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INVITED LECTURE - Study of neptunium sorption on clay and clay minerals using X-ray absorption spectroscopy

Friday 21 September 2012 09:00 (20 minutes)

In several European countries argillaceous rocks have been considered as potential host rock for the construction of radioactive waste disposal repositories. Among potential migration paths, sorption and diffusion of radionuclides are the most important processes for the migration of these elements beyond the engineered barriers of the repository.
We have investigated the sorption of neptunium(V), the most mobile and soluble species of Np, on clay minerals [1] as well as on natural Opalinus clay (Mont Terri, Switzerland) [2]. The interactions between Np(V) and the clay and clay minerals have been studied by batch experiments as a function of several chemical parameters and by synchrotron-based techniques.
Before presenting the results for Opalinus clay, the most important findings of batch and EXAFS studies of Np(V) sorption on pure clay mineral phases will be briefly reviewed. Since Opalinus clay has a heterogeneous composition, the sorption of Np(V) has been studied by microfocused synchrotron X-ray fluorescence (μ-XRF) mapping, microfocused-XANES spectroscopy (μ-XANES) and microfocused X-ray diffraction (μ-XRD) at the Swiss Light Source. By combining these synchrotron-based techniques, the following results were obtained [3]: 1) Considerable amounts of Np(IV) were detected in areas that are enriched in Np, even when the sorption experiments with Np(V) were conducted in air. 2) Under anaerobic conditions, a correlation between Np and Fe was observed in the μ-XRF maps, indicating that the reduction of Np(V) is caused by an iron(II)-containing mineral that could be identified as pyrite by μ-XRD. The reduction of Np(V) to the less mobile Np(IV) is an important process that increases the sorption of this long-lived radiotoxic element and retards its migration through the clay.

References

[1] S. Amayri et al., Radiochim. Acta 99 (2011) 349
[2] D.R. Fröhlich et al., Radiochim. Acta 99 (2011) 71
[3] D.R Fröhlich et al., Anal. Bioanal. Chem. (submitted)

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