



Reduction of uranium under abiotic iron reducing conditions - A macroscopic and spectroscopic study

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The mobility of uranium in natural environments is directly dependent on redox conditions. Adsorption onto iron oxides and formation of aqueous carbonate complexes are antagonistic processes affecting the mobility of U(VI). U(IV) is considered to be virtually immobile due to the low solubility of U(IV) minerals such as uraninite or pitchblende. This study investigates possible redox transformations of uranium under transient redox conditions. Specific focus lies on the fate of U as reductive dissolution of iron oxyhydroxides by S(II) is initiated. In batch experiments sulfide was incrementally added to a lepidocrocite suspension containing adsorbed U(VI). The partitioning of uranium was monitored during the progressing transformation of lepidocrocite into FeS. Synchrotron-based X-ray absorption spectroscopy was used to resolve the oxidation state of uranium. Upon addition of sulfide intermediate release of U from the solid to the solution was observed. The mobilization of U was followed by immobilization in a later stage of the experiment. XAS reveals that this immobilization coincides with reduction of U(VI) to U(IV). Consequently, reduction of U(VI) and precipitation of U(IV) solids, due to a shift from oxidic to sulfate reducing conditions is possible. However, kinetic effects might lead to an intermediate mobilization of U that should be considered for the risk assessment of nuclear waste repositories and the remediation of sites, contaminated with radionuclides.