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ENRICHED URANIUM RECOVERY FLOWSHEET IMPROVEMENTS

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ABSTRACT

Savannah River uses 7.5% tributyl phosphate (TBP) to recover and purify enriched uranium. Adequate decontamination from fission products is necessary to reduce personnel exposure and to ensure that the enriched uranium product meets specifications. Initial decontamination of the enriched uranium from the fission products is carried out in the 1A bank, 16 stages of mixer-settlers. Separation of the enriched uranium from the fission product, Zr-95, has been adequate, but excessive solvent degradation caused by the long phase contact times in the mixer-settlers has restrained the Zr-95 decontamination factor (DF).

An experimental program is investigating the replacement of the current 1A bank with either centrifugal contactors or a combination of centrifugal contactors and mixer-settlers. Experimental work completed has compared laboratory-scale centrifugal contactors and mixer-settlers for Zr-95 removal efficiencies. Feed solutions spiked with actual plant solutions were used. The Zr-95 DF was significantly better in the mixer-settlers than in the centrifugal contactors.

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As a result of this experimental study, a hybrid equipment flowsheet has been proposed for plant use. The hybrid equipment flowsheet combines the advantages of both types of solvent extraction equipment. Centrifugal contractors would be utilized in the extraction and initial scrub sections, followed by additional scrub stages of mixer-settlers.

INTRODUCTION

An experimental program was begun at the Savannah River Laboratory (SRL) in the spring of 1985 to evaluate methods to increase the Zr-95 decontamination factor (DF) in the enriched uranium recovery operation at Savannah River Plant (SRP). Some of the incentives for this work are to reduce operator exposure from radiation, reduce waste, and improve product quality. The increased DF would also allow processing of shorter cooled fuel and could increase capacity of the operation (Slide 1).

ZIRCONIUM CHEMISTRY

The aqueous phase chemistry of zirconium is complex. Zirconium is known to exist as many different chemical species in the aqueous phase. Its extractability is dependent upon the process temperature, acidity, the amount of time the organic and aqueous phases are contacted, and the amount of available tributyl phosphate (TBP) in the organic phase (Slide 2).[1]

The organic phase chemistry of zirconium is even more complicated. Zirconium can complex with the organic phase in several ways. The most common complex, and the easiest one to remove from the organic phase, is the normal extracted species, $Zr(NO_3)_4$ * 2TBP. Zirconium can also form a solvated species in the organic phase with three or more TBP molecules; and, in addition, it can form complexes with the various TBP and diluent degradation products. The distribution coefficients for zirconium in the degradation complexes are greater than for the normal species, and the degradation complexes are much more difficult to scrub from the organic phase (Slide 3). Therefore, it is important to reduce the amount of solvent degradation products formed, as well as the phase contact time of the organic and aqueous phases in the extraction section of the flowsheet.[1,2,3]

LABORATORY STUDIES

The experimental work was conducted in a large glove box facility located at SRL. This facility is capable of handling only trace levels of plant solutions. Experimental equipment consisted of a 16-stage centrifugal contactor unit approximately 1/50,000 plant scale. A 16-stage, 1/10,000 plant scale, mixer-settler unit was also used. The stage residence time of the mixer-settler approximates that of actual plant equipment. The centrifugal contactor residence time approximates what would be typical if contactors were installed as proposed (Slide 4). A 16-stage flowsheet, patterned after SRP's enriched uranium recovery flowsheet, was used for the experimental work (Slide 5).

Initially, the experimental work evaluated the Zr-95 scrubbing efficiencies of the equipment. Experimental runs were made using a feed solution spiked with actual plant solution. New TBP solutions were used for this work (Slide 6). Temperature effects were evaluated for both types of equipment, with results presented for centrifugal contactors (Slide 7). In agreement with previous studies, scrubbing of the zirconium improved as temperature increased for both types of solvent extraction equipment. Residence time effects were observed by comparing the organic phase zirconium profile for each type of equipment. Increased scrubbing results of all experimental runs were expressed as zirconium DF's (Slide 9).[4] The mixer-settlers provided greater DF's in all runs as compared with the centrifugal contactors.

Solvent degradation effects were then investigated by injecting a spike of actual plant solvent into the system (Slide 10). A comparison of the organic phase zirconium profiles in the mixer-settlers revealed reduced scrubbing efficiencies with plant solvent present (Slides 11 and 12). Expressed again as zirconium DF, the mixer-settlers' performance decreased by a factor of 4.5, but was still better than the centrifugal contactors' performance.

SUMMARY

It was concluded from this work that to improve the zirconium DF, it will be necessary to increase scrub temperatures; increase phase contact time in the scrub section to allow for the removal of zirconium species complexed with higher orders of TBP and solvent degradation products; and decrease the amount of solvent degradation products generated. Therefore, a hybrid equipment flowsheet that would combine the advantages of both types of solvent extraction equipment is proposed (Slide 13).

HYBRID EQUIPMENT FLOWSHEET ADVANTAGES

Centrifugal contactors would be used in the extraction section to reduce the amount of solvent degradations products formed. This would also reduce the amount of waste generated by solvent washing and increase the DF. In the initial scrub section, centrifugal contactors would also be used because experimental results showed good scrubbing of the zirconium in three to four stages of centrifugals. This would also further protect the solvent from radiolytic degradation. Finally, to provide the long phase contact time necessary to scrub zirconium complexes from the organic phase, four to 12 stages of mixer-settlers would be used (Slide 14). An example of a hybrid equipment flowsheet is given showing how the equipment would be arranged (Slide 15).

PROGRAM

Initial experimental testing has been completed on a larger laboratory mixer-settler capable of handling the flow rates of the centrifugal contactors. Five, 4-stage units are currently being fabricated; and it is anticipated that by the third quarter of 1986, the unit will be assembled and tested. An extension to the original glove box facility used to house the experimental equipment is underway. This expansion will house another 16-stage centrifugal contactor unit and three, 4-stage, large-scale mixer-settler units. Initial testing of the hybrid equipment flowsheets using plant solution spikes will begin in the fourth quarter of 1986. After all preliminary experimental work with tracer solutions is completed, the flowsheets that show the most promise will then be tested using actual plant solutions. This should begin in early 1987 (Slide 16).

ACKNOWLEDGMENT

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