

# Characterization of U(VI) Sorption-Desorption Processes and Model Upscaling

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## Introduction and Project Objectives

In order to apply predictive reactive transport models to the Hanford site, detailed knowledge of the speciation of contaminants is required. Important speciation parameters include: (1) oxidation state; (2) the local molecular structure surrounding contaminant elements; (3) the type and binding of a contaminant ion sorption complex (if adsorbed); (4) the type(s) of phase within which a contaminant is structurally incorporated [e.g., present in a three-dimensional precipitate(s)]; (5) the phase associations of a contaminant; (6) the microscopic distribution of a contaminant within sediments and soils. In the present study, we have used synchrotron-based X-ray spectroscopic methods to study the speciation of U and Cu in contaminated soil and sediment samples from the Hanford Site. To complement and complete our initial XAFS investigation of U speciation in contaminated vadose zone sediments below tank BX-102, we have also performed mXRD studies of two sediment sample to identify the specific U(VI)-silicate phase present. Samples from the 300 Area were examined by mSXRF to determine the microscopic distribution and element associations of Cu and U. These samples were also analyzed by U LIII- and Cu K-edge XAFS spectroscopy to determine the chemical speciation of these elements. Conclusions to Date (1) Uranium occurs primarily as sodium-boltwoodite  $[\text{Na}_2(\text{UO}_2)_2(\text{SiO}_3\text{OH})_2 \cdot 3\text{H}_2\text{O}]$  in the BX-102 sediment samples. (2) The dissolution kinetics of sodium-boltwoodite will be a major control on the future transport of U beneath tank BX-102. (3) In the 300 Area soils and sediments, uranium occurs as U(VI) and copper as Cu(II). (4) U and Cu are often found together or adjacent to one another; however, these elements don't appear to be spatially associated with Fe. (5) U appears to be bound to carbonate groups, and is likely contained in a  $\text{CaCO}_3$  mineral (calcite or aragonite). (6) It is unclear what phase(s) Cu is bound to or associate with, although the data are consistent with an association with  $\text{CaCO}_3$  minerals. (7) Future U release in the 300 Area will be controlled by the dissolution of  $\text{CaCO}_3$  minerals.