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Formation of surface U(V) by reduction of Ca-U-Carbonate complexes with nanoscale zero-valent iron

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While nanoscale zero-valent iron (nZVI) has the potential to be used as a permeable reactive barrier for the remediation of uranium-contaminated groundwaters, there remains some uncertainty regarding the processes of uranium immobilization and the nature of the precipitated phase(s). In this study the reduction of aqueous Ca-U(VI)-carbonate complexes by fresh, aged and corroded nZVI particles representing particles with three types of oxide shells were investigated at circumneutral pH. XAS, XPS and kinetic studies were applied to obtain information about the structural and chemical composition of the uranium-containing reaction products. Kinetic experiments demonstrated quick uranium removal by all types of nanoparticles with the uranium removal rate proportional to the extent of iron oxide layer formation indicating that the first stage is sorption onto iron oxide (magnetite) surface sites, followed by reduction by the zero-valent iron core. The final oxidation state of uranium depended on the mass ratio between iron and uranium. At 1:1 mass ratio uranium formed nanosized uraninite particles, while surface U(V) complexes were formed at higher iron loadings. U(VI)CaCO₃ complexes were reduced more slowly than binary U(VI)CO₃ complexes. We observed that the formation of U(V) complexes occurred only in presence of carbonate, which can be attributed to the stabilisation of surface U(V) by carbonate. Based on U and Fe EXAFS results structures for the U(V) complexes were proposed.

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