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Consolidated Fuel Reprocessing Program

SOLVENT EXTRACTION FLOWSHEET STUDIES WITH IRRADIATED FUEL FROM THE FAST FLUX TEST FACILITY

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A small batch (~2 kg) of irradiated fuel from the Fast Flux Test Facility (FFTF) was recently used as feed for some solvent extraction experiments that were made in the Solvent Extraction Test Facility (SETF). This fuel, which was from series DEA-1, had been irradiated to a burnup of ~0.26 TJ/kg (~3000 MWd/t) and cooled for ~1 year. The SETF is located in one of the heavily shielded hot cells of the Transuranium Processing Plant at the Oak Ridge National Laboratory. This facility has been used during the past several years to demonstrate the suitability of conceptual flowsheets for the solvent extraction processing of irradiated light-water reactor and fast breeder reactor fuels. Results of these experiments have provided information on uranium and plutonium separation and recoveries, fission product decontamination, comparisons of flowsheet options, and general operability of the system.

Equipment Description

Three solvent extraction contactors are used in the SETF; each is a 16-stage mixer-settler which has a liquid holdup of about 80 cm^3 per stage.

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Associated hot-cell equipment includes several effluent catch tanks and equipment for feed preparation and product purification. The feed preparation equipment includes a batch dissolver, a feed adjustment tank, a variety of filters which are used to clarify the feed solutions, and a feed metering and flow rate control system. The product purification equipment consists primarily of an anion exchange column. In addition, a precipitator tank, a filter, and a calcination furnace for conversion of plutonium products to the oxide form are available in a nearby hot cell.

Feed Preparation

The FFTF fuel pins were cut into ~25-mm-long pieces with a single-pin shear. The fuel was dissolved by immersing the pieces in ~7 kmol/m^3 (M) HNO_3 and heating the solution at ~370 K (~95°C) for ~14 ks (4 h). The resulting dissolver solution was (1) clarified by means of filtration, (2) sparged with NO_x gas to ensure that the plutonium was in the tetravalent state, and (3) adjusted with dilute HNO_3 to the desired acidity and heavy metal concentrations. In addition, the adjusted feed was reclarified as it was fed to the mixer-settlers by means of an in-line filter.

Solvent Extraction Tests

Two first-cycle flowsheet options for the Breeder Reprocessing Engineering Test facility were tested. In each flowsheet the uranium and plutonium were coextracted in the first contactor (A-bank) with 30 vol % tri-n-butyl phosphate (TBP) in a normal paraffin (NPH) diluent; then the pregnant solvent was scrubbed with two HNO_3 reagent streams. The conditions for the scrub section (reagent stream concentrations and stages) were varied slightly for each run. The losses of uranium and plutonium to the aqueous waste were typically ~0.003% and ~0.01%, respectively. Fission product decontamination factors (DFs) in the extraction bank for ^{95}Zr, ^{106}Ru, and ^{144}Ce were in the range of 90-700, 600-3000, and >1 x 10^5, respectively, for the two tests. The partitioning of the uranium and plutonium, however, was done using either (1) reductive stripping of the plutonium followed by uranium back-extraction with organic solvent or (2) costripping followed by selective uranium extraction.

In the first method, which is very similar to the conventional Purex process, partitioning was accomplished in B-bank by using (1) a dilute HNO_3 strip solution containing hydroxylamine nitrate (HAN) to reduce the plutonium, and (2) a TBP-NPH scrub to back-extract the uranium. This flowsheet was initially tested with a HAN/plutonium mole ratio of only ~2, and significant plutonium losses were measured to the uranium product stream. In a retest with synthetic feed containing uranium and plutonium, but no fission products, it had a HAN/Pu mole ratio of ~4. This run resulted in a plutonium loss of only ~0.009% and a uranium DF of ~2 x 10^4; however, analyses of the interstage samples showed that significant reoxidation and refluxing of plutonium were occurring.

In the second method, the heavy metals were first costripped from the solvent in B-bank with dilute HNO_3 containing HAN and hydrazine (HAN/N_2H_4/Pu ratio of 2/0.6/1) to reduce the plutonium. Then the aqueous product was routed to C-bank where the uranium was selectively extracted into a fresh TBP-NPH stream, leaving the plutonium in the aqueous effluent. The results from this run indicated good uranium-plutonium recovery and separation. The plutonium losses to the two waste solvent streams, from B-bank and C-bank, were 0.001% and 0.006%, respectively, and the uranium DF was ~6000. Although some reoxidation and reflux of plutonium also occurred in the partition contactor for this run, the situation seemed to stabilize at a low level early in the run.

Both partitioning methods were troubled to varying degrees with reoxidation and refluxing of plutonium, regardless of whether HAN alone or HAN in combination with hydrazine was used for reducing the plutonium. The organic back-scrub method, which used only HAN, had more extensive reoxidation problems but yielded the more concentrated plutonium product, ~30 kg/m^3. The selective extraction method, which used HAN plus hydrazine, had less reoxidation of plutonium but yielded a less concentrated product, ~9 kg/m^3. The plutonium concentration in these flowsheets may be a significant factor in the rates of reoxidation. A comparable quantity of hydrazine in the organic back-scrub flowsheet would probably have decreased the reoxidation but may not have eliminated it; similarly, increasing the plutonium concentration in the selective uranium extraction flowsheet may have resulted in more reoxidation.