

Nucleation behavior of U(VI) phosphate in the presence of mineral surfaces

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The addition of phosphate to U(VI)-contaminated shallow subsurface environments has been tested as a method of uranium immobilization primarily through the precipitation of sparingly soluble uranyl phosphate minerals. In the presence of phosphate and sediments, multiple mechanisms can control uranium mobility including adsorption of U(VI) onto mineral surfaces by ternary complexation¹ and surface nucleation of uranyl phosphate precipitates.² In this study, the transition between U(VI) phosphate adsorption and precipitation was investigated in the presence of various mineral surfaces, including goethite, montmorillonite, and sediments from Hanford, WA. U(VI) sorption isotherms on goethite and montmorillonite at pH 8 show a nucleation barrier to precipitation at elevated phosphate concentrations (100 or 1000 $\mu\text{M PO}_4^{3-}$). Solid-phase analysis by extended X-ray absorption fine structure spectroscopy (EXAFS) was used to characterize the structure of U(VI). Changes in the coordination number of the equatorial oxygen shell (U-O_{eq}) were observed with increasing uranium concentration prior to uranyl phosphate precipitation. Similar behavior was observed with both goethite and montmorillonite. No indication of a unique pre-nucleation cluster was found in the transition between U(VI) adsorption and precipitation by EXAFS or by laser induced fluorescence spectroscopy. Precipitate formation at the mineral surface was also examined using transmission electron microscopy. Results of this work will help to inform the design of phosphate-based uranium remediation strategies for soil and shallow groundwater systems.

[1] Singh A., et al. (2012) *Environ. Sci. Technol.* **46**, 6594–6603. [2] Singh A., et al. (2010) *Geochim. Cosmochim. Acta* **74**, 6324–6343.