OAK RIDGE NATIONAL LABORATORY LIBRARIES
3 4456 0549421 1

CENTRAL RESEARCH LIBRARY
DOCUMENT COLLECTION

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

UNION CARBIDE

CENTRAL RESEARCH LIBRARY
DOCUMENT COLLECTION
LIBRARY LOAN COPY
DO NOT TRANSFER TO ANOTHER PERSON
If you wish someone else to see this document, send in name with document and the library will arrange a loan.

ORNL-TM-508

COPY NO. -16

DATE - 3/1/63

CHEMICAL TECHNOLOGY DIVISION MONTHLY REPORT FOR FEBRUARY 1963

F. L. Culler, Jr.

ABSTRACT

Process development studies are reported on Power Reactor Fuel Processing (Shear-Leach Process Development, Processing of Breeder Reactor Fuels, Hydrochlorination of Uranium-Aluminum Alloy Fuel, Chlorination of Uranium Carbide--Thorium Carbide--Graphite Fuels, Solvent Extraction Flowsheet Development for UC Fuel, and Protactinium Adsorption Studies); Thorium Utilization Program (Fuel Cycle Development: Fuel Preparation); Waste Disposal Development (High-Activity-Waste Calcination, Low Activity Wastes, and Foam Separation); Chemistry Research (Protactinium Chemistry); and Transuranium Element Production (Analytical Chemistry Development, Corrosion Studies, Design of Experimental Facilities, HFIR Target Development, Chemical Process Development, Process Equipment Development, and Transuranium Facility); and Isotope Development Program (Cm^{242} Preparation).

NOTICE

This document contains information of a preliminary nature and was prepared primarily for internal use at the Oak Ridge National Laboratory. It is subject to revision or correction and therefore does not represent a final report. The information is not to be abstracted, reprinted or otherwise given public dissemination without the approval of the ORNL patent branch, Legal and Information Control Department.

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

CONTENTS

Page

Power Reactor Fuel Processing 5

Shear-Leach Process Development 5

Processing of Breeder Reactor Fuels 5

Hydrochlorination of Uranium-Aluminum Alloy Fuel 5

Chlorination of Uranium Carbide--Thorium Carbide--Graphite Fuels 6

Solvent Extraction Flowsheet Development for UC Fuel 6

Protactinium Adsorption Studies 8

Thorium Utilization Program 10

Fuel Cycle Development: Fuel Preparation 10

Waste Disposal Development 12

High-Activity-Waste Calcination 12

Low-Activity Wastes 12

Foam Separation 13

Chemistry Research 15

Protactinium Chemistry 15

Transuranium Element Production 16

Analytical Chemistry Development 16

Corrosion Studies 16

Design of Experimental Facilities 17

HFIR Target Development 18

Chemical Process Development 19

Process Equipment Development 20

Transuranium Facility 21

OAK RIDGE NATIONAL LABORATORY LIBRARIES

3 4456 0549421 1

CONTENTS

Page

Isotope Development Program 23

Cm^{242} Preparation 23

POWER REACTOR FUEL PROCESSING

Shear-Leach Process Development. -- Bench-scale, batch leaching tests are being made with sheared pieces of stainless-steel-jacketed UO_2-ThO_2 fuel to develop a method that ensures good contact between the leachant (nitric acid) and the UO_2-ThO_2 fines produced during shearing. In an initial test, fines were suspended successfully by discharging air through a draft tube; the sheared pieces were leached at a satisfactory rate. Leaching of the UO_2-ThO_2 was 99.95% complete in 14 hr of contact with boiling (117°C) recirculating 12 M HNO_3--0.04 M F--0.10 M Al. The gross recirculation rate in the 18-liter system was about 40 liters/min.

Processing of Breeder Reactor Fuels. -- The experimentally determined instantaneous dissolution rate of high-density (85 to 93% of theoretical) UO_2-PuO_2 pellets in nitric acid or in mixtures of nitric and hydrofluoric acid agreed closely with values calculated from the rates for UO_2 pellets and PuO_2 pellets of the same theoretical density. The addition of oxidizing agents to nitric acid did not increase the dissolution rate of PuO_2 pellets significantly.

Multiple extraction and Martin-technique experiments indicate that not more than 1% of Pa^{233} tracer is unextractable into 30% tributyl phosphate in dodecane from 0.5 M $Al(NO_3)_3$ containing 2 or 5 N HNO_3 and that the distribution coefficients are about 2 and 6, respectively.

