



ROBL-CRG

Experiment title:Thorium L_{III}-edge EXAFS measurements of Th treated montmorillonite**Experiment number:**

20_01_007

Beamline:

BM 20

Date of experiment:

from: 10.9.1999 to: 14.9.1999

Date of report:

20.02.2000

Shifts: 12**Local contact(s):**

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Received at ROBL:

20.03.2001

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Report:**Aims of the experiment and scientific background**

Clay minerals are present at significant level in argillaceous rock formations under consideration in many European radioactive waste disposal programs. Thus, an atomic/molecular level understanding of sorption mechanisms of radionuclides on clay surfaces is of fundamental importance for safety assessment of nuclear waste repositories.

On smectitic clays radionuclides and other metal ions can sorb as surface complex on edge sites and/or interlayer sites. XAFS studies with Ni and Co have shown, that another possible sorption mode is the formation of mixed lamellar phases. These mixed phases consist either of neoformed layer silicates [1] or hydrotalcite-like double hydroxide compounds [2-3]. The aim of this study is to use EXAFS to determine sorption mechanisms of Th on montmorillonite. Montmorillonite is an important smectitic mineral responsible for the retention of metals in the geosphere. Furthermore, the clay is used as a backfill material in the Swiss concept for a high level radioactive waste repository and thus, metal sorption on montmorillonite has been investigated in our laboratory in great details [4].

Experiments + Results

Samples were prepared in a glove box by reacting Th and montmorillonite at pH 5.0 and at high ionic strength (0.1 M NaClO₄) to block cation exchange processes. The initial Th concentrations for the samples were 3.5·10⁻⁴ M, 1.0·10⁻⁴ M and 4.0·10⁻⁴ M Th. After a

reaction time of 14 days the samples were centrifuged and the wet paste was transferred into a sealed Plexiglas sample holder. Th-L_{III}-edge fluorescence and transmission EXAFS spectra were recorded for samples containing 15, 50 and 207 $\mu\text{mol/g}$ Th sorbed onto the montmorillonite. Figure 1 shows the dependence of the k^3 -weighed Th spectra on the amount of Th sorbed onto montmorillonite. The corresponding radial distributions functions (RDF's) are shown in Figure 2. The first peak represents an oxygen shell at ~ 2.4 Å. The position and height of the peak does not depend on the Th concentration. The second RDF peak of the sample with the lowest concentration results from a Th-Si shell, suggesting specific binding of Th onto the montmorillonite surface. Figure 2 further reveals that with increasing Th concentration the second shell peak is shifted to longer distances in real space. This peak is most likely caused by an increasing Th-Th backscattering contribution. Detailed data analysis using theoretical approaches and further Th EXAFS studies with montmorillonite are currently ongoing.

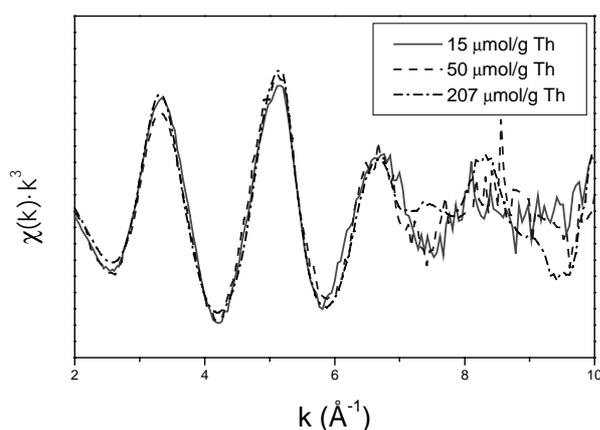


Figure 1: k^3 -weighed Th L_{III}-edge EXAFS spectra Th sorbed onto montmorillonite at 3 different Th-concentrations

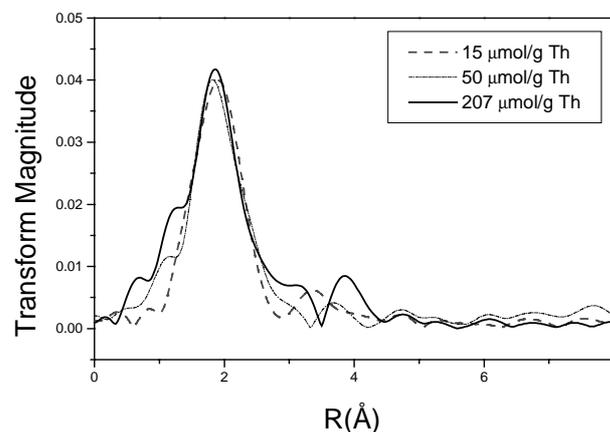


Figure 2: Concentration dependence of the Th L_{III}-edge RDF of Th sorbed onto montmorillonite

References

- [1] Schlegel M.L., Manceau, A., Charlet, L., and Hazemann, J.L. 2000. *American Journal of Science*. in press.
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- [3] Scheidegger A. M., Strawn D. G, Lamble, G. M., and Sparks, D. L. 1998. *Geochim. Cosmochim. Acta.* **62**, 2233-2245.
- [4] Baeyens B, and Bradbury M. H. 1997. *J. Contam. Hydrol.* **27**, 199-222.