Overcoming Kinetic Barriers to Actinide Recovery in ALSEP

PI: Mark P. Jensen, Colorado School of Mines
Program: Fuel Cycle Technologies, FC-1.2b

Collaborators: Artem Gelis, Argonne National Lab
Shubham Vyas, Colorado School of Mines

ABSTRACT:

A key objective of future nuclear fuel cycles is to minimize the transuranic content of high level waste to increase the capacity of nuclear waste repositories. This requires the transuranic actinide elements americium and curium to be separated from fission product lanthanides, which is very difficult. New separations processes are being developed to perform this separation, but the chemical reactions that enable the separation of Am and Cm from the lanthanides are sometimes too slow to allow efficient, cost effective separations. Despite the importance of this problem to actually deploying a real separation system, work on such actinide/lanthanide separation systems has historically focused on improving the thermodynamic selectivity of the processes to the near exclusion of the equally essential kinetic considerations. The objectives of this proposal are to identify specific kinetic bottlenecks that slow down a new solvent extraction process for separating trivalent actinides from lanthanides, known as ALSEP, and to investigate a novel strategy for overcoming the kinetic barriers that are limiting its deployment.

Motivated by very recent results in systems closely related to the ALSEP separations process, our central hypothesis is that the undesirably slow kinetics encountered in aspects of the ALSEP process, such as slow Am stripping, originate in the poor ability of the aqueous complexants to penetrate and react in the interfacial zone containing the metal-extractant complexes. To test our hypothesis we will systematically study the rate of partitioning of selected trivalent actinide and lanthanide elements in the ALSEP processes at room temperature and at a process-relevant temperature to develop baseline mechanistic models for ALSEP that identify the likely kinetic bottlenecks. Interfacial tension measurements, advanced interfacially-sensitive spectroscopies, and molecular dynamics simulations also will be employed to understand the kinetics of lanthanide and actinide partitioning in these systems. Parallel to these baseline studies, we will study fundamental ideas for altering the reaction rate in the interfacial zone by systematically tuning the chemistry on both sides of the interface and testing aqueous ligands that should be better able to react at the interface.

The results of our experiments, the mechanistic models, and potential improvements to the current implementations of the ALSEP process represent critical input to ongoing efforts in existing DOE programs for materials recovery within the Fuel Cycle Technologies program. Identifying specific places where the actinide/lanthanide separations are kinetically limited will enable rational approaches to improve the chemistry of these separations currently done by trial and error. Significant improvements in the extraction kinetics can greatly simplify the complexity and cost of future plans for material recovery.