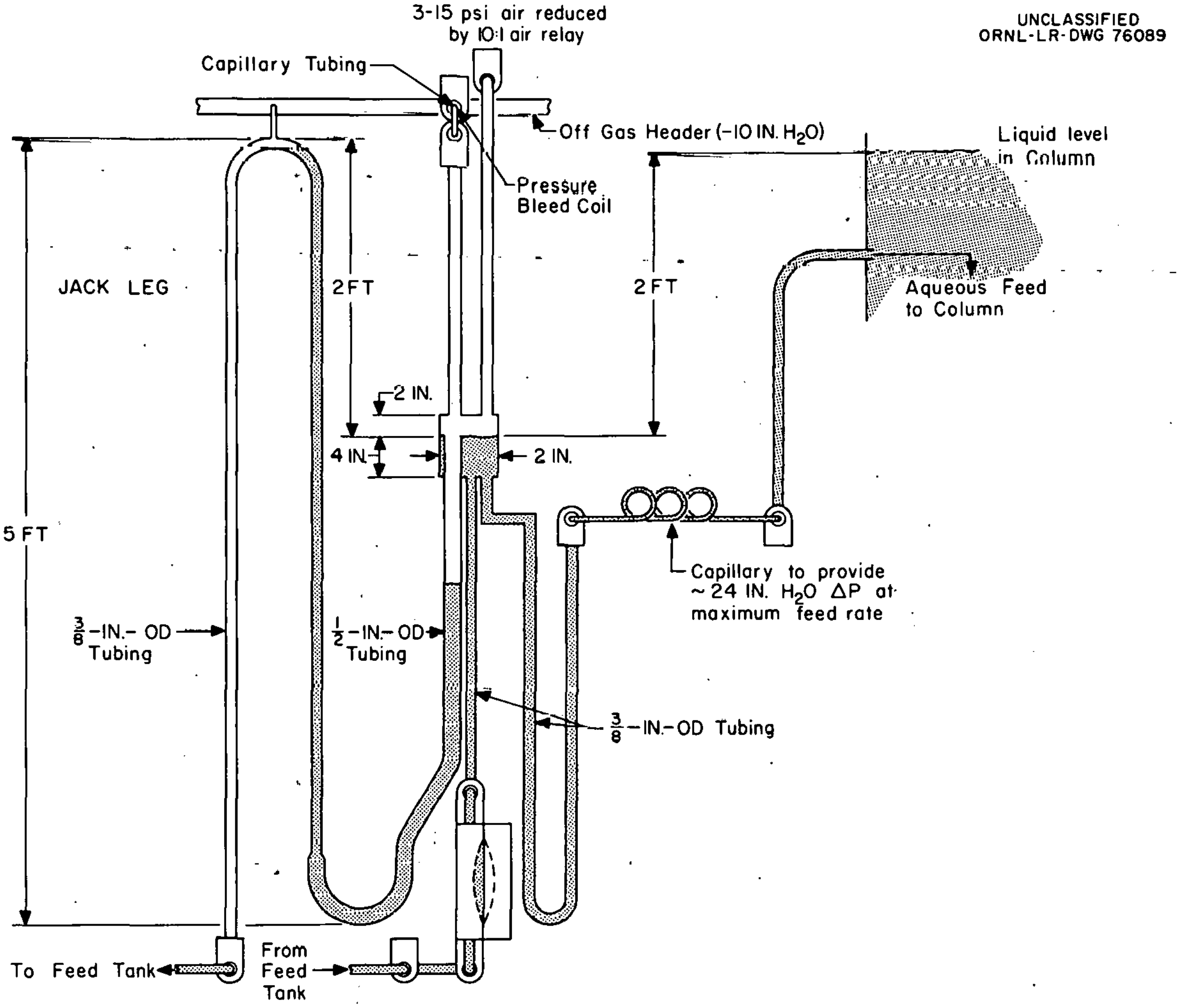

Fig. 5.4. Feed Metering System.

2. By removing air from the flow splitter, all flow is recycled to the feed tank; none enters the column.
3. By applying air pressure in excess of 24 in., all flow entering the flow splitter is forced into the column. This provision is desirable for adding cold feed from outside the cell.

A glass and plastic tube mockup of the metering system demonstrated that the concept was workable and established the tubing sizes required at critical points.

5.2.6 Detailed Equipment Design at ORGDP

Delays in completing design work on the development equipment in Building 4507 have moved the start date for detailed design work on TRU chemical processing equipment back to January 1, 1963. Several meetings have been held to familiarize ORGDP personnel with the present plans. Concept packages, including drawings and written criteria are being assembled and transmitted to ORGDP. A group of four engineers and seven design draftsmen will be available by January 1.

