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Speciation of reduced U(IV) in contaminated sediments and laboratory reactors

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In situ groundwater remediation or waste storage strategies often have the goal of limiting the mobility of a contaminant by decreasing its solubility in the passing aqueous phase. The chemical or enzymatic reduction of U(VI) to U(IV) presents an approach to achieve this goal by exploiting the lower solubility of uraninite (UO2) relative to that of U(VI) species. This property of U has led to extensive research aimed at understanding the mechanism of U(VI) reduction and the stability of uraninite. However, recent evidence from U LIII-edge x-ray absorption spectroscopy suggests that uraninite may not be the predominant form of U(IV) in reduced sediments on the time scale of remediation activities. We will present an example from contaminated sediments at the Oak Ridge Integrated Field Research Center,1 followed up by several laboratory studies where we find that factors such as the mechanism of enzymatic U(VI) reduction,2 groundwater components such as phosphate or carbonate,2 and the ratio of U to specific mineral surface sites,3 can prevent the precipitation of uraninite and result in the formation of complexed or adsorbed U(IV) species. The spectroscopic features allowing the identification of these species from XANES and EXAFS measurements will be highlighted. The predominance of non-uraninite U(IV) species in reduced sediments presents new analytical and transport modelling challenges that will need to be addressed in future research.

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