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Products of abiotic U(VI) reduction by biogenic Fe(II)-bearing minerals

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Uranium is a subsurface pollutant of concern in the US, former East Germany and many other countries worldwide. In the US, the inventory includes an estimated 475 billion gallons of contaminated ground water and 75 million cubic meters of contaminated sediments. In former Eastern Germany, an estimated 100 million tons of uranium-mined material was reportedly disposed improperly. The need for site remediation is urgent and the scale of the problem, enormous.

Reductive immobilization of hexavalent uranium [U(VI)] to uraninite (UO_2) by stimulation of indigenous dissimilatory metal-reducing bacteria (DMRB) has been extensively investigated as a remediation strategy for subsurface U(VI) contamination. These bacteria gain energy by reducing oxidized metals as terminal electron acceptors, often using organic electron donors. Thus, when evaluating the potential for in situ uranium remediation, it is important to understand how the presence of competitive electron acceptors and corresponding biogenic products affect U(VI) remediation and the long term reactivity of reduced uranium. Iron, an abundant metal in the subsurface, represents a substantial sink for electrons from DMRB and the reduction of Fe(III) leads to the formation of biogenic Fe(II) phases. Indirect abiotic U(VI) reduction by reactive forms of biogenic Fe(II) minerals is a potentially important process for uranium immobilization. The DMRB Shewanella putrefaciens CN32 was used to synthesize two biogenic Fe(II) minerals: magnetite and vivianite. The present work elucidates abiotic molecular scale redox reactions between biogenic magnetite, vivianite and uranium. While both biogenic magnetite and vivianite reduced U(VI) completely, X-ray absorption spectroscopy (XAS) analysis indicated dramatic differences in speciation of the reduced uranium phase in each case. Biogenic magnetite favored formation of structurally ordered crystalline UO_2 and biogenic vivianite led to the formation of a U(IV)-sorption complex even though UO_2 has generally been considered to be the sole end product of U(VI) reduction. Biogenic magnetite treated with phosphate also reduced U(VI) to a U(IV)-sorption complex indicating the interference of phosphates in UO_2 formation.

The U(IV)-sorption complex could be potentially more labile than UO_2 in sub-oxic environments and thus may affect the long-term stability of reduced uranium species post remediation. This observation suggests a new paradigm for U(VI) reduction, one in which the speciation of reduced uranium is dependent on the bulk mineralogical composition in the surrounding environment.