

## Actinide and Lanthanide Separation Studies for Spent Fuel Processing

L. K. Felker, P. V. Bonnesen, D. O. Campbell, E. D. Collins

*Oak Ridge National Laboratory: 1 Bethel Valley Rd., P.O. Box 2008, Oak Ridge, TN, 37831-6384, felkerlk@ornl.gov*

### INTRODUCTION

The Oak Ridge National Laboratory (ORNL) is participating in several areas of the Advanced Fuel Cycle Initiative Program. One of these areas is the group separation of the actinide (An) and lanthanide (Ln) series elements during the post-irradiation processing of the various types of reactor fuel. The objective of this processing step is to provide a segregated minor actinide fraction for further transmutation and a fission product fraction for disposal as waste. The most successful methods have used ion exchange or solvent extraction from concentrated LiCl solutions or ion exchange chromatography with specific organic complexing agents; both approaches lead to serious waste management problems.

A review of literature on this subject revealed that the current work of the European Commission studies<sup>1</sup> focused on a number of new reagents for accomplishing this actinide and lanthanide separation. The more promising reagents for this group separation were the dithiophosphinic acids and the triazinylpyridines. A study was initiated to investigate the separation of the actinide and lanthanide fission products using these types of reagents.

### DESCRIPTION OF WORK

Two of the reagents of interest, *bis*-(chlorophenyl)-dithiophosphinic acid and 2,6-*Bis*,6-di-*n*-propyl-1,2,4-triazin-3-yl-pyridine (BTP) were synthesized at ORNL in the Chemical Sciences Division and tested in the Nuclear Science and Technology Division using a highly irradiated plutonium target processing product fraction as the source of the minor actinides and rare earth fission products. Two additional dithiophosphinic acid reagents, diphenyl-dithiophosphinic acid and Cyanex® 301, were obtained from commercial vendors for comparison testing.

A synergist was added to the extractant system to increase the distribution of the actinide elements. Two synergists were tested in combination with the dithiophosphinic acid reagents. The synergists were trioctylphosphine

oxide and Cyanex® 923, which is a mixture of four trialkylphosphine oxides. The organic diluents for the extraction system were diethylbenzene and diisopropylbenzene for the dithiophosphinic acids and a dodecane/octanol mixture for the triazinylpyridine compound.

Batch contacts with the extractant and the aqueous feed solution were performed at phase ratios (o/a) of 0.2 for various contact times and acid concentrations. Analysis of the equilibrated phases yielded gamma count rates for americium (241 and 243) and europium (154 and 155). A spike solution containing 100 g/L rare earths as cerium, neodymium, and europium was prepared to increase the rare earth concentrations in the diluted aqueous feed to simulate typical processing concentrations. Distribution coefficients and separation factors were calculated from the average count rates for each element.

### RESULTS

Distribution coefficients and separation factors were measured for Eu and Am over a range of nitric acid concentrations from ~0.1 to 1 M, using two different dithiophosphinic acids in synergistic systems with TOPO in DEB. The values obtained were useful for a practical solvent extraction separation process, with extraction of Am and rejection of Eu. There was general agreement with the European work, although we used a different diluent. In the higher ranges of acidity there was precipitation or gel formation in the organic phase in some tests, and this phenomenon is not well understood. This is also in accord with European observations. This suggests that the primary problem with regard to application of the dithiophosphinic acid extractants may be their physical behavior rather than their chemical separation properties. This area will receive more emphasis in future work.

Similar testing is in progress with the BTP extractant. The Cyanex® 301 is being tested in parallel with the BTP for comparison purposes with the other dithiophosphinic acids.

## REFERENCES

1. “New partitioning techniques for minor actinides,” C. Madic, M. J. Hudson, J. O. Liljezin, J. P. Glatz, R. Nannicini, A. Facchini, Z. Kolarik, and R. Odoj, European Commission on Nuclear Science and Technology, EUR 19149 EN, 2000