Contribution ID: **13** Type: **Oral**

Microscale analysis of U(VI) uptake by argillaceous rocks using synchrotron radiation X-ray methods

Wednesday 21 May 2014 14:30 (25 minutes)

Argillaceous rock formations, due to their high clay content and low permeability, have significant radionuclide retention/retardation capacities, and are therefore in the focus for high level and long lived nuclear waste (HLW) repositories in several countries. In Hungary two geological sites (Gorica Block and W-Mecsek Anticline Block in the perianticlinal structure of W-Mecsek Mountains) of the Boda Claystone Formation (BCF) have been selected for the study of potential host rocks for HLW. The aim of the measurements was to obtain information on the U(VI) uptake mechanism on the micrometer scale, in order to complete molecular scale sorption results and understand the behaviour of uranium dissolving from HLW and to identify the possible differences in uptake capacity between the two possible sites.
Synchrotron radiation microscopic X-ray fluorescence (SR μ-XRF) has sufficient sensitivity to study the metal uptake on the microscale [1]. SR μ-XRF measurements were performed on thin sections subjected to uptake experiments involving U(VI) using 5 μm spatial resolution. The thin sections were prepared on high-purity silicon wafers from geochemically characterized cores of both sites of BCF. Correlation analysis of μ-XRF elemental maps indicated that the U enrichment was not only correlated to the argillaceous matrix, but also the cavity filling minerals played an important role in the uptake. By using positive matrix factorization as a new multivariate approach the factors with higher uptake capacity could be identified. The uptake capacity of the different mineral phases could be quantified with additional mineralogical information [2]. The results were compared with cluster analysis when the regions dominated by different mineral phases were segmented. The multivariate approach based on μ-XRF to identify the minerals was finally validated using microscopic X-ray diffraction.
Our results revealed that in the sample, taken from W-Mecsek Anticline Block, where dolomites have ankerite rims and U-bearing rings, newly formed FeOOH precipitations were observed, which partly replaced the ankerite at near neutral pH (6.8). In this sample the dissolution of ankerite was followed by FeOOH formation, which easily can bind U(VI) due to the enhanced specific surface area and high adsorption capacity. The oxidation of Fe(II) required to the formation of FeOOH is caused by partial reduction of U(VI) to U(IV). Eighty percent of uranium was taken up by clay minerals and 20% by FeOOH although the ankerite concentration is as low as 6% in W-Mecsek Anticline Block of BCF. The study demonstrated that the different mineralogy of Gorica Block and W-Mecsek Anticline Block significantly influences the U retention capacity of the Boda Claystone Formation [3].

[1] C. Walther, M.A. Denecke. Chemical Reviews 113 (2013) 995-1015.
[2] J. Osán, A. Kéri, D. Breitner, M. Fábián, R. Dähn, R. Simon, S. Török. Spectrochimica Acta Part B 91 (2014) 12–23.
[3] D. Breitner, J. Osán, M. Fábián, P. Zagyvai, C. Szabó, R. Dähn, M. Marques Fernandes, I.E. Sajó, Z. Máthé, S. Török. Submitted to Environmental Earth Sciences

Primary author: Dr OSAN, Janos (Hungarian Academy of Sciences Centre for Energy Research)

Co-authors: Ms KERI, Annamaria (HAS Centre for Energy Research); Dr FABIAN, Margit (HAS Centre for Energy Research); Dr DÄHN, Rainer (Paul Scherrer Institut); Dr TOROK, Szabina (HAS Centre for Energy Research)

Presenter: Dr OSAN, Janos (Hungarian Academy of Sciences Centre for Energy Research)

Session Classification: Actinides in Environmental and Life Sciences

Track Classification: Actinides in Environmental and Life Sciences