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ABSTRACT

Process development studies are reported on Power Reactor Fuel Processing (PuO_2 Pellet Dissolution, Chloride Volatility--Solvent Extraction of HTGR Fuel, Physical Properties of ThOCl_2 --Azeotropic HCl Solutions, Flowsheet Development of UC Fuel, Solvent Extraction Studies, Shear and Leach Studies, Fluoride Volatility Processing: Aluminum Alloy Flowsheet Development, Fluoride Volatility Processing: Retention of TeF_6 , and Fluoride Volatility Pilot Plant) and Chemistry Research (Chemical Applications of Nuclear Explosions, Ion Exchange Studies, Mechanisms of Separations Processes, Solvent Extraction Research, Process Chemistry of U and Th, Solvent Extraction Studies, and Volatility Studies).

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POWER REACTOR FUEL PROCESSING

PuO₂ Pellet Dissolution. -- Only 83% of a sintered PuO₂ pellet having a density 90% of theoretical was dissolved by refluxing 86 hr in 14 M HNO₃--0.05 M HF. There was no appreciable variation of the apparent surface area during the dissolution. The plutonium dissolution rate was about 1% per hour and was constant throughout the dissolution.

Chloride Volatility--Solvent Extraction of HTGR Fuel. -- Samples of HTGR fuel (7% Th--1.5% U--91.5% C) weighing about 30 g each were burned in oxygen at 800°C in 4 hr; the ash was chlorinated in 15% CCl₄--N₂ vapor at 500°C in 1 to 2 hr, and the nonvolatile chlorides were completely dissolved in cold water. The product solutions were evaporated to more than 500 g of thorium per liter and more than 100 g of uranium per liter and made 1 M in H₂O₂ at 80°C to oxidize slightly soluble UCl₄ to the more soluble UO₂Cl₂, to remove free chloride and to lower the freezing point to below room temperature.

Various methods of recovering uranium from the product solution by solvent extraction are being investigated. Over 99.9% of the uranium was extracted, using a volume of 30% TBP--Amsco equal to the volume of aqueous solution, in five batch extractions when the aqueous solution was made 1 M in HNO₃; seven batch extractions were required to achieve 99.9% uranium recovery when no nitrate was added. In both cases 92 to 96% of the thorium remained in the aqueous phase.

Physical Properties of ThOCl₂--Azeotropic HCl Solutions. -- Thorium oxychloride was found to be miscible in all proportions with azeotropic hydrochloric acid. The melting points of the solutions increased with increasing ThOCl₂ content and were found to be 35, 60, and about 140°C

at 4, 5, and 8 M ThOCl_2 , respectively. The melting and boiling point curves meet at approximately 8 M ThOCl_2 . The melting point of azeotropic 5 M ThOCl_2 -- HCl solution was lowered from 60°C to -20°C by adding 1.5 moles of H_2O_2 per mole of thorium at 80°C . The hydrogen peroxide reacted with chloride to form chlorine, which volatilized out of the solution. After the addition of the hydrogen peroxide, the solution was still stable at temperatures as high as the boiling point (more than 100°C) and could be boiled down to any desired concentration. The solution began to appear viscous at 25°C if boiled down to more than 5 M in thorium.

Flowsheet Development of UC Fuel. -- Laboratory development of an aqueous process flowsheet for sheared uranium monocarbide reactor fuel was continued. Unirradiated UC (200 g, 5.2% C) containing dicarbide impurity was hydrolyzed in 300 ml of water at 90°C . It took 8 hr for complete hydrolysis, as evidenced by cessation of gas evolution. After cooling, 300 ml of 15.8 M HNO_3 containing Pu(IV) tracer was added to dissolve the UO_2 slurry in 2 hr. A wine-red solution was produced, and it contained traces of yellow organic solids.

