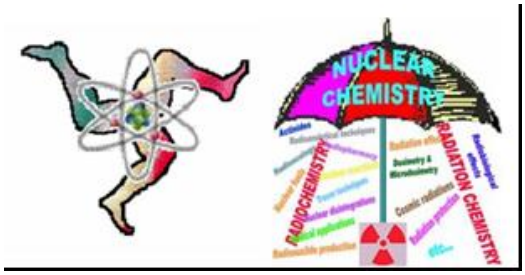

Contribution ID: **87** Type: **oral presentation**

Micro-XRF and micro-EXAFS investigations of radionuclide sorption in Boda Siltstone Formation

Wednesday 21 September 2011 12:35 (15 minutes)

Argillaceous rocks are being considered in many radioactive waste management programmes as suitable host rock formations for the deep geological disposal of radioactive waste. Clay minerals such as illite, smectite, illite/smectite mixed layers and kaolinite are important components in such rock types and can often make up 50 or more wt.% of the total mass. One of the most important characteristics of many clay minerals is their generally strong radionuclide retention properties.

The rocks investigated were prepared from geochemically characterized cores from drillings in Boda Siltstone Formation, Hungary. The samples are albitic clay stones that represent the most significant constituent of Boda Siltstone Formation. Two polished 30-60 μm thick thin sections were prepared, and the thin sections were placed for 72 hours into a synthetic ground water solution representative for Boda Siltstone Formation with 0.1 mM Ni2+ or 1 mM Ni2+ added. After the treating period the samples were dried up by high pressure Ar gas.

The micro-distribution and the local environment of nickel in the argillaceous rock samples were investigated applying micro x-ray fluorescence (micro-XRF) and micro extended x-ray absorption fine structure (micro-EXAFS) investigations at the micro-fluorescence Beamline L at HASYLAB (Hamburg, Germany). A polycapillary half-lens (X-ray Optical Systems) was employed for focusing a beam of 3 ×1.5 mm2 down to a spot size of 15 μm diameter. The EXAFS spectra were recorded in fluorescence mode at the K-absorption edge of Ni, recording the Ni-Kα fluorescent yield using an energy-dispersive silicon drift detector. A series of micro-EXAFS measurements were performed on the thin sections, at points of interest (POI) with varying Ni content.

Micro-XRF elemental maps collected at an excitation energy of 17500 eV showed an anticorrelation with Ca and Fe, and a correlation with K-rich phases. This indicates that mainly clay rich minerals are responsible for the Ni uptake in the Boda formation. The precise identification of the particular nature of the clay minerals responsible for the sorption would need additional micro-XRD investigations.

Using micro-EXAFS, the formation of precipitates and surface complexes could be clearly distinguished. The strong second peak in the radial structure functions (RSF) at R+ΔR = 2.7 Å indicates the formation of a precipitate from the Ni-containing synthetic ground water solution on the rock surface, and can mainly be attributed to Ni-Ni backscattering pairs. The micro-EXAFS spectra of a few POIs of the samples treated with 0.1 mM Ni2+, however, can be fitted without any Ni-Ni backscattering pair, indicating the formation of a surface complex. The obtained fitted structural parameters (5.3 Ni-O at 2.05 Å, 1 Ni-Al at 3.03 Å, and 2.1 Ni-Si at 3.3 Å) are consistent with Ni-uptake on edge sites of clay minerals.

The research leading to these results has received funding from the Swiss-Hungarian Cooperation Programme through Project n° SH/7/2/11 and from the European Community's Seventh Framework Programme (FP7/2007-2013) under grant agreement n° 226716.

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Session Classification: Session 9

Track Classification: Radioecology and Geochemistry