High Concentration Monoamide Separations: Phase Modifiers and Transuranic Chemistry

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ABSTRACT:

Recycle of desirable elements from used nuclear fuel at the industrial scale and the lowest cost for advanced fuel cycles requires new single cycle aqueous separations that can handle a variety of feeds and products. For example, HALEU recycle, MOX production, and proliferation resistant group actinide separations are currently at a low TRL in the United States. Solvent extraction systems built on monoamide extractants have demonstrated the potential to meet these diverse needs and goals, but these separations would be much more efficient if they could be intensified by using increased extractant concentration over what has previously been considered. However, the chemical properties and behavior of such high concentration monoamide (HiMA) systems are not known with any certainty. High extractant concentrations can enable additional efficiency by allowing higher metal loading of the organic phases, but we believe that this higher metal loading will come with a price. First, the higher concentrations of polar species in the organic phase will change solution properties such as viscosity, solubility of the extracted complexes, and phase disengagement. Second, it can encourage the extraction of otherwise undesirable species by shifting the extraction equilibria or favoring the formation of new extractable species. Third, HiMA systems could alter the redox properties of the solvent, which may require different approaches to control the oxidation states of the elements. This proposal seeks fundamental understanding of the features of HiMA extraction systems in these three areas to define the operating space and develop solutions to these limitations on this promising single cycle separation system.

The project will focus on the scientific understanding necessary to further develop HiMA separations to enable UNF recycling. Separations relevant to HALEU recovery (U only recovery), "standard" UNF processing (i.e. just U & Pu recovery) as well as GANEX-type separations (i.e. U, Np, Pu, Am) will be considered. Three primary tasks will elucidate essential behaviors of HiMA separations relevant to each of these classes of separations: (1) evaluation of modifier-containing HiMA systems and the effect of modifiers on essential solution properties such as viscosity, (2) evaluation of Np oxidation state speciation and chemical speciation in HiMA systems, and (3) evaluation of HiMA systems for proliferation resistant group actinide separations in a hexavalent GANEX (U-Am) framework. The chemistry of the HiMA systems will be explored using an array of experimental and computational approaches at the Colorado School of Mines and Pacific Northwest National Lab, including radiotracer measurements; UV-visible-NIR, vibrational, NMR, and X-ray spectroscopies; electrochemistry; and computational modeling to understand the systems. The project will fill critical knowledge gaps in the application of intensified monoamide extraction systems that will provide an opportunity to stabilize the fuel cycle resources necessary for advanced reactors and our current reactor fleet.