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Investigating the Fate of Plutonium: Speciation of Plutonium during Diffusion in Opalinus Clay

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With regard to the safe disposal of heat-generating radioactive waste in deep geological formations, detailed information on the interaction between the radiotoxic, long-lived radionuclides such as 239Pu (t1/2 = 24,110 a) and the host rock as important geological barrier are required. A combination of spa-tially-resolved synchrotron based microprobe techniques has been used to determine the chemical speciation of the redox-sensitive Pu migrating through heterogeneous Opalinus Clay (OPA, Mont Terri, Switzerland).

Long-term diffusion experiments were conducted with the redox-sensitive Pu(V) species under aero-bic conditions. Spatially-resolved molecular-level investigations were performed at the microXAS Beamline of the Swiss Light Source (SLS, Paul Scherrer Institut, Villigen, Switzerland). First, micro-scopic chemical imaging by micro-X-ray fluorescence (μ-XRF) demonstrated the complex nature of the reactive transport pattern. A strong correlation between the distribution of Pu and the geochemical heterogeneity of the rock matrix (represented by major elements such as Ca, Mn, Fe, or Sr) was ob-served. The chemical speciation of the redox sensitive Pu along the migration pathway was deter-mined by micro-X-ray absorption fine structure spectroscopy (μ-XAFS). Generally, in all investigated diffusion (and in complementary sorption) samples, Pu LIII-edge μ-XANES spectra on Pu hot spots demonstrated that Pu(IV) is the dominating species on OPA. Accordingly, the highly soluble Pu(V) was reduced by components of the OPA rock materials and converted to the less mobile tetravalent oxidation state.

For the first time, by means of μ-XRF, we succeeded to record a full 2D representation of a diffusion profile of Pu in a relevant geological medium. Moreover, in addition to the elemental distribution of Pu, fundamental chemical information could be obtained by μ-XANES. These spatially-resolved speciation measurements of Pu showed that Pu(V) was reduced progressively along its diffusion path to Pu(IV). To gain further information about the redox-reactive mineral component of the OPA rock, micro-X-ray diffraction (μ-XRD) measurements were employed in areas of interest. A correlation between Pu(IV) with the Fe(II)-bearing mineral siderite and the clay mineral illite was observed by μ-XRD.

The combination of these spatially-resolved x-ray microprobe methods is a powerful tool to determine the transport mechanisms of actinides in heterogeneous systems. The obtained information is an essen-tial part of the evaluation of the long-term safety of a repository and increases also the confidence in clay rocks as option for the geological disposal of radioactive waste.

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