SORPTION BEHAVIOUR OF DI- AND TRIVALENT RADIONUCLIDES ON OXIDESURFACES AT HIGH SALT CONCENTRATIONS

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Adsorption processes are important in retarding the potential migration of radionuclides from a nuclear waste repository to the biosphere [1–3]. Many studies have been carried out to characterize the interactions of key radionuclides on many surfaces for a wide range of solution compositions. The major body of the studies was performed with salt concentrations of 0.1M and below ([4] and references therein). For salt concentrations significantly higher than 0.1M there is no systematic data set to allow an extension of the present surface complexation approaches to higher salt contents within a variable charge model.

In this work, the sorption behaviour of various oxide minerals (quartz, ZrO_2(s) and FeOOH) at moderate to very high ionic strength conditions, from 0.1 to 5 M NaCl, is presented with respect to di- and trivalent cations. Quartz presents well-known surface properties, with surface functional groups also present on the surface of clays and clay minerals, while ZrO_2(s) has been used as an oxidized zircalloy template, and goethite, FeOOH(s) as a representative of Fe^{3+} solid phases (iron corrosion phases). As adsorbates of interest we have selected Eu(III) and Sr(II). The Eu(III) cation is often used as a chemical analogue for trivalent actinides like Am(III) and Cm(III), which are important radionuclides in nuclear waste. Sr(II), and specifically ^{90}Sr, is a part of the elements within the fast/Instant Release Fraction (IRF), which may be directly released to water in the case of canister failure.

A self-consistent Surface Complexation Model (SCM) has been developed for the full set of experimental data gathered in this work, from 0.1 to 5 M NaCl, using a coupled Pitzer/surface complexation approach. The Pitzer model was applied to aqueous species, while a Basic Stern Model was used for interfacial electrostatics of the system, which includes ion-specific effects via ion-pair formation. The dielectric constant of each respective solution was applied. Parameter fitting was done using the general parameter estimation software UCODE coupled to a modified version of FITEQL2 involving separate calculations of the respective ionic strength corrections.

It is expected that models as were derived within the present study can potentially be applied within the Safety Case for a nuclear waste repository in highly saline environments.

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References

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