The raw feed solution was treated with 0.04 M KMnO_4 for 4 hr at reflux. The MnO_2 precipitate was removed by filtration; the filtrate had the characteristic yellow uranyl nitrate color, a qualitative indication that the organic acids has been destroyed. The precipitate was washed three times with 50-ml portions of 4 M HNO_3 . Analysis showed that, before washing, the precipitate retained 0.03 and 0.64% of the total uranium and plutonium, respectively. Repeated washing reduced these losses to 0.028 and 0.13%, respectively; however, analysis indicated that additional or more efficient washing would have recovered more of the plutonium. The

filtered feed solution was treated with 0.1 M NaNO_2 for 1 hr at 40°C to reduce Pu(VI) to Pu(IV).

The feed was then processed batch countercurrently through four volumetric throughputs of the Purex first cycle of solvent extraction.

Solvent extraction performance was satisfactory, and no emulsions or interfacial "cruds" were observed. Uranium recovery was quantitative. The maximum uranium loss occurred during stripping ($4.5 \times 10^{-4}\%$ after seven stages). Plutonium extraction loss was 0.05% at the seventh extraction stage; however, the stage data show that plutonium was still being extracted at this point. Extraction coefficients for the last four stages suggest the presence of Pu(VI). Plutonium partition loss was nil after seven partition stages; no plutonium could be detected by pulse counting the BU stream.

The use of impure UC in this experiment represents a "worst-case" condition for uranium monocarbide fuel processing, since the actual reactor fuel is expected to be of much higher purity. The results indicate, however, that even such impure fuel as used in these tests can be processed satisfactorily.

Solvent Extraction Studies. -- An unidentified substance in slightly degraded di sec butyl phenyl phosphonate (DSBPP) in diethylbenzene (DEB) resulted in inaccurate analyses for uranium. Uranium analytical data obtained in the countercurrent stripping of uranium from 2.5% DSBPP in DEB containing 2.5 g of uranium per liter, after the uranium-loaded solvent was stored for four days, were in error by a factor of at least 2 for all except the first stage.

The distribution coefficient of protactinium tracer in 30% tributyl phosphate in n-dodecane decreased from 4.72 ± 0.46 (with $0.49 \pm 0.06\%$

unextractable) in 5 N HNO₃ to 0.78 ± 0.05 (with 66 ± 15% unextractable) in 5 N HNO₃ containing 0.01 N HF.

Laboratory countercurrent batch extraction experiments indicated that rongalite (CH₂HOSONa·2H₂O) is more efficient than ferrous sulfamate or uranium(IV) for selective stripping of plutonium from uranium in 30% TBP in n-dodecane. Experimental conditions were as follows:

Feed: uranium, 100 g/liter; plutonium, 6 x 10⁶ counts min⁻¹ ml⁻¹;
30% TBP in n-dodecane; 1 volume

Strip: (a) 0.06 M ferrous sulfamate, 0.05 M HNO₃; 0.083 volume; or
(b) 0.03 M uranium(IV), 0.1 M HNO₃; 0.083 volume; or,
(c) 0.066 M rongalite, 1.0 M HNO₃; 0.083 volume

Scrub: 30% TBP in n-dodecane, 0.1 volume

Stages: Six strip, six scrub

The rongalite reduced the plutonium in the uranium by a factor of 8000, whereas the ferrous sulfamate and uranium(IV) reduced the amount of plutonium by factors of 200 and 15, respectively. Reduction of plutonium with rongalite results in the liberation of SO₂, which decreases the amounts of solids in the aqueous waste.

Protactinium Adsorption Studies. -- The amount of protactinium adsorbed in 24-hr contacts with 10 g of unfired Vycor glass per liter of solution containing 2 x 10⁶ counts min⁻¹ ml⁻¹ Pa²³³ and 10 M HNO₃ ranged from 97.8 to 98.3% for three different batches of glass.

Shear and Leach Studies. -- Bench-scale leaching tests with 1/2-in. sheared lengths of stainless-steel-clad ThO₂-UO₂ unirradiated fuel in a 4-in.-diam cylindrical dissolver showed that, for a composite product containing about 125 g/liter of thorium and 9 M HNO₃, virtually the same

time (about 12 hr) was required to achieve 99.9% dissolution whether the dissolvent was employed in a single batch or in two successive half-batches.

