Organic Speciation and Interactions in ALSEP – One Step Partitioning Process of Minor Actinides, Lanthanides and Fission Products

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ABSTRACT:

The objective of the proposed research is to quantify the interactions of the extractant ligands in the Actinide-Lanthanide Separation (ALSEP) process organic phase. ALSEP proposes a trivalent minor actinide/lanthanide group extraction followed by a selective actinide strip to effectively partition minor actinides, lanthanides, and other fission products. This process utilizes HEH[EHP], an organophosphoric acidic ligand and either TODGA or TEHDGA, neutral ligands with diglycolamide functional groups collectively referred to as DGAs. Recently collected IR-spectroscopic data suggest that ALSEP ligands form inter-molecular adducts at a significantly lower equilibrium concentrations than those proposed for the TRUSPEAK process, HDEHP and CMPO. Quantifying these effects can be used to determine how an engineering scale process will perform and generate an accurate chemical model for the system.

The proposed work will investigate the process organic phase prior to aqueous phase separation and after separating aqueous phases of bare and metal-loaded acids. First, the polymeric tendencies of the pre-extraction organic phase will be determined using vapor pressure osmometry to determine the presence of any potentially competing adduct formations. Next, ^{31}P NMR will be used to accurately quantify the adduct formation constants in the organic phase at multiple temperatures typical of engineering processing operations. These investigations will be conducted for ligand combinations typical of ALSEP and TRUSPEAK for comparison. Aqueous phase extraction effects on ligand interactions will next be studied for both pure acid extraction and metal loaded acids. SAXS and EXAFS will be used to determine the differences in mixed extractant systems as opposed to pure DGA systems in terms of aggregate sizes and adduct contributions to extraction. Furthermore, comparisons of the amount of acid or metal extracted to the organic phase will be determined to determine how these behaviors affect an overall process. For metal extraction, slope analysis with varying extractant concentrations will also be performed to determine possible speciation changes.

This project will serve to generate a more accurate predictive chemical model for the operations of the ALSEP process. A full chemical model of this type is essential for operations of a large, complex chemical process to determine performance based on inputs and outputs to the system, and to correct process non-idealities quickly so as to preserve process integrity. This work is thus directly supportive of current DOE efforts of scaling a process for the efficient separation of minor actinides and lanthanides.