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Synchrotron radiation study of actinide sorption and diffusion in natural clay

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In several European countries, e.g., France, Germany and Switzerland, argillaceous rocks are considered as a potential host rock for the construction of nuclear waste repositories. Detailed information on the interaction between the clay and the actinides, which are major contributors to the radiotoxicity of spent nuclear fuel after storage times of more than 1,000 years, is required for the safety assessment of future nuclear waste repositories.

The interaction between redox-sensitive actinides (Np and Pu) and natural clay has been investigated in batch and diffusion experiments and by X-ray techniques. Opalinus Clay (OPA, Mont Terri Rock Laboratory, Switzerland), which was used as a reference for natural clay, consists of different clay minerals (66%), quartz (14%), calcite (13%), iron(II) minerals (4%) and traces of other minerals and organics. Due to this mineralogical heterogeneity, a combination of different synchrotron based techniques with micrometer resolution was used to study the sorption and diffusion of Np(V) and Pu(V), which have a high solubility at near neutral pH conditions. Different thin sections of OPA were contacted with 8 μM Np(V) or 20 μM Pu(V) solutions at pH 7.6 under aerobic conditions for several days. In the diffusion experiments, neptunium and plutonium were allowed to diffuse from a reservoir with 8 μM Np(V) or 20 μM Pu(V) into intact OPA bore cores during several weeks before they were removed from the diffusion cells. The thin sections and small pieces of the bore core were investigated at the microXAS beamline at the Swiss Light Source using monochromatic synchrotron radiation with a beam size of typically 3 x 1.5 μm^2 .

μ -XRF mappings of both sorption and diffusion samples showed heterogeneous distributions of neptunium and plutonium and other heavy elements (Ca, Mn, Fe, Sr) contained in OPA. Regions with high actinide concentrations, which frequently occurred in close vicinity to areas enriched in iron, were investigated by μ -XANES spectroscopy. By comparing these Np and Pu LIII-edge XANES spectra to those of reference spectra of neptunium and plutonium in different oxidation states, it was shown that the highly soluble pentavalent actinide species were reduced to the less soluble tetravalent oxidation state in all investigated samples. In some areas also Pu(III) was identified, although Pu(IV) remained the dominating oxidation state. Further information about the minerals present in areas enriched in neptunium or plutonium was obtained by μ -XRD. In addition to the clay mineral illite, siderite (FeCO_3) and pyrite (FeS) could be identified near actinide spots in several thin section samples, indicating that these Fe(II) minerals act as a reducing agents and causes the immobilization of neptunium and plutonium in OPA. The examples given in this presentation illustrate that μ -XRF, μ -XANES, and μ -XRD are a powerful combination of techniques for studying the speciation and migration behaviour of actinides in heterogeneous natural systems like argillaceous rocks with high spatial resolution.

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