Hydrochlorination of Uranium-Aluminum Alloy Fuel. -- Calculations with the GHTR code (using the IBM-7090 computer) to determine maximum temperatures obtainable in the hydrochlorination of U-Al alloy MTR fuel were continued. In these calculations it was assumed that all heat is lost by radiant heat transfer. The maximum steady-state temperature

decreased from 1073°C at a hydrochlorination rate of 12 mils/hr to 549°C at 6 mils/hr. At an average rate of 6 mils/hr, the 60-ml inside plates and the 180-mil side plates could be completely hydrochlorinated in 10 and 30 hr, respectively. The additional small contribution to heat removal by the heat capacity and convection of the gases was not considered. These results indicate that it will be possible to hydrochlorinate MTR fuel elements at rather low rates without exceeding the melting temperature of the fuel, about 650°C.

Chlorination of Uranium Carbide--Thorium Carbide--Graphite Fuels. -- Three 10-g samples of UC-ThC_2-graphite fuel containing 1.5% U and 7% Th were burned in oxygen at 800°C in 2.5 and 3.5 hr. The ash so produced was then chlorinated in 2 hr with 15% CCl_4--Cl_2 at 500 and 550°C for two of the 10-g samples and with phosgene at 500°C for the third. Complete chlorination was achieved in all three cases, and the nonvolatile products were completely soluble in water. The respective amounts of uranium volatilized from the combustion residues during chlorination represented 74, 84, and 64% of that in the residues. This volatilized uranium chloride was collected on the cool exit of the reaction tube.

These experiments indicate that it is possible to process this type of fuel in a nickel vessel, yielding an aqueous product solution containing both the uranium and the thorium. The complete separation of uranium and thorium might be made possible by using higher temperatures in order to completely volatilize the uranium as uranium chloride, but ceramic vessels would have to be used because metals corrode too rapidly.

Solvent Extraction Flowsheet Development for UC Fuel. -- Two alternative aqueous head-end procedures are being developed for sheared uranium

carbide fuel at ORNL: (1) hydrolysis with water at 80 to 100°C to form hydrated UO_2, followed by dissolution in nitric acid, or (2) direct dissolution of the UC in nitric acid. Further investigation of both procedures is required to determine whether they will yield a feed solution suitable for processing by conventional solvent extraction methods.

Hydrolysis of 20- to 60-g specimens of impure UC (about 5.1% C), containing some dicarbide, at 90°C for 2 hr followed by a 2-hr dissolution-reflux in 4 M HNO_3 yielded a raw feed solution containing 120 g uranium per liter in 3 M HNO_3, and waxy, yellow-brown solids. Nearly all the carbon from the UC was hydrolyzed to volatile paraffinic hydrocarbons. The water-washed, air-dried, amorphous organic solids (about 0.3 wt % of the UC) retained 0.003% U. These solids dissolved readily in acetone, though not in dodecane. Infrared spectroscopy indicated a very complex mixture of organic acids. Further analysis suggested the empirical formula: $(C_2H_2O_2 \cdot NO_3)_x$.

During exhaustive extraction of filtered and unfiltered raw feed solutions with 15% tributyl phosphate in Adakane, followed by exhaustive stripping, stable emulsions and interfacial matter were generated and persisted throughout these operations. Uranium recovery, however, was normal: 99.98%.

Pretreatment methods are being sought in which the organic acids may be chemically degraded to some form that does not interfere with solvent extraction. Scouting tests were conducted with unfiltered raw feed solutions treated with 0.1 M $KMnO_4$, $Ce(SO_4)_2$, $Na_2Cr_2O_7$, $NaNO_2$, and $(NH_2OH)Cl$, followed by 1 hr of reflux and then filtration. The filtrates were subjected to exhaustive extraction and stripping. Following the

permanganate treatment, no emulsification or interfacial matter were observed during extraction or stripping. Coalescence time was typical for a pure $UO_2(NO_3)_2$-HNO_3-tributylphosphate system, being 20 sec or less. Uranium extraction and stripping losses were 0.22 and 0.01%, respectively. Treatment with the other reagents appeared ineffective, compared with the permanganate treatment, suggesting that only partial degradation of the organic acids was achieved.

