Contribution ID: **224** Type: **Poster**

Sorption behaviour and speciation of neptunium(V) on Opalinus clay

Thursday, 22 April 2010 12:00 (20 minutes)

The sorption of Np on clay such as Opalinus clay (OPA) is one of many important processes that affect the migration and retardation of Np in the geosphere. Clay formations are under consideration in several European countries as a potential host rock for high-level radioactive waste repositories [1]. We studied the sorption of Np(V) onto OPA (from Mont Terri, Switzerland) both by batch experiments and X-ray Absorption Spectroscopy (XAS). The aim of this study was to obtain a mechanistic understanding of the interaction of Np(V) with the OPA surface. The obtained information are necessary for the safety assessment and detailed planning of nuclear repositories.
The batch experiments were done under aerobic and anaerobic conditions. The sorption of Np(V) on OPA was studied as a function of many chemical parameters such as pH (6-10), background electrolyte (synth. OPA pore water, saturated calcite solution, and 0.1 M NaClO4), partial pressure of CO2, and presence/absence of humic acid (HA). The sorption curves for 8·10-6 mol/L Np(V) in saturated calcite solution obtained under aerobic and anaerobic conditions, respectively, show that the adsorption edge occurs between pH 7-8. Maximum sorption was observed at pH 8.5 with 65% sorption under aerobic conditions and 80% under anaerobic conditions, respectively. Under anaerobic conditions the reduction of Np(V) to Np(IV) by Fe(II) minerals of OPA causes a stronger sorption. At pH > 9 Np sorption decreased due to the complexation of Np with carbonate in aqueous solution [2]. Molecular-level information was obtained by X-ray Absorption Fine Structure (EXAFS). EXAFS spectra were measured in fluorescence mode at the Rossendorf Beamline ROBL at ESRF. Several samples at pH 7.6 and 8.5 with a total Np(V) concentration of ~ 8·10-6 mol/L were prepared under aerobic and anaerobic conditions. The amount of Np sorbed in the EXAFS samples was in the range of 50-121 ppm. Np L3-edge X-ray Absorption Near-Edge Spectra (XANES) showed that in all aerobic samples the pentavalent oxidation state of Np was dominated. The Np L3-edge EXAFS spectra of the anaerobic samples prepared in different background electrolytes showed the same EXAFS pattern, indicating that Np(V)-carbonato complexes are formed at OPA surface. More details as well as additional results will be presented and discussed.

The authors acknowledge the ESRF for provision of synchrotron beam time and thank the ROBL team for assistance during EXAFS measurements. This work was financially supported by the BMWi under contract No. 02E10166. D.R. Fröhlich has been supported by a fellowship of DFG-GRK 826.

[1] NAGRA: Projekt Opalinuston - Synthese der geowissenschaftlichen Untersuchungsergebnisse, Entsorgungsnachweis für abgebrannte Brennelemente, verglaste hochaktive sowie langlebige mittelaktive Abfälle. Technical Report NTB 02-03, NAGRA, Wettingen/Schweiz, 2002.

[2] V. Neck, W. Runde, J. I. Kim, B. Kanellakopulos, Solid-liquid equlibrium reactions of neptunium(V) in carbonate solution at different ionic strength. Radiochim. Acta. 65, 29-37 (1994).

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Session Classification: Poster Session - Chemistry of Nuclear Fuel Cycle, Nuclear Waste Management

Track Classification: Chemistry of Nuclear Fuel Cycle, Radiochemical Problems in Nuclear Waste Management