Controlling Hexavalent Americium – A Centerpiece to a Compact Nuclear Fuel Cycle

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ABSTRACT:

Closing the nuclear fuel cycle could be simplified by recovering the actinides U through Am as a group. This could be achieved by converting U, Np, Pu and Am to the hexavalent state. Uranium, Np and Pu are readily oxidized to the hexavalent state. Generation of hexavalent Am in acidic solutions is more difficult, as the standard reduction potential of the Am(VI) /Am(III) couple (+1.68 V in 1 M HClO_4) is well outside of the electrochemical stability window of water. While the oxidation and separation of Am has been demonstrated under laboratory conditions, several issues could plague scale up and implementation of this separation with used fuel. Two primary concerns are considered. The first issue concerns the stability of the oxidized Am. The second involves the undesirable co-extraction of tetravalent f-elements with the hexavalent actinides. To address the first concern regarding Am redox instability, Am reduction will be monitored under a variety of different conditions to establish the means of improving the stability of Am(VI) in the organic phase. Identifying the components contributing most significantly to its reduction will allow thoughtful modification of the process. To address the second concern, we propose to apply branched chain extractants to separate hexavalent actinides from tetravalent f-elements. Both branched monoamide and organophosphorus extractants have demonstrated significant selectivity for UO_2^{2+} versus Th^{4+}, with separation factors generally on the order of 100. The efforts of this two-pronged research program should represent a significant step forward in the development of aqueous separations approaches designed to recover the U-Am actinides based on the availability of the hexavalent oxidation state. For the purposes of this proposal, separations based on this approach will be called SAn(VI) separations, indicating the Separation of An(VI).