Managing Zirconium Chemistry and Phase Compatibility in Combined Process Separations for Minor Actinide Partitioning

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ABSTRACT:

NEUP Program Supporting Fuel Cycle R&D Separations and Waste Forms call DE-FOA-0000799 requests long term R&D projects focusing on streamlining separation processes for advanced fuel cycles. An example of such a process relevant to the U.S. DOE FCR&D program would be one combining the functions of the TRUEX process for partitioning of lanthanides and minor actinides (from PUREX/UREX raffinates) with that of the TALSPEAK process for separating transplutonium actinides from fission product lanthanides. Experience teaches (and it has been demonstrated at the lab scale) that, with proper control, multiple process separation systems can be made to operate successfully. However, it is also recognized that considerable economies of scale could be achieved if multiple operations were merged into a single process based on a combined extractant solvent. Work is underway in the U.S. and Europe on developing several new options for combined processes (TRUSPEAK, ALSEP, SANEX, GANEX, ExAm are examples). There are unique challenges associated with the operation of such processes, some relating to organic phase chemistry, others arising from the variable composition of the aqueous medium. This project targets two general problematic issues in designing combined process systems: managing the chemistry of challenging aqueous species and optimizing the composition and properties of combined extractant organic phases. The primary focus areas of this research program are 1) developing improved information on the thermodynamics of problematic fission product zirconium and 2) developing a framework for an organic phase solvation model based on molecular-scale interactions between extractant molecules and organic solvent molecules. Both zirconium chemistry and "diluent effects" have been investigated in the prior literature. As each issue represents a particularly challenging obstacle to combined process development and the details of the science remain largely unresolved, this project aims to build on existing information with carefully designed and executed investigations of each subject and integration of the parallel thrusts where it is reasonable to do so. The research team includes Professor Nathalie Wall (PI), WSU who has recently reported results on the application of radiotracer-based solvent extraction studies of Tc(IV) complexes, Professor Kenneth L. Nash (WSU), Dr. Leigh R. Martin (Idaho National Lab), and Dr. Cécile Marie (CEA-Marcoule, France). The collaboration team brings expertise in actinide/lanthanide chemistry, separation science and the thermodynamics and kinetics of metal ligand interactions. Drs. Marie, Martin, and Nash have collaborated previously on similar investigations. The overarching objectives of this investigation are to improve the state of knowledge on zirconium chemistry in the nuclear fuel cycle, to add new analysis tools to support development of long term improvements in the predictability of fuel cycle separations systems, and ultimately to enable the creation of compact and efficient separations methods for advanced nuclear fuel reprocessing.