Group Actinide Separation by Crystallization: A Single-Technology Approach to Used Nuclear Fuel Recycle

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ABSTRACT:

Bulk removal of U, Np, Pu and Am, from used nuclear fuel (UNF) would be advantageous for future nuclear fuel recycle. A co-crystallization of the hexavalent actinides (Ans) with U(VI) has been demonstrated and a process based on this would have many benefits. By a simple adjustment of temperature, the solubility of UO_2^{2+} can be significantly altered, resulting in a uranium nitrate hexahydrate (UNH) precipitate. Other An(VI) species should behave in a similar fashion to that of U(VI), and a scheme to co-crystallize Np(VI), Pu(VI), and Am(VI) should be possible. This new approach involving crystallization would not only support a GANEX type paradigm, but it would also provide a technique that does not involve organic compounds and could affect the separation with only a thermal swing.

While the previous proof-of-principle separations are promising, there are numerous gaps in our fundamental understanding of the mechanism of removal of the An(VI) species, selectivity, kinetics, yield, and stage-wise strategy. This proposal will be focused on filling in these holes with a science-based approach to developing this new method of group Ans separations. To accomplish this goal, we will pursue three Specific Aims: (1) Develop thermodynamically based dissolution-recrystallization cycles for producing purified Ans streams at high yield and good kinetics, (2) Study the effects of different oxidation methods on the formation of the crystalline phase. (3) Develop an understanding of the molecular structure and bonding of the crystalline phase containing the An(VI) species.

We will deliver (1) a process method of utilizing dissolution-recrystallization cycle for increasing purity and kinetics; (2) a description of the compatibility of different oxidation methods with the crystallization process; (3) a set of UNH samples containing either Np(VI), Pu(VI), or Am(VI), as well as samples containing a combination of all three for XAS; (4) structural information of bonding environment of the An(VI) metal centers by XAFS; and (5) several students will be well versed in radiochemistry and An chemistry.

This new UNH crystallization based GANEX type separation approach, provides an elegant solution to the UNF recycling, which is inherently free of organic solvent and added complexants. We believe the development of this approach has the potential to open the door to a new paradigm of nuclear fuel recycling with a single, mature technology, crystallization. Moreover, this research will significantly impact the greater scientific community by expanding the basic understanding of Am(VI) chemistry through the structure and bonding data obtained through XAS. Finally, another one of the major benefits of this proposal is that a graduate student will be involved in carrying out these studies. They will be trained in An chemistry and radioisotope experimental chemistry to assure trained personnel for the future.