Development of Biodegradable Isosaccharinate-Containing Foams for Decontamination of Actinides: Thermodynamic and Kinetic Reactions between Isosaccharinate and Actinides on Metal and Concrete Surfaces

(Project Number: 82715)

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Research Objective

Actinide contamination of steel and concrete surfaces is a major problem within the U.S. Department of Energy (DOE) complex. For steel surfaces, the primary problem is contamination of sections of nuclear power reactors, weapons production facilities, laboratories, and waste tanks. For concrete, there are an estimated 18,000 acres of concrete contaminated with radioactive materials that need decontamination. Significant efforts have gone into developing decontamination technologies. Almost all current decontamination technologies rely on removal of the contaminated surface layer by mechanical means or by chemical methods using harsh chemicals. Some of the technologies are ineffective. Others are expensive, labor-intensive, and hazardous to workers. Still others create secondary mixed wastes that are not environmentally acceptable.

This project seeks fundamental information that will lead to the development of a new and more environmentally acceptable technology for decontamination of Pu on steel and concrete surfaces. The key component of this technology is isosaccharinate (ISA), a degradation product of cellulose materials that is biodegradable and binds strongly with Pu. Isosaccharinate will be incorporated into foams for use in decontamination of Pu from steel and concrete surfaces. To develop a fundamental basis for this proposed technology, we will 1) study the effect of pH and common ions (Na and Ca) on the speciation and thermodynamic reactions of ISA over wide ranges of pH and concentrations of Na and Ca, 2) develop thermodynamic and kinetic data for ISA reactions with Pu(IV) and Fe(III), and 3) determine the fundamental Pu concentration-controlling reactions involving steel and concrete surfaces and test contaminated surfaces using ISA-containing foams for Pu removal. Our ultimate goal is to develop a technology, based on sound fundamental principles, for decontaminating tetravalent actinides. The proposed research is a multi-laboratory effort that includes fundamental chemistry studies conducted at Pacific Northwest National Laboratory and Lawrence Berkeley National Laboratory, and foam formulation and decontamination research conducted at Sandia National Laboratories.

Research Progress and Implications

This report summarizes the results of the first 6 months of a 3-year project. Progress is described in the following paragraphs.

Isosaccharinate is not commercially available. Therefore, methods were perfected to produce large quantities of solid calcium isosaccharinate. Methods were also developed to change this calcium isosaccharinate to more soluble sodium isosaccharinate for use in studies involving development of equilibrium and kinetic data for important ISA reactions.

Preliminary tests to determine the possible efficacy of isosaccharinate to decontaminate steel and concrete surfaces containing uranium showed that isosaccharinate indeed holds promise as a decontaminating agent.

Experiments were planned to develop extensive thermodynamic data for complexation constants of isosaccharinate with a number of metals including actinides (trivalent, tetravalent, pentavalent and hexavalent) and matrix elements (e.g., Fe(II), Fe(III), and Ca) of contaminated surfaces.

Preliminary ^{13}C- nuclear magnetic resonance studies show that deprotonation of carboxylic acid groups is complete at pH 5 and that no further deprotonation reactions of ISA are apparent in the pH range from 5 to 12, which is contrary to that suggested in the literature.

Studies on the solubility of $NpO_2(am)$ as a function of pH at fixed ISA concentrations, and as a function of ISA and at fixed pH, showed that ISA strongly complexes Np(IV) in a large range of pH values, extending to as high as pH 12. These studies show that similar behavior can be expected with Pu(IV) systems and that ISA holds promise as an effective decontaminating agent for Pu.

Planned Activities

We will develop fundamental data over a wide range in pH values on the types of aqueous complexes and complexation constants of ISA with actinides and with metals such as Fe and Ca that compete with actinides for ISA complexation. Both the equilibrium and kinetic data for these important reactions will be developed.

We will formulate foams containing ISA, then test ISA solutions and ISA-containing foams for their effectiveness to decontaminate actinides from metal surfaces and concrete.