Heat transfer tests made on the steam-jacketed leacher, using 40 psi steam pressure, showed that the boil-up rate for the product solution rose from 0.09 to 0.14 lb/min as the air sparge, used to suspend $\text{ThO}_2\text{-UO}_2$ fines, was increased from 0 to 0.06 scfm. Increasing the sparge rate up to 0.19 scfm produced no further increase in boil-up rate but did increase the foaming.

Fluoride Volatility Processing: Aluminum Alloy Flowsheet Development. -- According to visually determined liquidus data, molten KF-ZrF_4 mixtures can dissolve sufficient AlF_3 to be useful as the solvent in volatility processing of aluminum-alloy or aluminum-clad fuel elements. At 600°C , the maximum acceptable temperature for the process, a solvent containing 63 mole % KF dissolved up to 15 mole % AlF_3 . Also, the addition of KF to this mixture permitted the dissolution of a substantial increment of AlF_3 without exceeding the 600°C temperature limit. The limit would not permit the use of more KF in the initial solvent. Recent results, obtained in a continuing investigation of $\text{KF-ZrF}_4\text{-AlF}_3$ phase equilibria, indicate that a single addition of 15 mole % KF during dissolution increased the solubility of AlF_3 from 15 mole % to 26 mole %.

Two immiscible liquids were found in the KF-AlF_3 binary system above 53 mole % AlF_3 at 980°C . The two-liquid region apparently extends into the $\text{KF-ZrF}_4\text{-AlF}_3$ ternary system but not to compositions of present interest as volatility solvents.

Laboratory dissolution studies further indicate the apparent feasibility of the KF-ZrF_4 system for aluminum-based fuels. The dissolution

of aluminum in KF-ZrF_4 salt at 600°C by hydrofluorination was found to proceed much more rapidly at 37 mole % ZrF_4 than was observed in previous work at 30 mole % ZrF_4 . Use of 70-30 mole % KF-ZrF_4 salt with 5 mole % AlF_3 was reported to give dissolution rates of 10 mils/hr, whereas in recent work with 63-37 mole % KF-ZrF_4 and no AlF_3 present, dissolution rates of 40 to 50 mils/hr were achieved. Although it is possible that the absence of AlF_3 promotes fast hydrofluorination, the more probable explanation is that the ZrF_4 reduction accounts for part of the aluminum dissolution. A black material or sludge is always formed, probably colloidal zirconium metal. This sludge sometimes adheres to the dissolving aluminum; at other times it disperses in the salt and in some cases leads to difficulty in passage of the HF through the salt. This problem does not appear serious, however, since several complete process dissolution tests have been successfully made, ending up at about 15 mole % AlF_3 , the approximate concentration limit imposed by the $\text{KF-ZrF}_4\text{-AlF}_3$ phase diagram in a single step. Tests have also demonstrated that fluorination of the salt at 550°C results in volatilization of more than 99.9% of the uranium (initial fuel, 3.6% uranium in aluminum).

Fluoride Volatility Processing: Retention of TeF_6 . -- More laboratory tests were made to determine the absorption of $\text{TeF}_6\text{-F}_2$ gas mixtures in 10% aqueous KOH solution. By use of two consecutive packed columns 1 in. in diameter and 20 in. long, with countercurrent flow, TeF_6 absorption efficiencies of 46, 68, and 67% were obtained at 25, 50, and 70°C , respectively, for both columns together. In contrast, more than 99% of the fluorine was trapped in the first column alone at all three temperatures. Tellurium hexafluoride thus appears to be somewhat more stable than fluorine to hydrolysis with aqueous caustic solution.