Protactinium Adsorption Studies. -- Studies on the adsorption of protactinium on unfired Vycor and silica gel were continued. A column of 80- to 100-mesh unfired Vycor, 1 cm in diameter and 8.6 cm long, adsorbed about 7.8 mg of Pa^{231} per gram of glass, or about 85% of the protactinium from 870 ml of a solution that was 0.5 M in thorium, 11.1 N in HNO_3, 0.074 M in aluminum, and about 0.03 M in fluoride. The Pa^{231} concentration was 54 mg/liter. The solution was passed through the column at a flow rate of 0.22 ml cm^{-2} min^{-1}.

During the run, the protactinium concentration in the emerging raffinate rose from 4 to 46% of the initial feed concentration. The 4% loss was constant for the first 400 ml of raffinate and was equivalent to a loading value of about 2 mg of Pa^{231} per gram of glass. The protactinium in this raffinate was not adsorbed on passing through a fresh column of unfired Vycor.

The loaded column was washed with 80 ml of 11 M HNO_3, which removed 13% of the adsorbed protactinium, and then the column was stripped with 0.5 M oxalic acid. The first 5 ml of oxalic acid removed 95% of the remaining protactinium, yielding a solution that contained 6.4 mg of Pa^{231} per milliliter, or about 120 times the concentration of the Pa^{231} in the

feed solution. More than 99% of the protactinium had been removed after a total of 30 ml of strip solution had passed through the column.

THORIUM UTILIZATION PROGRAM

Fuel Cycle Development: Fuel Preparation. -- The sol-gel process was modified to prepare rounded particles of thorium dicarbide and thorium-uranium dicarbide. Thorium oxide sols, and up to 10 atom % uranium--thorium oxide sols, 2 M in metal, were prepared. High-surface-area carbon (from 4 to 6 gram-atoms of carbon per gram atom of uranium plus thorium) was mixed into the sol. The sol was evaporated to a gel and was fired at 1580°C for uranium-thorium dicarbide and up to 1775°C for thorium dicarbide. Better than 99.8% conversions to dicarbides were obtained, with free-carbon residues of 4 to 7 wt % and unconverted oxide residues equivalent to 215 to 260 ppm of oxygen.

Rounded particles were prepared by dispersing the sol in carbon tetrachloride-isopropyl alchol mixtures, drying gently, then firing. Particles 50 to 250 μ in diameter were prepared, and they kept their shape during firing.

Uranyl nitrate was greater than 99% denitrated by steam at 180 to 250°C in 4 to 6 hr in a rotary calciner to produce alpha-UO_3 having crystallite sizes of 400 to 600 A and surface areas of 0.5 to 3 m^2/g. The nitrate was recovered entirely as nitric acid. The higher-surface-area UO_3 was reduced in the rotary calciner (in the presence of a 2:1 mole ratio of formic acid and hydrogen and palladium-impregnated thoria) at 100 to 400°C to UO_2 that had an average crystallite size of 127 A.

Two 22-in.-long irradiation capsules containing 5 wt % enriched uranium in sol-gel-produced ThO_2-UO_2 were put into the MTR. The capsules are scheduled to be moved to higher fluxes three times to accumulate an average burnup of 105,000 Mwd/tonne. About 4.5 kg of 5.0 wt % enriched

uranium in sol-gel-process ThO_2-UO_2 was prepared for irradiation specimens in the MTR. An operating procedure was written for the rotary denitrator, and successful routine operation was started.

WASTE DISPOSAL DEVELOPMENT

High-Activity-Waste Calcination. -- Hard, microcrystalline solids incorporating up to 45% Purex-waste oxides were prepared from simulated high sulfate and 1965 FTW (formaldehyde-treated waste) solution by the addition of phosphite or phosphate and sodium, calcium and/or magnesium. On a semiengineering scale, simulated Darex waste was fixed into a very hard, strong product that incorporated 25% waste oxides and represented a volume reduction of 5.17 from the waste solution. The feasibility of removing mercury from 1965 FTW by amalgamation with copper was demonstrated on a laboratory scale.

Type 304L stainless steel coupled to titanium 45A showed weight losses amounting to a maximum corrosion rate of 0.41 mil/month for 192 hr of exposure to the vapor above refluxing Hanford FTW (Purex waste) solution. Rates for specimens exposed to the solution and at the interface were slightly lower. Titanium 45A showed slight weight gains for coupled and uncoupled specimens under these conditions.

