Actinide sorption to aluminum (hydr)oxides: Influence of sorption site acidity

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The presence of the actinide elements in the geosphere poses a long-term environmental concern due to the toxicity and long half-lives of several isotopes. Therefore, understanding the mechanisms responsible for enhancing or retarding the mobility of the actinides in the environment is important for risk management. The environmental fate and transport of the actinides is particularly influenced by interfacial reactions such as adsorption and reduction.

The acid-base properties and hydration/dehydration mechanisms of corundum (α-Al_2O_3), γ-alumina (γ-Al_2O_3), and gibbsite (γ-$Al(OH)_3$) were previously studied [1]. The results showed that surface acidity follows the trend corundum > gibbsite > γ-alumina. This is presumably related to a difference in surface sorption sites $\equiv$AlOH, $\equiv Al_2OH$, and $\equiv Al(OH)_2$, where the greater proportion of aluminum at the surface leads to higher surface acidity. Therefore, metals that act as Lewis acids should preferentially sorb to γ-alumina. However, the relationship between sorption affinity and surface acidity has not been explicity established. The objective of this work was to examine actinide interactions with aluminum (hydr)oxide minerals with the intent of being able to characterize binary actinide-mineral systems based on fundmental thermodynamics.

Batch sorption experiments were used to monitor the amount of Eu(III), Th(IV), and Np(V) sorbed to corundum, gibbsite, and γ-alumina as a function of time, pH, and ionic strength. In addition, high-resolution transmission electron microscopy (HRTEM) imaging was used to identify possible surface precipitates and monitor for morphological changes in the solid phases. Batch sorption data and HRTEM observations were used to develop preliminary surface-complexation models describing actinide sorption to aluminum (hydr)oxide mineral phases.

This research supports the eventual investigation of plutonium sorption to aluminum (hydr)oxide minerals where, based on previoius research [2-3], surface-mediated reduction and strong inner-sphere surface complexation is expected.

[1] Yang *et al* (2007) *J. Colloid Interface Sci.* **308**, 395-404 [2] Hixon *et al* (2013) *J. Colloid Interface Sci.* **403**, 105-112 [3] Hixon *et al* (2010) *Radiochim. Acta* **98**, 685-692