5.2.7 Process-Line Disconnects -- (E. J. Breeding and T. S. Mackey)

A special investment-cast disconnect (Fig. 5.5) clamp was developed for the TRU plant process lines to allow removal of process equipment by opening the clamp by manipulators. The unit may be used singly, in a single row, or two rows back to back. By unitizing the design into one clamp assembly per connection, the quantity of casting was sufficient to justify their purchase when compared with the costs of a former design wherein several connections were machined into an integral block.

Line termini were designed, fabricated, and tested for stainless steel, Hastelloy-C, tantalum, and zirconium.

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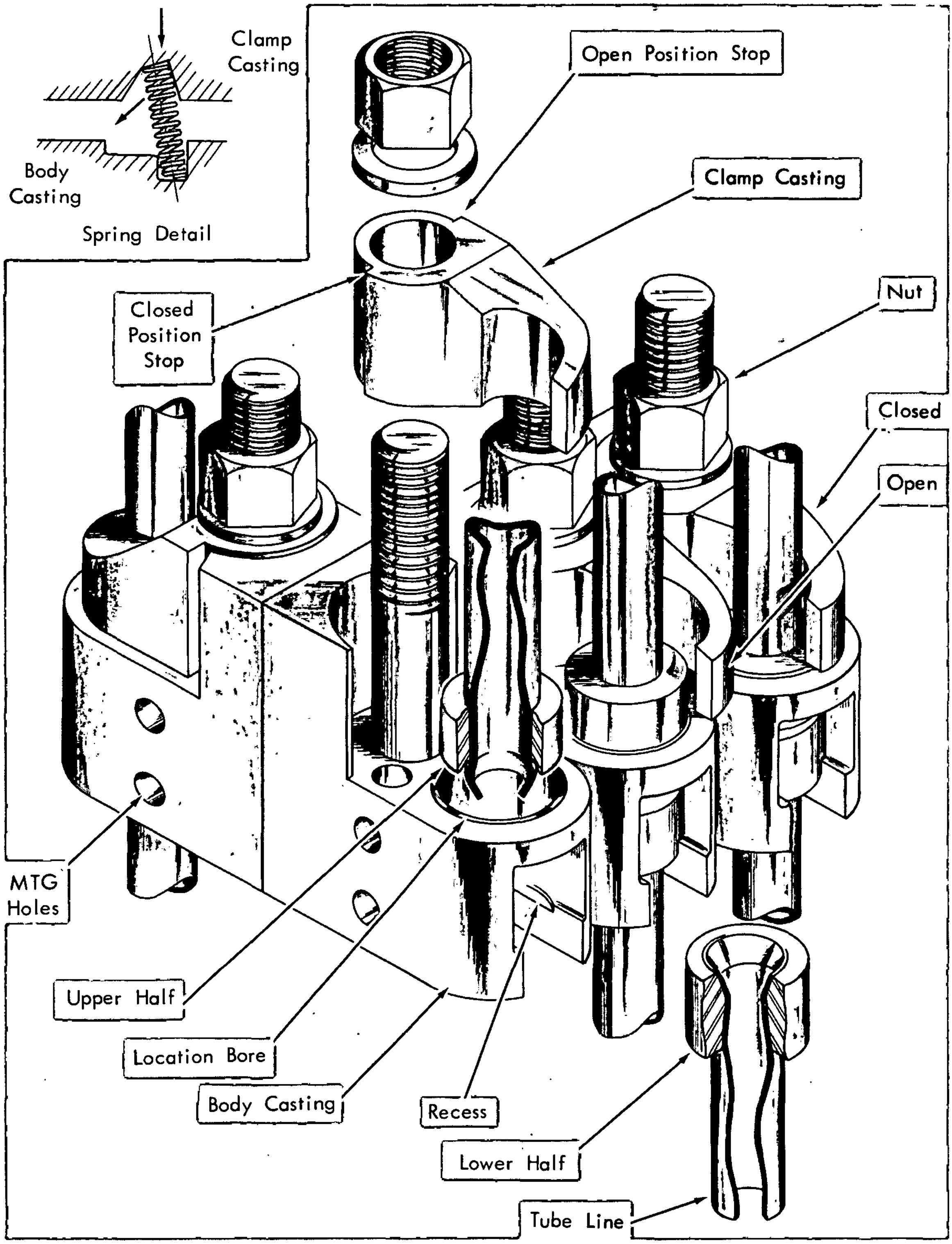

Fig. 5.5. Disconnect Assemblies, Grouped.

