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Microemulsions and Aggregation Formation in Extraction Processes for Used Nuclear Fuel: Thermodynamic and Structural Studies

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ABSTRACT

Advanced nuclear fuel cycles rely heavily on the successful chemical separation of various elements in used fuels. Numerous solvent extraction (SX) processes have been developed for the recovery and purification of metal ions from this material. However, the predictability of process operations has been challenged by the lack of a fundamental understanding of the chemical interactions in several of these separation systems. One example is the process for the separation of trivalent actinide—An(III)—from trivalent lanthanide—Ln(III)—elements, where gaps in the thermodynamic description of the mechanism make predictions very challenging. Recent studies of this system, in particular, as well as past studies of SX, in general, have suggested that aggregate formation, most notably reverse micellization, in the organic phase results in a transformation of its selectivity and efficiency. Aggregation phenomena have consistently interfered in SX process development, and, over the years, have become synonymous with an undesirable effect that must be prevented. Here we propose a 4-year research effort to derive the fundamental principles of solvation and self-organization in non-aqueous solutions as the experimental conditions of the organic solution become increasingly disposed towards aggregation. The proposed work will blend the thermodynamic, structural, and theoretical investigations of evolving solute architectures to enhance our understanding of the supramolecular chemistry governing aggregate formation, especially in challenging separation systems such as An(III)/Ln(III) partitioning. The relationships to be established between the structures and the energetics of the molecular interactions will outline new theoretical descriptions of metal ion transport phenomena between aqueous environments and highly organized non-aqueous media.

Specific objectives of the proposed research are as follows:

1. Investigate the important trends and relationships between the formation of water-in-oil microemulsions and the mechanisms of metal ion phase transfer.
2. Use 2-phase calorimetry to investigate thermodynamic signatures of progressive structural organization of the organic phase leading to third phase formation.
3. Use synchrotron radiation techniques, such as small angle X-ray scattering and X-ray absorption spectroscopy, to characterize and gain an improved understanding of the structure of reverse micelles, how and why they form and their role in metal extraction.
4. Compile the accumulated systems knowledge, perspectives and insights obtained from the coordinated experimental and theoretical efforts in a database to allow us to describe, understand and predict the mechanisms and formation of reverse micelle effects and stable water-in-oil microemulsions in the extraction of metal ions.