



# Physikalisches Kolloquium

Horst Geckeis, Forschungszentrum Karlsruhe

» Geochemical aspects of nuclear waste disposal «

*Einführung: Th. Müller*

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Geochemical processes leading to either mobilization or retention of radionuclides in an aquifer system are significantly influenced by their interaction with rock, sediment and colloid surfaces. Therefore, performance assessment of nuclear waste disposal requires the elucidation and quantification of those processes. State-of-the-art analytical techniques as e.g. laser - and X-ray spectroscopy are increasingly applied to study solid-liquid interface reactions and to obtain molecular level speciation insight.

We have studied the sorption of trivalent lanthanides and actinides (M(III)) onto aluminium oxides, hydroxides and purified clay minerals by the time-resolved laser fluorescence spectroscopy and X-Ray-Fluorescence spectroscopy [1, 2]. Chemical nature and structure of surface bound actinides are proposed based on spectroscopic information. Similarities of spectroscopic data obtained for M(III) sorbed onto  $\gamma$ -alumina, gibbsite and clay minerals suggest the formation of very similar inner-sphere surface complexes at pH > 5 with the general formula  $\equiv\text{S-O-An(III)(OH)}_x^{(2-x)}(\text{H}_2\text{O})_{5-x}$ . Those speciation data are found consistent with those predicted by surface complexation modelling. The applicability of data obtained for pure mineral phases to actinide sorption onto heterogeneously composed natural clay rock is investigated by experiments and by geochemical modelling.

The assessment of clay colloid borne actinide migration observed in the frame of *in-situ* experiments at the Grimsel underground laboratory [3] also calls for detailed information on actinide speciation. Combined spectroscopy and wet chemistry experiments provide a fairly consistent picture of the actinide speciation under given groundwater conditions and impart insight into the dynamics of actinide-colloid interaction. Notably for tetravalent actinides we observe significant impact of ageing on the rate of actinide desorption from clay colloids. Studies are ongoing to elucidate underlying mechanisms.

Above mentioned studies demonstrate the appropriateness of combining spectroscopy and 'wet chemistry' as a strategy to validate geochemical model assumptions and to reduce uncertainties.

[1] Rabung, Th., Geckeis, H., Wang, X.K., Rothe, J., Denecke, M.A., Klenze, R., Fanghänel, Th., Radiochim. Acta 94 (2006), 609-618

[2] Rabung, Th., Pierret, M.C., Bauer, A., Geckeis, H., Bradbury, M.H., Baeyens, B.; Geochimica et Cosmochimica Acta, 69 (2005) 5393-5402

[3] Geckeis, H., Schäfer, Th., Hauser, W., Rabung, Th., Missana, T., Degueldre, C., Möri, A., Eikenberg, J., Fierz, Th., Alexander, W.R., Radiochim. Acta, 92 (2004), 765-774

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**Freitag, 29.06.2007, 17 Uhr c.t.,**

**Universität Karlsruhe (TH), Otto-Lehmann-Hörsaal, Physik-Flachbau (Geb. 30.22).**

**Anschließend Nachsitzung im Gastdozentenhaus „Heinrich Hertz“**