5.3 Design of the TRU Process Plant

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

5.3.1 Building Design

All drawings and specifications were received from the architect-engineer for initial ORNL Title II review, and about 95% were reviewed and returned. The check-out of approved drawings has been initiated at ORNL prior to final approval in Philadelphia early in January.

Procurement of the materials that ORNL will furnish the Lump-Sum Contractor has commenced. The first procurement package will consist of Hastelloy C plate, sheet, strip, and bar stock, which will be purchased by the AEC-ORO, directly from the Haynes-Stellite Company. Other Hastelloy and stainless steel procurement packages are being readied for procurement also. ORNL will procure all Hastelloy and high-quality stainless steel material, whether installed as part of the lump-sum building contract or the CPFF process equipment installation. The Hastelloy will be supplied in prefabricated and heat-treated condition, while the stainless steel will be supplied largely in basic shapes (exceptions being valves) liquid eductors, disconnect units, etc.

ORNL will also obtain the detailed design and fabrication of the ten cell-viewing windows (lead-glass and oil), complete with the alpha-seal feature and equipment for replacement of the bulk shielding windows and the alpha-seal components from the cell wall. Conceptual design and preparation of the specifications is being done in the ORGDP Engineering Division.

It is estimated that the final-bid package will be issued to the AEC-ORO by the architect-engineer in late January 1963, and that the bid period and evaluation will require 14 weeks, permitting the lump-sum contract to be let in May 1963.

5.3.2 Mechanical-Equipment Design

Intercell Conveyor. -- Nine drawings concerning installation and assembly, general arrangement and fabrication details of the intercell conveyor housing were completed, approved, and are ready for inclusion in the Title II design package.

Design of conveyor internals including the drive, chain adjuster, idlers, and the dolly is complete, and the drawings are being processed for final approval.

Design modifications are in progress on the track and chain support and control components.

Equipment Transfer System. -- Detail design of the principal components of the transfer case and cubicle door is almost complete. Drawings of the box section and inner assembly were issued for approval. Detailing of operation and maintenance tools is in progress.

Fabrication of the hoist assembly and door lock mockup for development is almost 75% complete.

Cell Cubicle. -- Detail design concerning installation and assembly, general arrangement and fabrication of floor pan, back wall, and roof assemblies of the cell cubicle is complete, approved, and ready for inclusion in Title II design package.

Specifications covering fabrication and installation of these components have been completed and issued for approval.

5.4 Critical Path Scheduling

The first manpower-leveling studies were completed in September, and the results were used to set up working schedules for the various participating groups. An analysis of these schedules revealed the information shown in Table 5.1.

The schedules are reviewed regularly to ensure that they represent current conditions.

Table 5.1. Total Manpower Required (Man-days)

	Mechanical Equipment	Target-Fabrication Equipment	Chemical-Processing Equipment	Total
Developmental design	718	389	747	1854
Final design	1074	918	3608	5600
Total design	1792	1307	4355	7454
Developmental design scheduled for FY-63	637	381	740	1758
Percent developmental design to be completed in FY-63	89%	98%	99%	95%
Final design scheduled for FY-63	606	647	1311	2564
Percent final design to be completed in FY-63	56%	70%	36%	46%

In September 1962, ORNL agreed to procure certain special items included in the architect-engineer's design package. A critical-path analysis, using the best available delivery times, was made for the procurement of these special items. The results of this study, shown in Table 5.2, were transmitted formally to the AEC by a letter dated November 12, 1962.

Two of the items in Table 5.1 are critical; they are the embedded Hastelloy C spool sections and the shielding-window liners. It may be noted that if the delivery times for these two items could be reduced 30 days, no ORNL procurement items would appear on the critical path of the overall project.

On November 14, 1962, the second Transuranium status review meeting was held, and the respective arrow diagrams were individually reviewed by representatives of Catalytic Construction Company and ORNL. At the time of this meeting, the highlights of the master schedule were as shown in Fig. 5.6.

