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Ultra-trace determination of actinides in clay systems with accelerator mass spectrometry and development of a diffusion setup for reducing conditions

Clay rock is a potential host rock for the final disposal of high-level nuclear waste (HLW) in deep geological formations. In the host rock, reducing conditions are expected correlating with low actinide (An) solubility and strong An sorption on clay rock. Consequently, reduced An species are known to be rather immobile. In order to verify this, there is an interest in performing An diffusion experiments through natural clay under reducing conditions, resembling as much as possible conditions expected in a deep geological repository. Due to the very low expected An concentrations, an analytical method is needed which is able to determine An down to ultra-trace levels (≈ 10^{-19} mol/sample).

In the frame of this work, such an analytical procedure was tested. Samples simulating the sample matrix obtained after a diffusion experiment through natural clay were produced by spiking Opalinus Clay or Callovo-Oxfordian Clay and corresponding pore water samples with ^{233}U, ^{237}Np, ^{244}Pu, ^{243}Am, and ^{248}Cm in the concentration range 10^{-15}–10^{-19} mol per sample. Subsequently, a group extraction of these An and their determination with accelerator mass spectrometry (AMS) was performed [1].

First diffusion experiments with ^{233}U(VI) and ^{243}Am(III) in samples of the Opalinus Clay are in planning.

The results will provide relevant insight in the diffusion of strongly sorbing actinides at ultra-trace concentrations in clay matrices.

References:

[1] F. Quinto, et al. Anal. Chem. 2017, 89, 7182-7189.