Low-Activity Wastes. -- The scavenging-precipitation ion-exchange process employs a sodium hydroxide (0.01 M) precipitation followed by clarification and filtration as the head-end treatment. Phosphates in the waste stream hinder complete precipitation, producing a stabilized supersaturation of hardness ions. A carboxylic resin, Amberlite IRC-50, effectively removed this residual hardness prior to the secondary decontamination with the phenolic ion exchange resin, Duolite CS-100. Under the worst conditions, the hardness (70 ppm) was removed from about 1200 bed volumes of treated waste, postponing the breakthrough of the secondary column to the 1600-to-1800 bed volumes normally achieved when

there is less than 10 ppm of hardness in the treated waste containing 0.01 M Na^+. This treatment is superior to the one that removes hardness by means of the addition of sodium carbonate because that one reduces the loading of cesium on the CS-100 resin due to the competition between the added sodium and the cesium. A residual hardness of 40 ppm caused a 0.1% cesium breakthrough of the CS-100 resin at about 570 bed volumes in the presence of the usual 0.01 M Na^+ concentration. The addition of 0.005 M Na_2CO_3(0.01 M Na^+) reduced the residual hardness to less than 10 ppm but doubled the sodium concentration to 0.02 M. This increase in sodium concentration resulted in a 0.1% cesium breakthrough at about 900 bed volumes on the secondary (CS-100 resin) column. The Amberlite IRC-50 resin, like Duolite CS-100, can be regenerated with 0.5 M HNO_3.

Foam Separation. -- The removal of strontium from water by foam separation is much more efficient if the calcium concentration is in the 5-ppm range (calculated as calcium carbonate), rather than in the 65-ppm range, as it is in ORNL tap water or low-radioactivity-level waste. Accordingly, studies of a head-end calcium precipitation are continuing, and a 9-in.-diam sludge column was used with ORNL process-waste water as feed. The unit was operated continuously for two 20-day periods at flow rates from 10 to 60 gal ft^{-2} hr^{-1}. The precipitation of calcium and magnesium was induced by making the water 0.005 M each in NaOH and Na_2CO_3; a 2-ppm concentration of ferric iron was used as coagulant. The ratio of sludge-bed height to column diameter was in the range 0.5 to 1. Excellent operation was obtained at all flow rates. Dissolved hardness of the effluent water was 2.0 to 4.5 ppm, while turbidities ranged from 0 to 5 ppm. Radiochemical analyses showed

Sr^{90}, Co^{60}, Cs^{137}, Ru^{106}, and Ce^{144} decontamination factors of 10 to 15, 30, 1.1, 2.0, and 40, respectively. Chemical analyses showed the alkylbenzene sulfonate and phosphate concentrations in the waste water to be 0.75 and 1-2 ppm, respectively.

Subsequently, the effluent from this head-end-treatment process was tested for strontium removal by the foam separation process; a 6-in.-diam column was used. The flow rate was 40 gal ft^{-2} hr^{-1}; the gas-to-liquid flow-rate ratio was 10; the dodecylbenzene sulfonate (DBS) surfactant concentration was 275 ppm. A decontamination factor of 27 was obtained for strontium under these unoptimized conditions. The strontium distribution factors of (0.6 to 2.4) x 10^{-3} cm, calculated from column runs with clarifier effluent, agree with values obtained from laboratory studies on solutions that contained about 5 ppm of calcium.

Engineering studies were continued in 6-in.-diam foam columns with DBS as the surfactant. Screen-lined, perforated centrifuge bowls were tried as foam breakers, with the following results: The efficiency of foam condensation is greater for a low flow rate of dry foam than for high rate of wet foam. Screens of 100 or 120 mesh break foam more efficiently than do screens of 40 or 200 mesh. The capacity of the centrifuge increases as the rotational speed increases. However, the foam-condensing efficiency increases as rotational speed decreases, provided that the capacity of the rotating screen is not exceeded.

CHEMISTRY RESEARCH

Protactinium Chemistry. -- Careful measurements of protactinium distribution between 5 N sulfuric acid and Dowex 1 resin showed that at Pa^{231} concentrations greater than 2×10^5 counts min^{-1} ml^{-1} (0.004 mg/ml) in the acid, the concentration of protactinium in the resin increases with approximately the fifth (± 1) power of the protactinium concentration in the acid. In these tests, protactinium concentrations varied from 3×10^6 counts/min per gram of resin (0.06 mg/g) to 1.3×10^8 counts/min per gram (2.6 mg/g). At protactinium concentrations below 2×10^5 counts min^{-1} ml^{-1} in the aqueous phase and 3×10^6 counts/min per gram in the resin, the concentration in the two phases were approximately linearly related, although scatter in the data prohibited a definite conclusion.

