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Interaction of plutonium and neptunium with magnetite under anoxic conditions: Reduction, surface complexation, and structural incorporation

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For the redox-reactive fission products and actinides Se, Tc, U, and Np, it is assumed that the strongly reducing conditions in deep underground, anoxic nuclear waste repositories will reduce their mobility, since their lower-oxidation states commonly form solids of very low solubility. This is not necessarily the case for Pu, where the hexa- and pentavalent aquo-complexes prevalent at higher pe are replaced at lower pe by a tetravalent solid of low solubility, PuO2, but also by a trivalent aquocomplex at lower pH. FeII-bearing mineral phases, especially those with low bandgap, play an important role in this process, since they are able to catalyse redox reactions at their surface. Magnetite, FeIII2FeIIO4, is an important mineral in this context, since it forms by steel corrosion under anoxic conditions and is also prevalent as geogenic mineral. Therefore, we investigated the reaction of PuV, and in comparison also of NpV, with magnetite in sorption and coprecipiation experiments with X-ray absorption spectroscopy. We found that PuV is indeed reduced to the trivalent oxidation state in the presence of magnetite. The PuIII aquo-complexes are, however, strongly sorbed by forming tridentate surface complexes [1]. PuIII can also be partly incorporated by the structure of magnetite by rapid coprecipitation. During aging, however, it is expelled from the structure and readsorbed at the magnetite surface. This behaviour of Pu is then compared to that of Np.

Primary author: Dr SCHEINOST, Andreas (HZDR Institute of Resource Ecology)

Co-authors: Dr FELLHAUER, David (JRC-ITU); Mr ALTMAIER, Marcus (KIT-INE); Dr STEUDTNER, Robin (The Rossendorf Beamline at ESRF); Dr DUMAS, Thomas (CEA); Dr GAONA, Xavier (KIT-INE)

Presenter: Dr SCHEINOST, Andreas (HZDR Institute of Resource Ecology)

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