Actinide-Specific Interfacial Chemistry of Monolayer Coated Mesoporous Ceramics

Project ID: 65370

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Research Objective

The objective of this program is to design, synthesize, and evaluate high-efficiency, high-capacity sorbent materials capable of selectively sequestering actinides from complex aqueous mixtures. One of the central goals of this project is to understand the fundamental interfacial science required to develop novel mesoporous materials coated with organized monolayers of rationally designed ligands, custom-tailored for binding specific actinide cations. This capability addresses waste management by separation of actinides, a central concern of high-level waste (HLW) management at several DOE sites.

Research Progress and Implications

This report summarizes work after 1.5 years of a 3-year project. Selective isolation of actinides from a complex mixture is anticipated to increase assay accuracy, speed up assay turnaround, reduce waste volume (and costs), and enhance decontamination efficiency and safety. In Year 1, we developed powerful new SCF synthesis methods that significantly enhance our capabilities in the production of SAMMS; we developed a series of first generation sorbent materials and evaluated their capabilities using lanthanide models; and we developed a novel anion sequestration strategy. In Year 2, we have extended our SCF methods to include SC nitrogen (which has certain advantages over other SCFs); we have refined our interfacial ligand field to exploit chelators with extremely high affinity for the actinides; we have determined binding kinetics (3-5 minutes) using lanthanide models; we have started with competition experiments to evaluate competition by common competing metal cations; started to acquire actinide binding data; obtained detailed characterization of the binding of tetrahedral oxoanions to our anion SAMMS and applied these materials to the sequestration of pertechnetate.

Planned Activities

In Year 3, we intend to extend our actinide studies to include all of the actinides from Th through Am; we will evaluate the HOPO ligands in a SAMMS matrix; look at actinide competition studies, kinetics, waste simulants and ultimately possibly even actual waste samples (from Tank Farms, PFP or other Hanford facilities), and perform EXAFS characterization of the pertechnetate/SAMMS adduct.

Information Access

List publications and any other access to results (e.g., a Web page). http://www.pnl.gov/etd/product/samms/index.htm
http://www.acs.org/government/success/index.html

"Environmental Applications of Self-Assembled Monolayers on Mesoporous Supports (SAMMS)", G. E. Fryxell,* J. Liu and S. Mattigod, *Materials Technology*, **1999**, *14*, 188-191.

"Design and Synthesis of Selective Mesoporous Anion Traps" Glen E. Fryxell, Jun Liu,* Meiling Gong, Teresa A. Hauser, Zimin Nie, Richard T. Hallen, Mauxu Qian, and Kim F. Ferris, *Chemistry of Materials*, **1999**, *11*, 2148-2154.

"Environmental Applications of Interfacially Modified Mesoporous Ceramics" G. E. Fryxell *, J. Liu, S.V. Mattigod, L.Q. Wang, M. Gong, T.A. Hauser, Yuehe Lin and K. F. Ferris, and X. Feng; to be published in *Ceramics Transactions*, Volume 94, *Environmental Issues and Waste Management Technologies in the Ceramic and Nuclear Industries,* (in press).

"Synthesis and Applications of Functionalized Nanoporous Materials for Specific Adsorption" J. Liu,* G. E. Fryxell, S. Mattigod, T. S. Zemanian, Y. Shin, L. Q. Wang, to be published in "Access in Nanoporous Materials" Vol. 2 (in press).

"Designing Surface Chemistry in Mesoporous Silica" G. E. Fryxell* and Jun Liu an invited contribution to "Adsorption at Silica Surfaces" edited by Eugene Papirer, Marcel Dekker, (in press).

"Self-Assembled Monolayers on Mesoporous Supports for Metal Separation" S. Mattigod, G. E. Fryxell, X. Feng and J. Liu* in " Metal Separation Technologies Beyond 2000: Integrating Novel Chemistry with Processing", edited by K. C. Liddell and D. J. Chaiko, published by The Minerals, Metals and Materials Society, pp. 71-79, **1999.**

"Self-Assembled Monolayers on Mesoporous Silica, a Super Sponge for Actinides" X. Feng,* L. Rao, T. R. Mohs, J. Xu, Y. Xia, G. E. Fryxell, J. Liu, and K. N. Raymond, *Ceramic Transactions*, Volume 93, *Environmental Issues and Waste Management Technologies in the Ceramic and Nuclear Industries* (IV), edited by J. C. Marra and G. T. Chandler, **1999**, pp. 35-42.