Table 5.2 ORNL Procurement for Installation by Fixed Price Contractor[a]

	Dates Required from Critical Path Schedule Early	Late	ORNL Proposed Delivery Date[b]	Date by Present Schedule
1. Hastelloy C embedded spool sections	1/8/64	1/8/64	170	1/8/64
2. "H" quality stainless steel pipe, fittings, and disconnects for embedded spool sections	8/22/63	11/8/63	102	10/1/63
3. "H" quality stainless steel pipe, valves, fittings, and disconnects for installation in pipe tunnel	8/22/63	4/16/64	166	1/2/64
4. Hastelloy C spools for installation in pipe tunnel	2/28/64	7/13/64	207	2/28/64
5. Hastelloy material for field fabrication of waste pit and cell pit pans	10/29/63	1/27/64	122	10/29/63
6. Cubicle floor pans	5/1/64	5/1/64	230	4/1/64
7. Shielding window liners	11/20/63	11/20/63	145	11/20/63
8. Conveyor housing	2/17/64	2/17/64	195	2/12/64
9. Hastelloy C for tank pit pan screeds	9/6/63	1/8/64[c]	85	9/6/63

[a] Based on November 7, 1962, Master Schedule.

[b] Expressed as project days after contractor has access to job site (assuming present schedule of May 7, 1963, or later).

[c] The original critical-path schedule showed this item to be required by 9/6/63, but a rearrangement of the construction schedule agreed to on Nov. 14 permitted the change to 1/8/64.

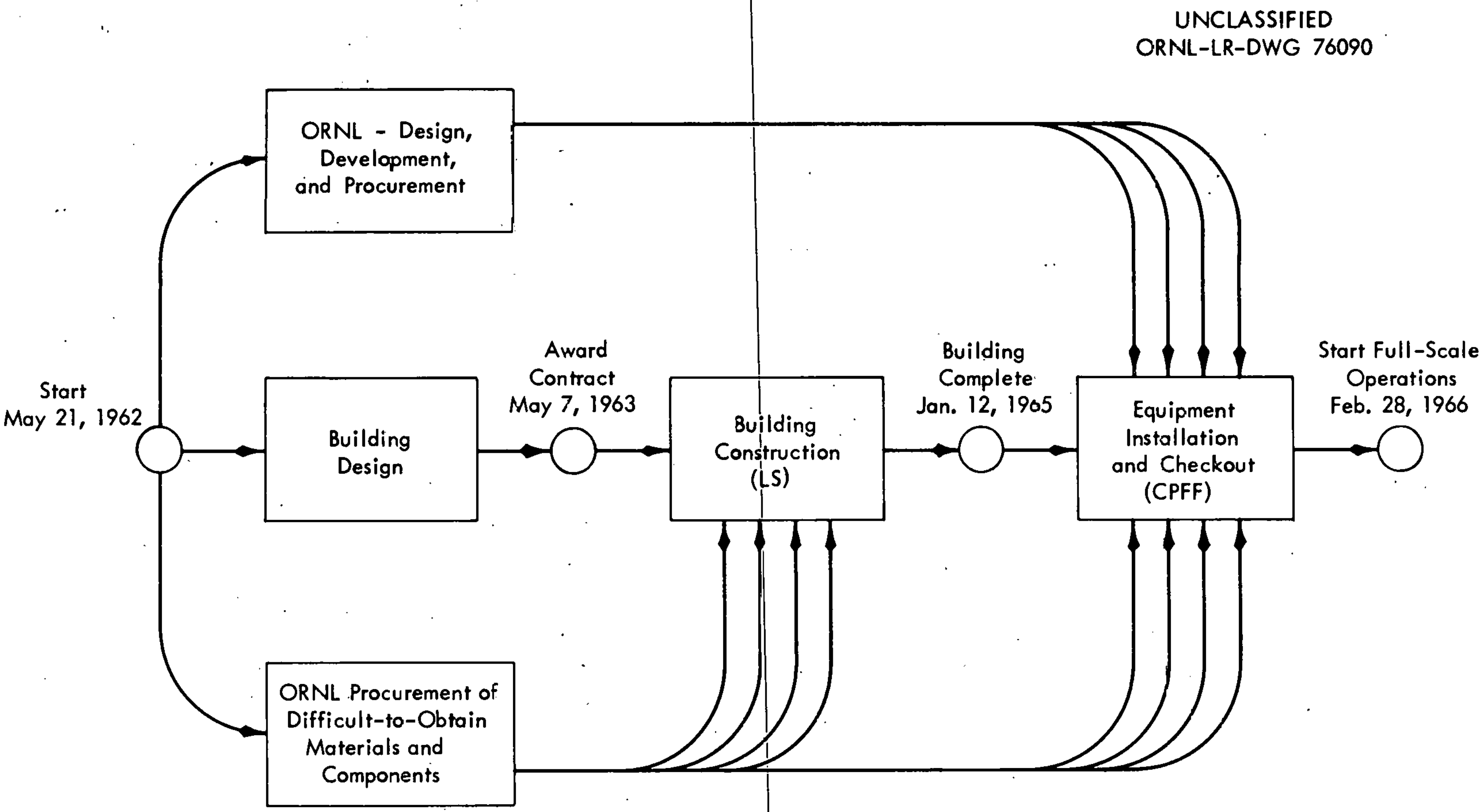

Fig. 5.6. Principal Items on the Master Schedule.