The extraction of protactinium from sulfuric acid into 0.03 N trilauryl amine--diethylbenzene solution showed a similar large increase in protactinium concentration in the solvent as the concentration of the protactinium in the aqueous phase was increased. Preliminary results indicate this to be a third- to fourth-power dependence. In both systems at least several days were required to reach equilibrium.

TRANSURANIUM ELEMENT PRODUCTION

Analytical Chemistry Development. -- A computer program was written for the analysis of absorption spectrophotometric data for systems containing a number of absorbing species that may or may not be at equilibrium. The program was tested by using synthesized spectrophotometric absorption data for two, parallel first-order reactions involving three components. The results were excellent.

The investigation of various methods for the determination of zirconium in various Tramex solutions was started. At least four methods seem to be applicable to the microgram quantities of zirconium that are present and also to hot-cell manipulation. These will be evaluated in terms of the effect of high radiation (both alpha and gamma), interfering ions, and sensitivity.

Both the 400-channel analyzer and the automatic alpha counter are being inspected and assembled by the Instrument and Control Division and should be operational by February 15.

Corrosion Studies. -- An investigation of the effect of oxidant species on the corrosion of Hastelloy C by 6 M HCl at 35°C was completed. Tests were run with (1) normal atmospheric oxygen in solution, (2) with oxygen aeration of the solution, and (3) with continuous addition (about 3 ml/hr) of 0.05 M H_2O_2 solution. The peroxide concentration in the test solution never exceeded 0.001 M. The best corrosion resistance was shown by a solution-exposed weld specimen with atmospheric oxygen present; after 1000 hr, the rate was 0.7 mil/month. With oxygen aeration and with peroxide addition, the rates for similar specimens increased to 5.2 and

4.0 mils/month, respectively, after 1000 hr. All specimens were preferentially attacked in heat-affected zones adjacent to welds. The severity of the attack was greatest in the oxygen-aeration test, where the measured penetration ranged between 28 to 32 mils. Rates after 1000 hr were nearly identical for unwelded specimens exposed to the vapor in the no-added-oxidant and in the peroxide-added tests, being 1.5 and 1.7 mils/month, respectively. The unwelded specimen in the vapor above the oxygen-aerated test corroded at a rate of 3.7 mils/month.

Corrosion data were obtained on welded Zircaloy-2 in mixtures of nitric and hydrochloric acids at 43°C and at boiling temperatures. When the solution was 6 M in nitric acid, with varying concentrations of hydrochloric acid up to 0.5 M, the corrosion rate was consistantly less than 0.1 mil/month in the liquid phase. A maximum vapor-phase corrosion rate of 0.2 mil/month was observed above a boiling solution of 6 M HNO_3--0.5 M HCl. There was no indication of preferential attack in the weld areas. However, accelerated corrosion was observed in both 3 M and 6 M HCl that were also 0.1 and 0.5 M in nitric acid. In 3 M HCl, the 0.5 M HNO_3 produced a corrosion rate of 5.5 mils/month at boiling and 0.7 at 43°C. In the 6 M HCl solution, 0.1 M HNO_3 produced the highest corrosion rates: 1 mil/month at 43°C and 10.8 at boiling. Again, no preferential attack in heat-affected zones was noted.

Design of Experimental Facilities. -- The design of cell 4 is 98% complete, and the design of the roof-area sample-handling equipment is in the checking stage. The design of prefabricated piping is 75% complete. Design of the sampling system and the shielding for the multichannel gamma spectrometer have not been started.

The construction of the makeup area is complete except for the installation of the glove box for the pump drive. The construction of the operation area is complete except for the installation of the fire protection system and the gamma spectrometer equipment. The equipment for the roof area has not been installed, but all components except the sample-removal box are fabricated. All structural supports are in the cell, and installation of the piping was started.

The cell tanks to be plated are now at Parma, Ohio, being prepared for application of the tantalum. A tantalum valve is being made in the ORNL shops, and, when completed, it will be tested for operability and leaktightness at the TRU mockup.

