Micro-XRF and micro-XRD study of the uptake of nickel and uranium by argillaceous rocks

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The present study focuses on the interaction of selected metals representing actinides and fission products with the host-rock of the planned high level radioactive waste (HLW) repository in Hungary (BSF). The aim is to identify the mineral phases responsible for the uptake of Ni and U by the host rock, using combined micro-XRF and micro-XRD. Small pieces of selected core samples from BSF were grinded (ca. 30 μm) and mounted onto a high purity Si holder and polished with 0.25 μm diamond paste. The samples were subjected to 72-hour sorption experiments with the elements of interest at different concentrations (0.1 mM or 1 mM in 0.1 M NaCl solution).

The measurements were performed at the micro-fluorescence Beamline L of HASYLAB. A Ni/C multilayer double monochromator and a single-bounce capillary were used to focus the incoming X-ray beam in a spot of 22 μm in diameter at 17.5 keV. Elemental maps were collected for adsorbed elements (Ni, U) as well as for the major and minor elements of the rock using a stepsize of 20 μm and 1 s counting time per pixel. Based on the micro-XRF maps, positions of interest for micro-XRD analyses were selected in order to investigate a correlation between the element of interest and elements representative for distinct minerals. Diffraction images were collected employing a MAR CCD165 detector using counting times of 60 to 120 s. The *XRDUA* software [1] was used to perform the azimuthal integration of the XRD images. For samples treated with a 0.1 mM Ni^{2+} solution, micro-XRF and micro-XRD revealed that mainly illite is responsible for the Ni uptake of the rock (Fig. 1) [2]. For samples treated with UO_2^{2+} or a 1 mM Ni^{2+} solution, no correlation was found between K-rich phases and the element of interest.

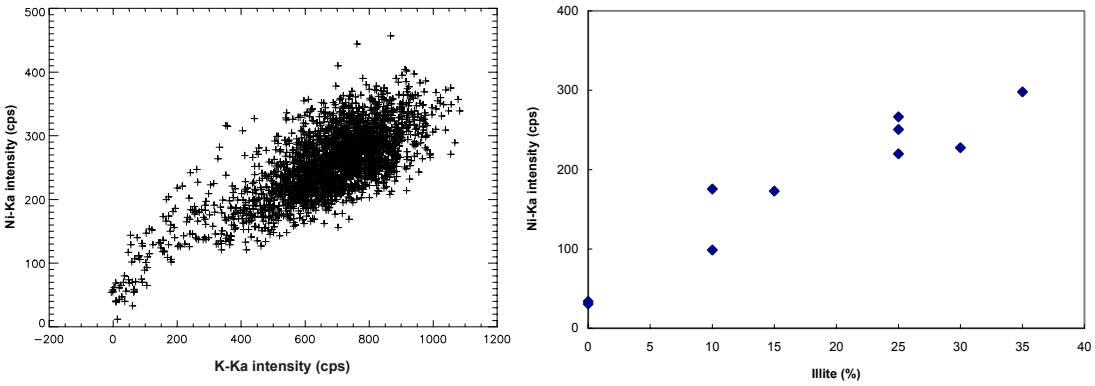

Figure 1: Scatter plot of K-Kα and Ni-Kα X-ray intensities for all pixels of the elemental map (left), and Ni-Kα X-ray intensity versus illite content derived from micro-XRD at selected positions (right)

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References

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