X-ray Studies of Interfacial Molecular Complexes in ALSEP Back-Extraction

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Program: FC-1.2: Materials Recovery

ABSTRACT:

Efficient separation of trivalent minor actinides (Am/Cm) from lanthanides in spent nuclear fuel is a primary objective for the optimization of nuclear fuel cycles. Processes based upon solvent extraction, such as ALSEP, address this challenge. Solvent extraction in nuclear fuel reprocessing relies upon the transport of metal ions across organic/aqueous (liquid/liquid) interfaces. Although the interaction of metal ions with aqueous complexants, buffers, and organic extractants at the organic/aqueous interface is likely to determine the efficiency and kinetics of extraction processes, little is known about the molecular complexes that form at the interface, or the mechanisms for ion transport across the interface. Optical spectroscopies and Lewis Cells have provided some equilibrium and kinetic information primarily from the bulk solution, but are limited in their ability to provide structural information near or at the interface between two immiscible solvents.

The objective of the proposed project is to apply state-of-the-art synchrotron X-ray techniques to achieve a fundamental understanding of the molecular-scale organization of bulk extractants and radiologically derived impurities, complexants, buffers, and metal ions at the organic/aqueous interface during ALSEP back-extraction. Synchrotron X-ray interface-sensitive techniques provide unique information that is unobtainable by other methods, and the knowledge obtained can be integrated with ongoing work in the development of the ALSEP process. This proposal focuses on ALSEP back-extraction because it is kinetically hindered, though the reasons for the slow kinetics are unknown and represent a critical knowledge gap in its application. Therefore we propose to map the unknown territory of the organic/aqueous interface in the presence of trivalent ions and resolve how those ions are complexed as they are transported across the organic/aqueous interface.

A new sample cell will be fabricated during the first year of the project that will be compatible with the safety requirements for X-ray studies that contain the trivalent actinide Am(III). This cell will be used to study the back-extraction of Am(III) separately and in combination with trivalent lanthanides Eu(III), La(III), and Nd(III), as well as separate studies of these lanthanides. Preparatory visual and interfacial tension measurements will precede the X-ray studies. Deliverables consist of scientific manuscripts describing the results of these studies, as well as reports and conference presentations. These investigations of interfacial molecular complexes formed during the process of back-extraction aim to fulfill the need for a fundamental understanding of the stripping of metals, including the role of complexants, buffers, and radiolytic degradation impurities in the transport of metal ions across the organic-aqueous interface in ALSEP, as requested by the NEUP workscope FC-1.2. The understanding gained during the project can lead to the development of more efficient and faster metal stripping that is relevant to the separation of actinides from lanthanides in the nuclear fuel cycle.