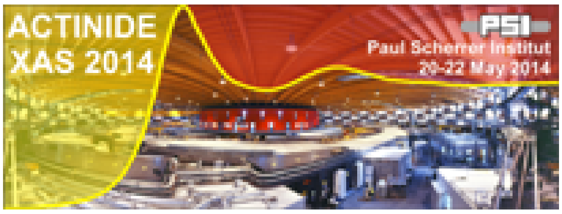

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Application of positive matrix factorization and microscopic X-ray fluorescence spectroscopy in the examination of argillaceous rocks

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Argillaceous rocks are considered in most European countries as suitable host rock formations for the deep geological disposal of high-level radioactive waste (HLW). The most important chemical characteristic in this respect is their generally strong radionuclide retention property due to the high sorption capacity. Consequently, the physico-chemical parameters of these processes have to be studied in great detail. Synchrotron radiation microscopic X-ray fluorescence (SR micro-XRF) has sufficient sensitivity to study these processes on the microscale without the necessity of the application of radioactive substances. The studies at the Environmental Physics Department of Centre for Energy Research of Hungarian Academy of Sciences focus on the interaction between the escaped ions and the host-rock surrounding the planned HLW repository. SR micro-XRF measurements were performed on thin sections subjected to sorption experiments using 5 μm spatial resolutions. Inactive Cs(I), Ni(II), Nd(III) and natural U(VI) were selected for the experiments chemically representing key radionuclides. The thin sections were prepared on high-purity silicon wafers from geochemically characterized cores of Boda Claystone Formation, Hungary. Samples were subjected to 72-hour sorption experiments with one ion of interest added. The micro-XRF elemental maps taken usually on several thousand pixels indicate a correlation of Cs and Ni with Fe- and K-rich regions suggesting that these elements are predominantly taken up by clay-rich phases. U and Nd was found to be bound not only to the clayey matrix, but the cavity filling minerals also played important role in the uptake. Multivariate methods were found to be efficient tools for extracting information from the elemental distribution maps even when the clayey matrix and fracture infilling regions were examined in the same measured area. By using positive matrix factorization as a new approach the factors with higher sorption capacity could be identified and with additional mineralogical information the uptake capacity of the different mineral phases could be quantified. The results were compared with cluster analysis when the regions dominated by different mineral phases are segmented. The multivariate approach based on micro-XRF to identify the minerals was validated using microscopic X-ray diffraction.

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