Mixed Ligand Metal Phosphonate-Phosphate Hybrid Ion Exchangers for the Separation of Lanthanides from Actinides and Americium from Curium

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ABSTRACT:

It is possible to recover 95% of the original fuel value from spent nuclear fuel rods. During fission of U-235 in generating electricity, a group of elements, termed actinides are produced along with another group of elements known as lanthanides. While the actinides, except for curium can be used as fuel, the lanthanides cannot. They absorb neutrons and thereby reduce the amount of electricity generated. Therefore, these elements along with a few others must be removed and treated as waste.

Our objectives are to cleanly separate the lanthanides and curium from the reusable actinides. Our ability to do these separations stems from our discovery of a remarkable group of ion exchangers that readily absorb elements (as ions) with high charge of 3+ or 4+ but sorb very little of those with lower charge (1+, 2+). Both the actinides and lanthanides exist as 3+ ions but the actinides can be oxidized to AnO2+ with charge of 1+. The lanthanides and also curium under these conditions remain in the 3+ state and can be separated from the actinides by ion exchange sorption.

The ion exchangers to be used for this project are metal-organic phosphonate-phosphates of general composition $M(O_3PC_6H_4PO_3)_{1-x/2}(HPO_4)x\bullet nH_2O$, M= Zr_4+, Sn_4+.They are three dimensional porous solids that are stable in air to 450 ºC. By comparison with similar materials that are stable to ionizing radiation, we believe that these ion exchangers will also be resistant to such radiation.

A portion of our studies will be directed towards determination of the structure of the ion exchangers. Due to their poor crystallinity, a variety of procedures such as Extended X-ray Absorbance Fine Structure (EXAFS), electron diffraction (ED), and atomic pair distribution function (PDF) will be obtained at Brookhaven and Argonne National Laboratories to retrieve useful structural data. This information will be utilized to uncover the reasons for their unusual sorption behavior and to develop useful materials for difficult separation procedures.

The bulk of the funding will be utilized for two graduate students and a post-doctoral research fellow at Texas A&M, for salary, travel expenses and living costs for when they are at the collaborating laboratories. Personnel at Texas A&M University will partner with Dr. David Hobbs at SRNL and Dr. Donald Reed at Los Alamos Carlsbad who will utilize their expertise and laboratories to handle the highly radioactive materials necessary to the project. The actinide portion of the research will be completed by Texas A&M students at the two national laboratories because they have the personnel and the facilities to handle the radioactive material. Finally, chromatographic procedures will be formulated by Dr. Linda Wang of Purdue University to effect the final steps in the process. Funds will also be allocated to SRNL and Los Alamos to cover the cost of radioisotopes and analytical procedures. These three colleagues will be collaborators with a monetary contribution of $65,000 total over the proposal lifetime.

The impact of this study is the development of an inexpensive, simple way in which to separate the lanthanides from the actinides found in the spent nuclear fuel rods and to recover a large portion of the usable fuel. This project will determine the feasibility of ion exchangers to effect complete separation of lanthanides from actinides without radiolytic degradation. In addition we will utilize the structural data to design new sorbents for difficult separations and test their recyclability. In the process, several students will receive training to handle radioisotopes and knowledge of lanthanide and actinide chemistry required for the future of nuclear energy.