At the time of the November 14 meeting, the building-design phase had 30 days of float; however, due to design slippages and requests for more time on certain activities, about 25 days of the float will be consumed. This slippage places the building design and subsequent activities (i.e., drawing review, issue for bids, bid time, and award contract) on the critical list along with ORNL procurement of the embedded Hastelloy C spool sections and shielding-window liners.

The following improvements are planned:

1. The result of manpower leveling in the master schedule.
2. A more efficient system for reporting and disseminating information to project personnel.
3. Establishing an acceptable procedure whereby ORNL progress on the capital project is reported formally to the AEC on a monthly basis.
4. Integrating a cost program into the present schedule arrangement.

6. TRU CORROSION STUDIES

J. L. English J. C. Griess

6.1 Tantalum

The sensitivity of tantalum to hydrogen pickup in a number of TRU-related environments (solution and vapor) was investigated. Table 6.1 presents the results of the tests, most of which were run for 1000 hr. Included among the tests were tantalum-plated specimens and tantalum specimens coupled with aluminum to simulate a condition in which aluminum-clad target rods in contact with a tantalum vessel will undergo dissolution in hot hydrochloric acid solution.

With the exception of the plated specimens, corrosion rates were negligible. The latter specimens picked up considerable quantities of hydrogen due to complete dissolution of the stainless steel base metal. Dissolution resulted from contact with the hot 6 M HCl solution through defects in the tantalum plate. Both the 3- and 10-mil-thick tantalum plates were extremely brittle after completion of the tests. Some hydrogen pickup occurred also in the tantalum specimens coupled with type 1100 aluminum. There was no evidence of embrittlement, however, as determined by bending the specimens 180° after test. Neither specimen showed any signs of cracking during bending.

Table 6.1. Results of Hydrogen-Pickup Tests with Tantalum[a] in TRU-Related Environments

Environment		Time (hr)	Hydrogen Content (ppm) Solution	Vapor
Oxygenated 3 M HCl at 105°C		1000	<1	<1
Oxygenated 6 M HCl at 43°C		1000	3	-
Oxygenated 6 M HCl at 105°C		1000	<1	<1
Oxygenated 10 M LiCl--0.1 M HCl at 130°C		1000	2	<1
Oxygenated 10 M LiCl--0.1 M HCl--0.0023 M $LaCl_3$ at 130°C		1000	<1	<1
H_2O_2-Containing 10 M HCl--0.0023 M $LaCl_3$ at 35°C		1000	<1	<1
Coupled with type 1100 aluminum in 6 M HCl at 105°C	(1)	7	64	-
	(2)	6.5	54	-
Purposely damaged tantalum plate in oxygenated 6 M HCl at 105°C	(1)	100[b]	3100	-
	(2)	350[c]	2000	-

[a] Unexposed control material contained between 1 and 2 ppm of hydrogen.

[b] Three-mil-thick defected plate; stainless steel base metal dissolved completely during test.

[c] Ten-mil-thick defected plate; stainless steel base metal dissolved completely during test.

6.2 Hastelloy C

An investigation was completed to determine the effect of oxidant on the corrosion of welded and unwelded Hastelloy C by 6 M HCl solution at 35°C. Three different environments were examined: (1) no added oxidant, (2) oxygen aeration of the test solution, and (3) continuous addition of hydrogen peroxide to the test solution. Weld specimens were exposed in the solution; unwelded specimens were exposed to the vapor above the solution. Tests were of 1000-hr duration, with replacement of solutions at 100-hr intervals.

Figures 6.1 and 6.2 show weight-loss vs exposure time as semilogarithmic plots for solution and vapor specimens, respectively. In the absence of an added oxidant, the corrosion rate in solution was appreciably less than rates obtained in tests with oxygen aeration and peroxide addition. After 1000 hr, rates were 0.7, 5.2, and 4.0 mils/month, respectively, for the no-added-oxidant, oxygen-aeration, and peroxide-addition tests. All specimens underwent preferential attack in heat-affected zones adjacent to welds, with the most severe attack taking place in the oxygen-aeration test. The measured depth of penetration varied between 28 and 32 mils.