Fluoride Volatility Pilot Plant. -- Five processing runs were completed in the Fluoride Volatility Pilot Plant on Zr-U alloy following a 30-week shutdown for equipment decontamination and modification. Three of the runs, TU-13 to TU-15, were performed to study the performance of the equipment and also to study selective desorption techniques as a method of improving product quality (by separating MoF_6 from the UF_6). Two runs, R-7 and R-8, were made to study fission product behavior and uranium decontamination from fission products while processing 13-month and 6-month decayed alloy.

Dissolution rates ranged from 2.0 to 2.9 kg/hr during cold runs (based on 90% completion of the dissolution to minimize tail-out effects) with HF utilization efficiencies per pass through the dissolver of 24 to 34%. Higher dissolution rates and HF utilizations (3.4 and 3.7 kg/hr, and 41 and 49%, respectively) were obtained during hot runs, but the increase may not be a result of fuel burnup.

The uranium was separated as UF_6 from the molten fluoride melt in all runs by reaction with fluorine at 500°C . The average fluorine utilization efficiency was about 5% during cold runs, and 14 and 7% during hot runs R-7 and R-8, respectively. Nonrecoverable uranium losses, mainly in the waste salt, were less than 0.46% in all runs.

A selective desorption procedure was used during all runs to improve product quality by separating molybdenum from the UF_6 . The procedure consisted in desorbing MoF_6 from the UF_6 at 150°C and collecting the molybdenum (and the milligram quantities of uranium evolved) on a chemical trap. No product improvement was obtained during cold runs since up to 2000 ppm of molybdenum collected in the product. However, the high

molybdenum contamination was attributed to HF dilution of the product in these runs (up to 30 wt %) and not to ineffective molybdenum removal during desorption. Only 9 ppm of molybdenum collected with the R-7 product, which was not contaminated with HF. Significantly, this was the lowest molybdenum level in the product during the zirconium-uranium alloy program (about twentyfold below maximum AEC molybdenum specifications for enriched UF₆ return).

Total cation impurity levels in the product from cold runs TU-13 through TU-15 were about 3200 ppm, or tenfold higher than AEC specifications (because of HF co-contamination). However, cation specifications were met in run R-7 on 13-month decayed alloy (total cations less than 171 ppm). During this run, Tc⁹⁹ and Np²³⁷ were removed from the UF₆ to less than 1 and 4 ppm, respectively, in the product by complexing with MgF₂ at 100°C. A substantial quantity of uranium (at least 200 g) was also retained on the bed by both physical adsorption and deposition in the presence of moisture.

During run R-7, on 13-month decayed alloy, overall-process decontamination factors ranged from greater than 10⁵ to greater than 10⁹ for volatile fission products (Sb¹²⁵, I¹³¹, Te, Ru¹⁰⁶, and Nb⁹⁵), and from greater than 10⁸ to greater than 10⁹ for nonvolatile fission products (Zr⁹⁵, Sr⁹⁰, and Cs¹³⁷). These factors include uranium decontamination during both major steps of the process, dissolution and product recovery, and represent the ratio of the fission product activity in the fuel to the corresponding activity in the product. The specific fission product activities in the fuel were evaluated from reactor data, with the fuel burnup calculated by the CRUNCH code on the IBM 7090 computer. Product

activities were measured by standard techniques; when the fission product activity was below the analytical limit of detection, the lower limit was used as the product activity for the decontamination factor calculation. Without exception, the dissolution decontamination factors (DF's) were small compared to the product recovery DF's. Dissolution DF's ranged, for volatile fission products, from 10 for I^{131} to 750 for Ru^{106} . Strontium-90 and Cs^{137} decontamination during dissolution was negligible (DF of 2 or 3).

Considerable I^{131} and Nb^{95} (about 80% and 97%, respectively) remained in the salt during fluorination to separate the uranium. About 85 and 95% of the Ru^{106} and tellurium, respectively, in the fluorinator feed salt followed the UF_6 during fluorination.

The UF_6 product from run R-7 met AEC specification for gamma activity. Beta specifications were also met within the limit of error of the counter. In all cases, specific fission product activities were at or below the analytical lower limit of detection in the product.

