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Speciation and surface complexation modelling of Np(V) sorption on montmorillonite

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The sorption of Np(V) on Na-montmorillonite (STx-1) has been studied by batch experiments, spectroscopic measurements, and surface complexation modelling with the aim to contribute toward a better understanding of the sorption of 237Np (t1/2 = 2.1 × 106 a) in the near field (bentonite backfill material) and far field (argillaceous rocks) of high-level nuclear waste repositories. Batch experiments were performed in the absence of inorganic carbon and under air-equilibrated conditions with 0.1 and 0.01 M NaClO4 as background electrolyte, 8 × 10-12 and 9 × 10-6 M Np(V), and 3 ≤ pH ≤ 10. At pH > 8 the presence of inorganic carbon has a strong influence on the sorption behavior of Np(V) due to the formation of aqueous Np(V) complexes with carbonate.

Neptunium LIII-edge extended X-ray absorption fine structure (EXAFS) measurements on Np(V)/montmorillonite samples with Np(V) loadings in the range of 0.3-3.5 µmol/g have been performed to determine the speciation of Np at the solid-liquid interface. The EXAFS spectra of samples prepared under ambient air conditions (pCO2 = 10-3.5 atm) revealed the formation of Np(V)-carbonate complexes at the montmorillonite surface.

The results of the batch experiments obtained under CO2-free conditions could be modeled using the two site protolysis non-electrostatic surface complexation and cation exchange (2SPNE SC/CE) model described in [1]. For modeling the sorption behavior of Np(V) on montmorillonite in the air-equilibrated system, the aqueous complexation of Np(V) with carbonate [2] was included and the following additional surface complexation reaction was required: ≡SOH + NpO2+ + CO32- ⟺ ≡SONpO2CO32- + H+.

[1] M.H. Bradbury and B. Baeyens, Modelling the sorption of Mn(II), Co(II), Ni(II), Zn(II), Cd(II), Eu(III), Am(III), Sn(IV), Th(IV), Np(V) and U(VI) on montmorillonite: Linear free energy relationships and estimates of surface binding constants for some selected heavy metals and actinides, Geochim. Cosmochim. Acta 69, 875-892, 2005.
[2] Chemical Thermodynamics of Neptunium and Plutonium, (Eds. J. Fuger et al.) Elsevier, Amsterdam 2001.

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