Some difference in corrosion behavior was observed on the unwelded vapor specimens. The rates were similar for the no-added-oxidant and peroxide-added tests after 1000 hr, 1.5 and 1.7 mils/month, respectively. However, the specimen in the oxygen-aerated test corroded at a rate slightly more than double these values, 3.7 mils/month.

6.3 Homalite CR-39, Homalite CR-100, and Polypenco K-51

Tests of 1000-hr duration at 80°C were completed with Homalite CR-39 (allyl diglycol carbonate), Homalite CR-100 (polyester), and Polypenco K-51 (polycarbonate) in a number of TRU-related organic processing solutions. Included among the latter were: (1) 1 M mono-2-ethylhexylphenylphosphonic acid in diethylbenzene, (2) 30% Alamine 336 chloride in diethylbenzene, and (3) 30% di-2-ethylhexylphosphoric acid in normal decane.

Good resistance was exhibited by all materials in the three solutions. Dimensional and weight changes during test were relatively minor. Some slight darkening took place on the two grades of Homalite, and the Polypenco K-51 specimens showed no change in appearance during exposure.

7. ANALYTICAL METHODS DEVELOPMENT

L. T. Corbin   J. C. White

7.1 Determination of Americium

F. L. Moore

During this reporting period, development and evaluation of an analytical method that is specific for the determination of americium

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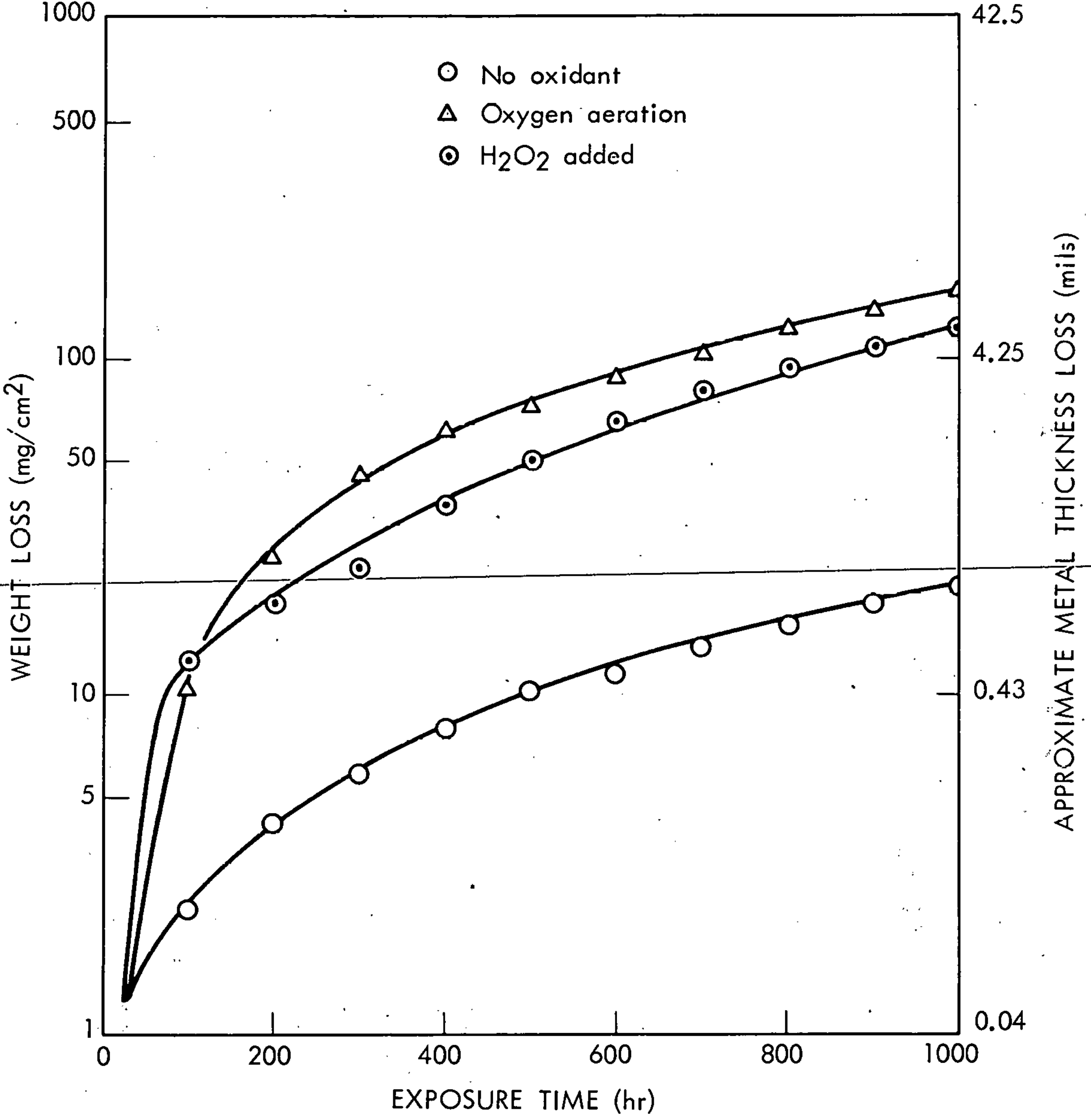