HFIR Target Development. -- Pellet-pressing studies showed that the addition of about 20 vol % of ultrafine aluminum powder to 63 vol % of the 20-μ metal particles produces a continuous-metal matrix when pressed with 17 vol % of oxide. The ultrafine metal powder and the ceramic powders were blended first and then mixed with the coarser metal power, finally being cold pressed. A very uniform dispersion of the oxide in the metal matrix was obtained.

The use of a continuous-acting press with rapid load rates produced a superior surface on the aluminum cylinders. The cap powder was also better densified and more resistant to erosion during ultrasonic cleaning. The use of the fast-acting press may permit less stringent dimensional requirements in the fit-up between the die and sleeve.

End closures on simulated target capsules were made by using a braze alloy consisting of Al--12% Si. Several advantages can be cited for braze joints, compared with welded joints. Among the advantages are the

following: The bond area is ten times larger when the joint is brazed; it is a simple operation that easily produces a good bond; and, the joint can be inspected. Several different types of braze-joint designs are being evaluated.

Construction is nearing completion on the area for the TRU mock-up assemblies. The plutonium-target line will be the first equipment assembled in this laboratory. Fabrication of the pellet inspection and loading equipment is about 80% complete in the shops at the Oak Ridge Gaseous Diffusion Plant. Designs were issued for comments and checked for correctness on the cubicle-3 transfer arm and the Blendor dispenser. Comments were favorable, and the drawings were returned for correction.

Chemical Process Development. -- Laboratory tests indicate that the Tramex feed adjustment step can be satisfactorily accomplished by evaporation. Simulated dissolver solutions containing 2 M HCl were evaporated, without aluminum precipitation, to yield a 10 M LiCl--0.2 M $AlCl_3$ solution containing about 0.1 M HCl, which had a boiling point of 129°C. Unfortunately, the depletion of acid from such solutions by radiolysis due to alpha radiation is high. With Cm^{242} at about 2 w/liter, a G value for hydrogen ion depletion of 5.5 was observed for a synthetic feed solution; it was 4.1 for an actual feed. The latter value is equivalent to the destruction of 0.5 mole of HCl per liter per day at a process level of 10 w/liter.

By pressurizing the column to suppress gas-bubble formation, Am^{241} and Cm^{242} were separated by elution from Dowex 1-8X with 4 M $LiNO_3$ at pH 1.5. The single-stage separation factor was 1.7. Use of this procedure with irradiated Cf^{252}, however, gave almost no separation of californium,

einsteinium, and fermium. Single-stage separation factors between adjacent elements were less than 1.1. Almost complete separation of americium and californium was obtained in a test which used preferential stripping from 2-ethylhexyl phenylphosphonic acid [2-EH(ØP)A] into hydrochloric acid solutions. An ion exchanger was prepared for this test by adsorbing the 2-EH(ØP)A on powdered glass and using it as a static exchanger bed. The americium was preferentially stripped with 2 M HCl, and a single-stage separation factor of 18 was obtained.

Tertiary amine extraction was found to be an attractive alternative method for recovering americium and curium from nitrate raffinate solutions of the ion exchange process for the recovery of plutonium. By adjusting the nitrate solution to 3 M $LiNO_3$--1 M $Al(NO_3)_3$ and then extracting with 0.6 M Alamine 336·HNO_3 in diethylbenzene, distribution coefficient of 8.0 was obtained for americium and 5.5 for curium. By stripping the actinides from the amine with hydrochloric acid, the conversion from the nitrate to chloride systems can be accomplished by this procedure.

Process Equipment Development. -- A system composed of three pulse columns was used to study the Tramex process. When the column was fitted with only sieve plates (1/32-in. holes, 5% free area, 1/4-in. spacing), the smallest stage height observed was 28 in. for the scrub section of the process. This stage height was reduced to 18 in. by providing four coalescence sections consisting of 2-in. spaces filled with Teflon Raschig rings (1/4 x 1/8 in.). This stage height is adequate for the proposed TRU pulse column if it can be provided by a packing more radiation resistant than Teflon.

In tests to determine resistance to gamma radiation, 11 protective coatings remained servicable after exposure to more than 10^9 rads when tested in deionized water. In air, all coatings were more resistant to radiation, and 16 coatings were servicable after exposure to more than 3×10^9 rads.

