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Adsorption of U(VI) onto zirconia (ZrO_2): a batch, spectroscopic, and modeling investigation

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The safety of high-level radioactive waste repositories depends on understanding radionuclide interactions with corrosion products in the repository near field. Zirconia (ZrO_2), a corrosion product of zircaloy cladding on nuclear fuel rods, acts as an important first barrier against radionuclide release.

A combination of experimental approaches was applied to elucidate the adsorption behavior of U(VI) at the water–zirconia interface under CO_2(g)-free conditions. pH-dependent batch sorption experiments, at varying ionic strength, U(VI) concentration, and solid-to-liquid ratio, were complemented by sorption isotherms (pH 3.5 and 4.3). Sorption of U(VI) increased with pH, starting at pH 3 and reaching maximum uptake at pH ≥ 6. The lack of ionic-strength dependence indicates predominantly inner-sphere surface complexation. Electrokinetic measurements corroborated this interpretation, showing a shift of the isoelectric point of pristine ZrO_2 toward higher pH in the presence of sorbed U(VI). The observed shift of adsorption edges to lower pH at higher solid-to-liquid ratios indicates heterogeneous surface reactivity involving both strong and weak sorption sites, consistent with the isotherm experiments.

Under acidic conditions, U(VI) predominantly formed a mononuclear bidentate inner-sphere surface complex according to *in situ* attenuated total reflection Fourier-transform infrared spectroscopy and extended X-ray absorption fine structure spectroscopy analyses. Increasing pH favored the predominance of a polynuclear U(VI) hydrolysis surface species, while laser-induced luminescence spectroscopy at alkaline pH indicated the additional formation of a mononuclear U(VI) hydrolysis species.

Macroscopic and molecular-level data were employed to parameterize a charge-distribution multi-site complexation model, resulting in thermodynamic constants that improve predictions of U(VI) environmental fate in repository performance assessments and contribute to safety evaluations of extended interim spent nuclear fuel storage [1].

[1] I. Demnitz-Jessat et al., New insights on the U(VI) Uptake on ZrO_2: a combined batch, spectroscopic and modeling approach, *Journal of Hazardous Materials*, **2026** (in preparation).