

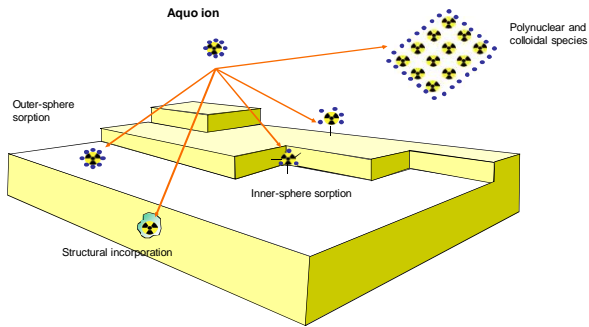
SORPTION OF TRIVALENT ACTINIDES (An^{3+}) ON MINERAL SURFACES

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OBJECTIVE

to understand the sorption mechanism at a molecular scale



SCOPE

An^{3+} : Pu^{3+} , Am^{3+} , Cm^{3+}

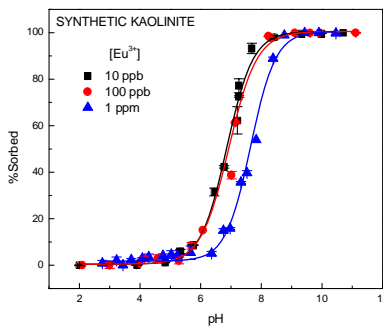
Minerals (S): kaolinite, gibbsite, γ -alumina

Sorption reaction: $S-OH + An^{3+} \leftrightarrow S-O-An^{2+} + H^+$

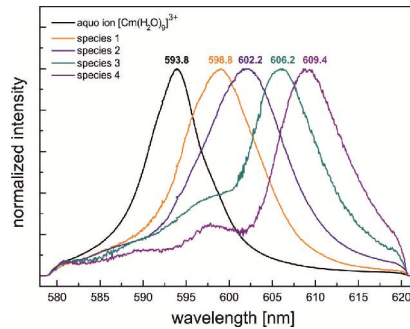
Methods:

- Batch sorption studies
- Laser-induced fluorescence spectroscopy (TRLFS) to identify sorbed surface An^{3+} species
- Nuclear magnetic resonance spectroscopy (NMR) to identify sorbing hydroxyl groups on the mineral surfaces

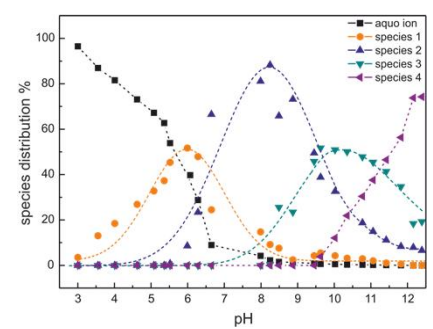
EXAMPLE: Cm^{3+} (Eu^{3+}) SORPTION ON KAOLINITE



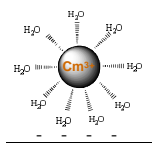
Batch sorption-% vs pH



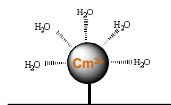
Fluorescence spectra of identified species



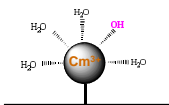
Distribution of species as a function of pH



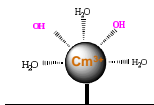
outer-sphere complex



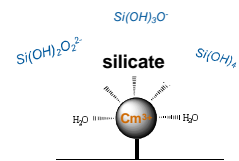
species 1
[>O-Cm(H₂O)₅]²⁺



species 2
[>O-Cm(OH)(H₂O)₄]⁺



species 3
[>O-Cm(OH)₂(H₂O)₃]



species 4
[>O-Cm(OH)_x(silicate)]

COLLABORATORS:

Karlsruhe Institute of Technology – INE – Germany (TRLFS)

Institute of Chemical Physics and Biosphysics, Tallinn, Estonia (NMR)

Scientific publications:

- N.Huittinen, Th.Rabung, J.Lützenkirchen, S.C.Mitchell, B.R.Bickmore, J.Lehto, H. Geckeis, *Sorption of Cm(III) and Gd(III) onto Gibbsite, α -Al(OH)₃*, J. Colloid and Interface Science 332(2009)158.
 N.Huittinen, Th. Rabung P. Andrieux, J. Lehto, H. Geckeis, *A Comparative Batch Sorption and TRLFS Study on the Sorption of Eu(III) and Cm(III) on Synthetic and Natural Kaolinite*, Radiochim. Acta 98(2010)613.
 N.Huittinen, P.Sarv and J.Lehto, *A ¹H NMR Study on the Specific Sorption of Y(III) and Eu(III) on γ -Al₂O₃*, J. Colloid and Interface Science 361(2011)252.
 N.Huittinen, Th. Rabung, J.Lützenkirchen, M.Hakanen, J.Lehto and H.Geckeis, *New Insight into Cm(III) Interaction with Kaolinite – Influence of Mineral Dissolution*, Geochim. Cosmochim. Acta 99(2012)100-109.
 N.Huittinen, P.Sarv and J.Lehto, *A ¹H and ²⁷Al NMR Investigation of Y(III) and Eu(III) Interaction with Kaolinite*, Appl. Clay Sci. 80-81 (2013) 182-188.
 N.Huittinen, *Sorption of Trivalent Actinides onto Gibbsite, γ -alumina, and kaolinite – A Spectroscopic Study of An(III) Interactions at the Mineral-Water Interfaces*, Doctoral Dissertation, University of Helsinki, 2013.