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## Uptake of Np(V) by zirconia: a combined batch, spectroscopic, and surface complexation modeling study

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The interactions of long-lived actinides, such as the transuranium element neptunium, with corrosion products in the near-field of a repository are important processes that have to be considered when assessing the safety of a nuclear waste repository. As a main corrosion product of the zircaloy cladding material of spent nuclear fuel rods, zirconia ( $\text{ZrO}_2$ ) constitutes a first possible barrier against the release of radionuclides.

To gain a detailed understanding of the surface processes in the Np(V)–zirconia system, a comprehensive, multi-method approach was applied. The Np(V)– $\text{ZrO}_2$  system has been studied on the macroscopic level by conducting pH-dependent batch sorption experiments under varying conditions (ionic strength, Np(V) concentration, and solid-to-liquid ratio (m/V)). In addition, a Np(V) sorption isotherm at pH 6 was collected. The results revealed that Np(V) sorption onto  $\text{ZrO}_2$  was affected by pH, Np(V) concentration, and solid-to-liquid ratio. Uptake of Np(V) increased with pH, starting around pH 3 with maximum sorption reached from pH 6. The shift of the sorption edge towards lower pH with increase of the m/V ratio points to the presence of different kinds of sorption sites. This is supported by the Np(V) sorption isotherm results, where the shape suggests strong and weak binding sites. Furthermore, Np(V) uptake was found to be independent of ionic strength and zeta potential measurements revealed a shift towards higher pH values of the isoelectric point of the neat  $\text{ZrO}_2$  in the presence of Np(V). Hence, the formation of Np(V) inner-sphere surface complexes is indicated.

Molecular information about the surface species were obtained by Extended X-ray Absorption Fine Structure Spectroscopy (EXAFS) and *in situ* Infrared (IR) Spectroscopy, revealing the predominant formation of inner-sphere Np(V) surface complexes. A short Np–Zr distance derived from EXAFS suggests the presence of Np(V) bidentate inner-sphere complexes on the  $\text{ZrO}_2$  surface.

These information obtained on a macroscopic and a molecular level can be used to restrict the number of surface species as well as their denticity in a surface complexation model. The thermodynamic surface complexation parameters of the Np(V)–zirconia system derived in this study will help to make more reliable predictions about the fate of Np(V) in the environment.

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