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Interaction of U(VI) with Fe(II)-bearing montmorillonite: Implications for uranium immobilization in the subsurface environments

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Uranium is a major component of nuclear waste that can mobilize in underground repository environments due to various dynamic geochemical factors. Moreover, geogenic uranium contamination is present in many regions of the world, including the Indo-Gangetic plains in India where high dissolved uranium content in both shallow and deep aquifers has become a major public health issue. The fate and transport of uranium in subsurface environments can be controlled by their adsorption on common minerals, including phyllosilicates.

Montmorillonite is a 2:1 phyllosilicate that is both naturally present in the subsurface and a constituent of bentonite clay, a sealant material used in nuclear waste repositories. Montmorillonite is comprised of two types of “adsorption” sites- the interlayer fixed-charge cation exchange sites and the pH-dependent variable-charge edge sites, which behave differently under various pH and background ionic strengths. Montmorillonite contains a considerable pool of iron atoms in its structure which upon reduction forms Fe(II)-bearing montmorillonite, relevant in most subsurface and/or underground environments.

Reduction of U(VI) on Fe(II)-bearing montmorillonite following its adsorption can effectively control the mobility of uranium. The extent of reduction of U(VI) by Fe(II)-bearing montmorillonite was studied as a function of pH and background sodium ion concentration using X-ray absorption spectroscopy and chemical extractions. Complete reduction of U(VI) to U(IV) was found at a low sodium concentration both at acidic and near-neutral pH. However, at high sodium concentration, only 70% of U(VI) was reduced at near-neutral pH, thus highlighting the role of the type of adsorption sites on extent of reduction and associated immobilization of uranium. Bicarbonate-induced mobilization of surface bound U(VI) and U(IV) species on the montmorillonite surface was also tested. The results indicate easier leachability of surface bound U(VI) than surface U(IV) species. A surface complexation model interpreting the equilibrium adsorption of U(IV) on montmorillonite was developed which illustrates U(IV)-montmorillonite surface speciation. These findings establish the potential importance of structural Fe(II) in low iron content phyllosilicates in controlling uranium mobility in subsurface and underground deep repository environments. Moreover, if designed properly, Fe(II)-bearing phyllosilicate based permeable reactive barriers can potentially control uranium mobilization in known uranium contamination sites.