Ten of the commercial disconnect clamps were tested for misalignment tendencies. Measurements made at 60 lb-ft of torque gave a misalignment ranging from 1.1 to 1.8°. The acceptable limit is 0.5°. The contact surface of the upper clamp arm was then machined at 1.5°. Subsequent misalignment tests indicated acceptable values for all clamps except one, which gave 0.8° of misalignment.

Five 3/8-in. tantalum disconnects made by forming the cones on the tubing itself leaked excessively during tests. The sealing surfaces of these specimens will be burnished with a rolling device and then retested. This procedure should improve the sealing of the tantalum disconnects.

Transuranium Facility. -- The Title II design for the Transuranium Facility was completed, and the lump-sum contract portion of the construction job is being submitted for bids. It is expected that construction will begin by July 1, 1963. Procurement of special materials and equipment for the facility by ORNL was begun. Orders were placed for Hastelloy C and stainless-steel tube plate, tubing, and fittings. Requests for bids were issued for several other fabricated items.

Design of chemical process equipment was started in the Design Group at the Oak Ridge Gaseous Diffusion Plant. A design of a Zircaloy-2 dissolver was issued for comment. Design is in progress on the following: the arrangement and mounting of disconnects in the tank pit, the hot

disconnect well, line bundles and track support, the service plugs for bringing lines into the tank pit, and the in-cell sampler.

Satisfactory bids were received and orders placed for Zircaloy-2 for the dissolver and one solvent extraction equipment rack.

A test ion exchange column was designed and is being built. The final 15 engineering flowsheets are complete and ready for checking.

ISOTOPE DEVELOPMENT PROGRAM

Cm^{242} Preparation. -- The recommended conditions for the Tramex process were modified in three respects to obtain the decontamination necessary for the processing of Cm^{242}: (1) Aluminum chloride was added to the feed to simulate the aluminum used as target material. (2) More-concentrated hydrochloric acid (8 M) was substituted for the stripping solution to improve the zirconium, niobium, and iron decontamination. (3) A scrub of freshly prepared Alamine $336 \cdot HNO_2$ was added to improve ruthenium decontamination. Ruthenium, zirconium, niobium, and rare earth decontamination factors were $\geq 10^4$ in laboratory mixer-settlers with simulated feed containing tracers.

Although irradiation tests of 0.6 M Alamine $336 \cdot HCl$--diethylbenzene indicated no difficulties up to 300 whr/liter, the loss of acid in Tramex feed by radiolysis at high activity levels poses a potentially serious problem. Experimentally determined G values for acid destruction were between 4.0 and 5.6 molecules per 100 ev. Unless methods for averting this acid loss can be found, it will be necessary to add makeup acid to prevent the precipitation of hydroxides.

Stable Tramex feeds of low acidity, which are required for actinide extraction, 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.

INTERNAL DISTRIBUTION

1. R. E. Blanco
2. J. O. Blomeke
3. J. C. Bresee
4. K. B. Brown
5. F. R. Bruce
6. F. L. Culler
7. D. E. Ferguson
8. H. E. Goeller
9. A. T. Gresky
10. M. J. Skinner
11. E. G. Struxness
12. W. E. Unger
13. M. E. Whatley
14. H. B. Whetsel
15. R. G. Wymer

16-17. Central Research Library
18. Document Reference Section
19-31. Laboratory Records
32. Laboratory Records (RC)

EXTERNAL DISTRIBUTION

33. P. C. Aebersold, Washington AEC
34. B. Anderson, COO
35. E. L. Anderson, Washington AEC
36. W. Belter, Washington AEC
37. S. Bernstein, UCNC, Paducah
38. J. A. Buckham, ICPP
39. J. T. Cristy, HOO
40. A. Friedman, Washington AEC
41. R. R. Hayward, AI
42. K. K. Kennedy, IDO
43. S. Lawroski, ANL
44. J. A. Lieberman, Washington AEC
45-46. J. A. McBride, ICPP
47. B. Manowitz, BNL
48. J. W. Morris, SRP
49. H. Pearlman, AI
50. W. H. Reas, HAPO
51. B. Schwartz, Washington AEC
52. J. M. Simmons, Washington AEC
53. C. M. Slansky, ICPP
54. C. E. Stevenson, IDO (ANL)
55. V. R. Thayer, du Pont, Wilmington
56. L. Topper, NYOO
57. A. F. Voigt, ISC
58. W. E. Winsche, BNL
59. L. L. Wyman, NBS
60-74. DTIE
75. Research and Development, ORO