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Americium Separation by Electrochemical Oxidation for Nuclear Fuel Cycle Applications

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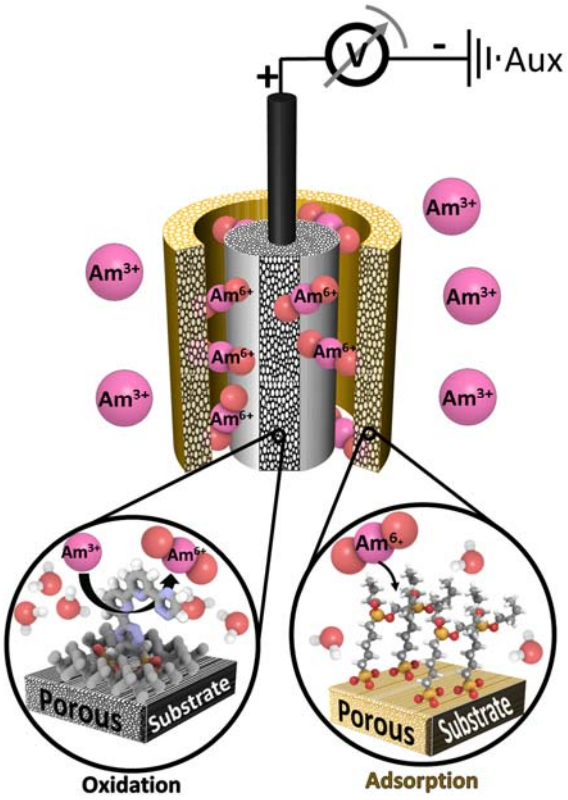
ABSTRACT:

Separation of americium from nuclear waste streams, including other minor actinides (MA) and the lanthanides, continues to place a significant technical burden on the nuclear fuel cycle. Separation of Am(III) from the lanthanides and MA in the common trivalent oxidation state in acidic solutions by conventional extraction techniques presents a significant challenge given their similar atomic radii. In the research proposed here a separation scheme will be explored, involving oxidation of Am(III) to Am(VI) followed by adsorption of Am(VI) onto a solid support. It will build on our initial results on the electrochemical oxidation of Am(III) to Am(VI) at high surface area, derivatized oxide electrodes with selective separation and concentration of Am(VI) as AmO_2^{2+} from the nuclear waste stream at a second, solid support.



Electrochemical oxidation at high surface area oxide electrodes will be facilitated by derivatization of the electrodes with surface-bound ligands to concentrate Am(III) ions and redox mediators to facilitate interfacial electron transfer. Existing electrodes based on high surface area fluoride-doped tin oxide (FTO) and oxide-coated reticulated vitreous carbon (RVC) will be investigated with extensions to ultra-high surface area porous metal oxide aerogels which are ideally suited for flow applications. Surface derivatization of the electrodes, will be accomplished by using ligands known for Am(III) binding, synthesized with phosphonic acid derivatives as anchoring groups for surface attachment. Stabilization of porous materials and of ligand surface binding will exploit well-developed atomic layer deposition (ALD) techniques or by electrochemical deposition of diamond like carbon (DLC). Liquid-solid extraction systems will be developed to remove Am(VI). These will include the development of sorbent materials surface functionalized or pore impregnated with known ligands and materials designed for Am(VI) liquid-liquid separation or uranyl separation. The procedures described here have the potential to impact the nuclear fuel cycle directly. They will also provide procedures for preparing high oxidation state Am in the absence of oxidizing agents enabling fundamental studies of intrinsic value, and advances in its coordination chemistry.