Characterization of Actinides during HEDPA Leaching for Aluminum Dissolution in Tank Waste Sludges

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The nuclear waste sludges in underground tanks at Hanford contain most of the actinides as well as non-radioactive materials. Among these materials, aluminum oxide is particularly problematic to the vitrification of high-level waste sludges because it is present in large amounts and it is not as easily removed from the sludge by baseline leaching processes as expected. Besides, interactions of actinides with aluminum oxide have significant implications in designing the strategies for sludge leaching. The objective of this study is to examine the behavior of actinides in the leaching of aluminum oxide with 1-hydroxyethane-1,1-diphosphonic acid (HEDPA), a complexant that forms strong complexes with actinides and aluminum in acidic to basic solutions.

Batch adsorption/dissolution experiments were conducted to examine the interactions between lanthanides/actinides and the aluminum oxyhydroxide boehmite (γ-AlOOH) in 1.0M NaCl suspensions containing HEDPA. In the pH range 4 to 9, complexation of Al by HEDPA significantly enhanced dissolution of boehmite. This phenomenon was especially pronounced in the neutral pH region where the solubility of aluminum is limited by the formation of sparsely soluble aluminum hydroxides. At high pH levels, dissolution of boehmite is inhibited by HEDPA. Systems without HEPDA present exhibited higher concentrations of aluminum in the aqueous phase than those with HEPDA. Both enhancement and inhibition of boehmite dissolution are assumed to be due to differing modes of coordination between HEDPA and the boehmite surface. Data are discussed in terms of the complexation of aluminum and lanthanides/actinides with HEDPA and the effect of complexation on leaching of the lanthanides/actinides from the boehmite surface. The results of the leaching of lanthanides/actinides from aluminum (oxyhydr)oxides by HEDPA (and simultaneous dissolution of the aluminum solid) are of significant importance to the development of pretreatment processes for nuclear waste sludges.

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