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Chemical Speciation of Strontium, Americium, and Curium in High Level Waste: Predictive Modeling of Phase Partitioning During Tank Processing

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Research Objective

In this research program, Pacific Northwest National Laboratory (PNNL) and Florida State University (FSU) are investigating the speciation of Sr and Am/Cm in the presence of selected organic chelating agents (ethylenediaminetetraacetic acid (EDTA), N-(2-hydroxyethyl)ethylenediaminetriacetic acid (HEDTA), nitrilotriacetic acid (NTA), and iminodiacetic acid (IDA)) over ranges of hydroxide, carbonate, ionic strength, and competing metal ion concentrations present in high level waste tanks. The fundamental understanding of chemical speciation reactions gained from these studies is also used to propose methodologies for removal of Sr and Am/Cm from organic chelates present in high level tank waste, via competition, displacement or other reactions, without the need for the development of costly and potentially hazardous organic destruction technologies.

Research Progress and Implications

As of May 1998, these studies show that given the expected range in chelate concentrations present in tank waste and the fact that carbonate is present in significant concentration in all tanks, it is likely that the chemical speciation of Sr will be almost certainly unaffected by the presence of IDA and probably also unaffected by the NTA as well. Only HEDTA and EDTA appear to be present at high enough concentration and form strong enough complexes to significantly impact the speciation of Sr in tank waste. In addition, competition with other metal ions present in such solutions, in this case Ca, can effectively displace Sr from the strong chelating agents HEDTA and EDTA, depending upon the concentration of Sr and the competing metal ions. This result indicates that metal ion displacement may represent an acceptable alternative to costly and hazardous organic destruction technologies in reducing the impacts of organic chelates in tank processing, especially given the fact that the chelators represent only a small fraction of the total organic carbon in tank waste. Predictive thermodynamic relationships have been developed for Sr in the presence of mixed hydroxide-carbonate systems and in the presence of competing metals (Ca^{2+}), along with a detailed mechanistic understanding of the important speciation reactions in these system

Considerable progress has also been made in the study of the effects of hydrolysis and carbonate complexation on the displacement of trivalent actinides and actinide analogs from the organic chelates: EDTA, HEDTA, NTA, and IDA. Studies on the solubility of Eu (a trivalent actinide analog) compounds [i.e. $Eu(OH)_3(c)$] have been completed as a function of base concentration in the presence of four organic chelates: EDTA, HEDTA, NTA, and IDA. These studies have shown that high base concentration can displace Eu(III) from all of the organic chelates studied. The effective NaOH concentration for the displacement reaction being dependent on the nature of the specific chelate studied and the chelate concentration. Such data are being used to develop accurate thermodynamic models for trivalent actinide species under high base conditions.

Planned Activities

Plans for FY99 call for completing the temperature dependent thermodynamic model for Sr in the mixed chelate systems, completing the studies that have been initiated on the $EuPO_4(c)$, publishing the results on the $Eu(OH)_3(c)$ in mixed chelate systems, and the studies of solution phase calorimetry. Plans for FY99 also call for extending the CE/MS studies to high ionic strength solutions. Support for studies in outer years will be required to: 1) extend the trivalent actinide model to higher

temperatures (>250° C), 2) unravel the speciation of tetravalent actinides (especially Pu(IV)), and 3) verify the modeling predictions and speciation measuremets (CE/MS) in tank waste, specifically complex concentrates from the Hanford site.

Other Access To Information

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