

Sorption of Eu(III) and Am(III) on magnetite in NaCl brine

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In deep underground nuclear waste repositories groundwater may contact the emplaced waste packages inducing corrosion of steel containers. Corrosion products represent an additional chemical barrier that may immobilize radionuclides (RN) and thus limit migration of RN to the far-field. For a site in saline formation the aquatic systems are expected to have high ionic strength (IS), which may affect RN retention processes in the chemical barriers. At present, hardly any RN sorption data onto Fe corrosion products have been reported under such saline conditions. In this study, the adsorption of Am(III) and its chemical analogue Eu(III) on magnetite, which is expected to form as a major steel canister corrosion product, was investigated under anoxic conditions as a function of ionic strength set by NaCl.

The sorption of Eu(III) on magnetite (solid/liquid ratio = 0.5 g/L) was investigated batch wise in NaCl brine with IS = 1, 3 and 5 M, as a function of pH_m (Eu concentration = $5.1 \cdot 10^{-10}$ M) and as a function of Eu concentration (10^{-9} M to 10^{-4} M) at constant pH_m 6 and 7. The pH_m range in the experiments was limited to pH_m 5.5 as a lower limit due to dissolution of magnetite and to pH_m 10 as an upper limit. A high immobilization capacity of > 90 % was found above pH_m 6, with $\log K_D$ values of 5.3 L/kg at IS = 1 M and 4.4 L/kg at IS = 5 M. It was concluded from batch experiments that higher IS provides slight but non-significant decrease of the sorption capability of Eu on magnetite. Results from batch sorption experiments were analysed with a surface complexation model. Surface acidity constants and site densities were determined by modelling potentiometric titrations of the solid. Information on the chemical nature of the surface-sorbed species was obtained by X-ray absorption spectroscopy (XAS) at the Am L_3 edge for Am(III) contacted with magnetite in NaCl brine with IS = 1 and 5 M at pH_m 6 and 7 respectively. Preliminary XAS results are consistent with the Eu(III) batch sorption experiments outcomes, confirming negligible effect of ionic strength on the sorption.

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