CHEMISTRY RESEARCH

Chemical Applications of Nuclear Explosions. -- Acid leaching of water-insoluble residue from a composite Gnome sample showed that 80% of the plutonium and the total rare earths are leached by 10 ml of 5 N HNO_3 per gram of residue at room temperature (27°C) with 73% dissolution of the residue. Increasing the acid excess to 100 ml of 5 N acid per gram only increased the plutonium and gross beta extraction by about 10% and the dissolution of residue by about 10%. The data indicate that dissolution of the residue is necessary to leach appreciable quantities of activity from the residues. The amount of activity and dissolved residue leached increased nearly linearly when the acid volume was increased from 3 to 10 ml per gram, but it increased only 10% for a tenfold increase in acid volume thereafter. This indicated that all readily soluble components are dissolved by 10 ml of acid per gram of residue. Increasing the leaching temperature from room to reflux temperature increased the amount of plutonium and total rare earths leached by 10%. Below about 30 volumes of acid per gram, the samples were difficult to filter. Varying the nitric acid concentration at a constant 30 volumes of acid per gram increased the amount of plutonium and total rare earths leached from 77 to 92% and from 84 to 91%, respectively, when the acid concentration was increased from 1 N to 10 N. Acid consumption was 9 to 13 meq/g.

Ion Exchange Studies. -- An examination was made of effluent-water solutions collected from previously reported flowing-water irradiation experiments of Dowex 50W cation exchange resin. Each liquid volume was reduced by a factor of about 50 in a vacuum distillation unit that

maintained the solution at or below room temperature to minimize the possibility of volatilizing or decomposing any organic constituents. Chemical analyses showed that most of the concentrates of each run contained soluble sulfates and sulfonates. Visually, the first concentrates of each run were colorless; remaining concentrates become progressively darker, varying from light straw to dark brown and containing increasing quantities of suspended solids. Ultracentrifuge and light-scattering studies indicated that these suspended solids had a density of about unity and a molecular weight of several million.

One interpretation of the available data on the radiation degradation of this cation exchange resin is as follows. Initially, the only degradation process involves cleavage of active sulfonate groups from the resin. These dissolve, while being partly converted to sulfate, in the flowing-water stream as indicated by the occurrence of maximum acidity in the effluent stream at the beginning of each exposure period. The resin matrix does not start to break up until a substantial fraction of active sulfonate sites is lost. Then, along with the formation of water-soluble radiolysis products, small particles of the matrix, of several millions in molecular weight, begin to break off the beads and enter the effluent stream in some manner that does not lead to fragmentation of the resin beads.

Mechanisms of Separations Processes. -- The transpirational vapor pressure method is being used to obtain the activities of tributyl phosphate (TBP) in two-phase systems of uranyl nitrate, tributyl phosphate, and water at 25°C. Tributyl phosphate transpired is being determined by P^{32} -beta counting of the labeled phosphate. As the uranium

concentration in the organic phase decreases from 1.59 to 1.43 M, the vapor pressure of TBP increases from 0.0044 to 0.055 μ Hg; the corresponding rational activities, based on a vapor pressure of anhydrous TBP of 0.527 μ , increase from 0.0083 to 0.104. The standard deviations of these values have not yet been established.

Analysis of previously reported data show that the excess partial molar free energy of TBP in water-saturated TBP is +256 cal/mole of TBP. As nitric acid is added to a two-phase system, the excess partial molar free energy of TBP decreases but becomes negative only when the concentration of acid in the organic phase is about 1 M, corresponding to a ratio $(\text{HNO}_3)_{\text{org}}/(\text{TBP}_s)$ of about 0.3. The sign of this excess free energy and its magnitude suggest that there is a weak repulsion between TBP and water molecules; they do not support the concept of formation of a complex such as $\text{TBP}\cdot\text{H}_2\text{O}$.

