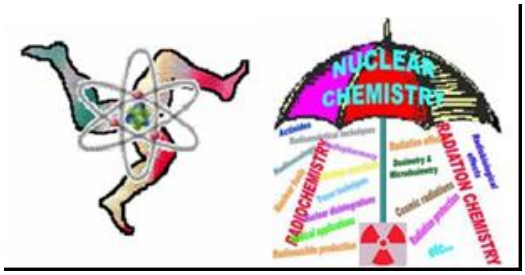

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Plutonium speciation during sorption on natural clay

Tuesday 20 September 2011 09:55 (15 minutes)

Plutonium will be a major contributor to the radiotoxicity of spent nuclear fuel after storage times of more than 1,000 years due to the long half-lives of Pu-239 and Pu-240. Several European countries are considering argillaceous rocks as potential host rock formation for high-level nuclear waste repositories. Plutonium has a very rich and complex chemistry in aqueous solution, where up to three Pu oxidation states can coexist. Therefore, detailed studies on the migration behavior of Pu are required for assessing the safety of deep geological disposal of nuclear waste in clay formations.

We investigated the sorption of Pu in oxidation states III-VI on Opalinus clay (OPA, Mont Terri, Switzerland) using several synchrotron-based techniques: X-ray absorption fine structure spectroscopy (EXAFS, μXANES), X-ray fluorescence (μXRF), and X-ray diffraction (μXRD). Pu L3-edge EXAFS measurements on OPA suspensions showed that inner-sphere surface complexes of tetravalent Pu dominate its speciation in the clay, independent from the initial Pu oxidation state in the aqueous phase. Similar results have been observed previously for the sorption of Pu on kaolinite [1], which is with 22 ± 2% an important clay constituent of OPA.

To determine which mineral phases of OPA are responsible for the uptake of Pu, thin sections of OPA bore cores were exposed to different Pu solutions and analyzed with high spatial resolution (≤ 1.5 μm) at the SLS MicroXAS Beamline. μXRF mapping revealed areas that were enriched in Pu. Pu-μXANES measurements showed that Pu(IV) is the major oxidation state in these areas. According to μXRD, clay minerals (kaolinite, illite), Fe(II) minerals (siderite, pyrite), calcite, and quartz dominate the mineralogy of the Pu-rich areas.

[1] T. Reich et al., Application of XAFS to Actinide Environmental Science, AIP Conf. Proc. 882 (2007) 179.

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