Fig. 6.1. Effect of Oxidizers on Corrosion of Solution-Exposed, Welded Hastelloy C by 6 M HCl at 35°C.

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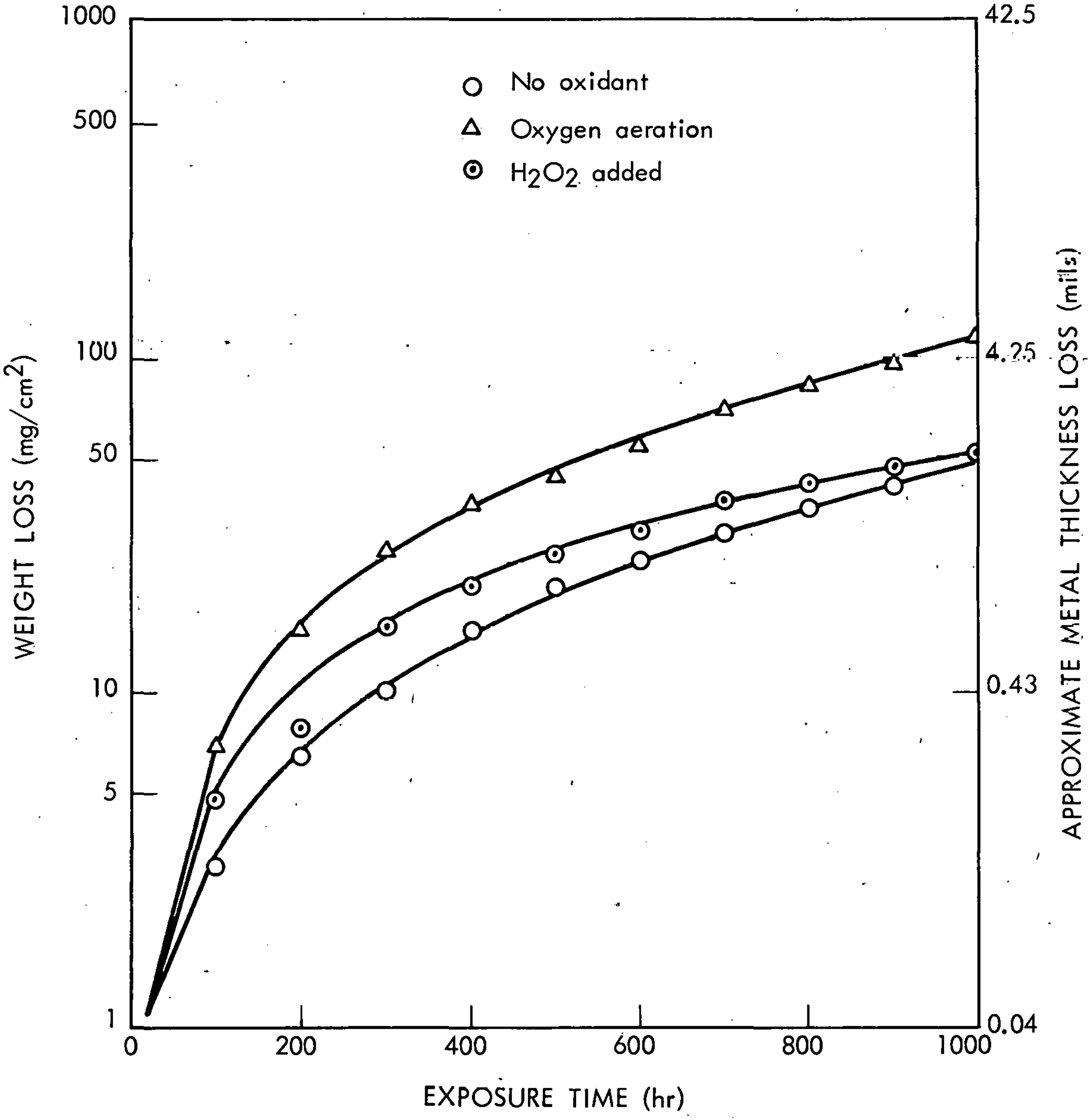

