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Sorption studies of Np(V) onto the zircaloy corrosion product ZrO_2

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The interactions of dissolved radionuclides, such as the actinide neptunium(V), with corroded phases in the near field of a repository are crucial processes to be considered in a safety assessment of a nuclear waste disposal for highly radioactive waste. Zirconia (ZrO_2), the corrosion product of the zircaloy cladding material of spent nuclear fuel rods, represents one of the first barriers encountered by mobilized radionuclide ions.

In this study, the interactions between Np(V) and monoclinic zirconia were studied at room temperature on a macroscopic and molecular scale. The influence of different parameters (time, pH, ionic strength, [Np(V)]) was investigated by means of batch sorption experiments to gain information on the macroscopic level. Starting at pH 3, an increased uptake of Np(V) with increasing pH was observed, reaching its maximum at pH $\geq$ 7. The Np(V) sorption was found to be independent of ionic strength, indicating Np(V) inner-sphere complexation on the ZrO_2 surface. This finding was supported by electrophoretic mobility measurements at different Np(V) concentrations. A shift of the isoelectric point of the neat ZrO_2 to higher pH values in the presence of Np(V) suggests the same Np(V) binding mode.

In situ ATR-FTIR spectroscopy was applied to gain a deeper understanding of the Np(V) sorption processes on a molecular level. These experiments allow a thorough characterization of the surface speciation including the number of occurring species, their denticity, and their reversibility of formation. Subsequently, this information will be used for the surface complexation modelling (SCM) to parametrize a comprehensive description of the Np(V)-ZrO_2 system, which in turn will contribute to a more reliable prediction of the fate of Np(V) in the environment.