Solvent Extraction Research. -- Large separations between the lanthanide and transplutonium element groups were obtained by extraction from chloride-free solutions. The extractant was di(2-ethylhexyl)phosphoric acid in a hydrocarbon diluent with or without a modifier. The aqueous phase was a mixed solution of a sodium salt of ethylenediaminetetraacetic acid, N-hydroxyethylethylenediaminetriacetic acid, or diethylenetriaminepentaacetic acid with any of several common carboxylic acids, particularly glycolic acid. Cerium extracted better than americium by factors up to more than 50, while europium was usually still more extractable. Differences within each group were small. The presence of nitrate did not decrease separations. Distribution coefficients, separation factors, and the order of preference were dependent

on the identity and both relative and absolute concentrations of the pair of mixed acids and on the composition of the diluent.

Hydroxamic acids, $R \overset{\text{O}}{\parallel} \text{C} - \overset{\text{H}}{\text{N}} - \text{OH}$, showed potential for extraction of zirconium, niobium, hafnium, and strontium. Strontium was extracted by 0.1 M octanohydroxamic acid in hexone from 0.5 M NaNO_3 , with a maximum extraction coefficient of $E_a^{\text{O}} = 8$ and separation factor from sodium of $SF_{\text{Na}}^{\text{Sr}} \approx 50$ at pH 10.5. Cesium was not extracted. The available hydroxamic acids decomposed when exposed to nitric acid or alkaline solutions but appeared to be stable in extraction from hydrochloric acid.

It was shown previously that tributyl phosphate (TBP) when used in aromatic rather than aliphatic diluents gave improved performance with regard to radiation stability, uranium extraction power, and separation of uranium from fission products. Similar results have now been obtained with a wider range of aromatic diluents. Solutions of TBP in diethylbenzenes, butylbenzene, or trimethylbenzenes extracted uranium from 2 M HNO_3 with coefficients about 50% higher than those obtained with TBP in Amsco 125-82 solutions. Separation factors of uranium/hafnium were about 2 times higher.

Process Chemistry of U and Th. -- In mineral beneficiation studies on Conway granite, about 70% of the thorium was recovered in a heavy mineral separate which comprised 8% of the ore weight. Magnetic fractionation of the ore gave a magnetic product representing about 20% of the ore weight and containing about 60% of the thorium.

Solvent Extraction Studies. -- The flow capacity of pulsed columns at a high flow ratio (100:1) of aqueous to organic phase is virtually the same as that at a more conventional ratio of 5:1. For the system,

30% tributylphosphate-Amsco vs 4 M NaNO_3 , the total flow rate in a standard sieve plate (1/8 in. holes, 23% free area) column, operated with the aqueous phase continuous, increased from 520 to 3100 gal hr^{-1} ft^{-2} as the pulse frequency was decreased from 90 to 35 cpm. The flow capacity of a nozzle-plate (1/8 in. holes, 10% free area) column operated with the organic phase continuous was significantly less, ranging from 190 to 820 gal hr^{-1} ft^{-2} as pulse frequency was decreased from 70 to 35 cpm.

Volatility Studies. -- The results of several stirred reactor tests have demonstrated that mechanical agitation does not significantly improve the volatilization of PuF_6 from 31-24-45 mole % LiF-NaF-ZrF_4 upon fluorination at 600°C. In all cases the half-reaction time value was 3 to 5 hr, or about the same as in unstirred melts. In one 4-hr test where optimum conditions prevailed, the stirrer was the centrifugal-tube-type operated at 2200 rpm.

Using an alundum reactor and an alundum fluorine inlet dip tube, a half-reaction time of 10 hr was obtained in another unstirred PuF_6 fused-salt-volatilization test. This run also indicated no significant difference from the normal case since use of about twice the usual amount of salt probably led to the long half-time.

From the results of the stirring tests and the alundum-vessel run, it appears that the slow rate of PuF_6 volatilization is due neither to diffusion-rate-controlling mechanisms nor to back reaction of the oxidized plutonium with the nickel container wall. Tests are now being planned to determine whether a higher fluorine pressure (at the same flow rate) or a change in the salt composition will result in some improvement in the volatilization rate.

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