Fig. 6.2. Effect of Oxidizers on Corrosion of Vapor-Exposed, Unwelded Hastelloy C by 6 M HCl at 35°C.

was completed. This new method is based on the oxidation of trivalent americium to the sexivalent state with silver-catalyzed peroxydisulfate. Fluoride stabilizes the americyl (AmO_2^{2+}) ion during a series of scavenges with lanthanum fluoride. Excellent separation of americium tracer is effected from Cm, Bk, Cf, Pu, U, Th, Cs, Sr, Zr, Nb, Ru, Fe, Al, and the lanthanides. A recent report[12] gives details, efficiency, and application of the method.

7.2 Absorption Spectrophotometric Studies of Transuranic Elements

D. A. Costanzo

Computer methods are being investigated to make possible the analysis of spectrophotometric absorption spectra of solutions containing more than one absorbing species, any or all of which may not be at equilibrium. The program will be used to determine the concentration of all absorbing species in the system at any given time and to determine the reaction kinetics of systems that are not at equilibrium. A detailed description of the problem was written and submitted to the Mathematics Panel to be programmed for the IBM 7090 computer.

The Cary model 14PM recording spectrophotometer was serviced and repaired, and the glove box system was installed and is being equipped.

Micro absorption cells and other specially designed absorption cells are being fabricated and should be received in the near future.

7.3 Determination of Americium

H. C. Meyer J. H. Cooper

Six dissolution samples of irradiated americium were analyzed according to the method developed by Moore.[12] Using standard carrier techniques, the carrier recoveries ranged from 97.5 to 99.8%, and the

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[12] F. L. Moore, Separation of Americium from Other Elements: Application to the Purification and Radiochemical Determining of Americium, ORNL-CF 62-9-88 (Sept. 25, 1962).

variation between duplicates was ±3%. The results agreed with those obtained by existing methods to within ±5%.

Due to the lack of samples for more stringent evaluation, the method, as presently established, was accepted as a standard radiochemical method for the TRU program.

7.4 Determination of Fission Yields of Americium

E. I. Wyatt R. R. Richard

Routine radiochemical methods were used on the six samples of irradiated americium for the determination of fission yields for mass numbers 89, 91, 140, and 144. The results disagreed slightly with published data, with mass 89 and 91 being higher by nearly 20%, and 140 and 141 lower by almost 10%. Further work will be done along this line when more samples are available.

7.5 Preliminary Calculations of Fission Product Yields of Americium

E. I. Wyatt R. R. Richard

To facilitate the development of radiochemical methods for the analysis of typical TRU fuel rods, a series of fission product yields were calculated. Sample fuel rods were: 6 g of Am^{241} irradiated for 49 days at a flux of 3×10^{14}, cooled 10 days; and a Pu^{242} target irradiated for 6 months, 1 year, 1.3 years, and 1.8 years and cooled 30 and 60 days. Preliminary calculations of the yields are shown in Table 7.1.

Table 7.1. Results of Preliminary Calculations of Fission Product Yields

Fission Product	Am^{241}		Pu^{242}	
	(μg/ml)	(mc/ml)	(μg/ml)	(mc/ml)
Sr	0.32	4.55×10^{-2}	0.18 - 0.46	2.72×10^{-2}-6.51×10^{-2}
Y	0.26	6.48	5.94×10^{-3}-0.10	0.17 - 2.57
Zr	3.37	11.86	3.51 - 9.48	0.80 - 10.87
Nb^{95}	0.18	6.93	4.26×10^{-2}-0.33	1.67 - 11.69
Mo^{99}	--	3.69	-	-
Ru	8.56	39.74	34.47 - 75.69	38.33 - 103.64
Cs^{137}	2.06	0.16	13.39 - 28.70	1.06 - 2.27
Ba^{140}	0.31	22.64	6×10^{-5}-0.34	4.7×10^{-4}-24.71
Ce	5.06	27.01	8.33 - 49.01	15.06 - 81.37
TRE's	14.27	90.16	51.27 - 118.6	29.8 - 167.6

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