Influence of ionic strength on neptunium(V) sorption onto gibbsite

Tao Wu, S. Amayri, T. Reich

Institut für Kernchemie, Universität Mainz, D-55099 Mainz, Germany

Neptunium is one of the most important elements from the safety point of view due to its hazardous nuclide Np-237 with a long half-life of 2.1×10^6 years. For the safety assessment and design of nuclear waste repositories, studies on the kinetics, thermodynamics, redox behaviour and speciation of Np in the rock formation of the repository and the aquifer, after a possible release from the repository, are required. Montmorillonite and other clay minerals like kaolinite contain octahedral layers of [AlO₆] as structural units. Different experimental data for the sorption of Np(V) are available for various oxides and hydrous oxides of aluminum [1], montmorillonite and kaolinite [2-4]. To better understand the role of these aluminium layers for the sorption of Np(V) onto clays, we are investigating Np(V) sorption on the reference mineral gibbsite (α -Al(OH)₃).

The batch experiments were done in the presence of CO_2 , 60 h equilibration and 26,000 rpm centrifugation for 1 h. The sorption of 8.0×10^{-6} M Np(V) onto gibbsite (Merck, 1.22 m²/g) was studied in the solid-to-liquid ratio range of 4-20 g/L. The batch experiment shown in Fig. 1 was done in pH 9.0. The Np uptake increased with increasing solid-to-liquid ratio more than 50% at 20 g/L.



Fig. 1 Sorption of 8.0×10^{-6} M Np(V) as function of solid-liquid ratio at pH 9.0.

The sorption of 8.0×10^{-6} M Np(V) at 15 g/L gibbsite was studied in the pH range 6.0-10.0. The batch experiments shown in Fig. 2 were performed at ionic strength of 0.01 and 0.1 M NaClO₄. The uptake of Np(V) by gibbsite strongly increased above pH 6.5 and reached its sorption maximum at pH 8.5 with 50% sorption. Above pH 8.5, the amount of Np(V) sorbed onto gibbsite decreased to 5% at pH 10.0. The formation of neptunium-carbonato complexes above pH 8.5 reduces the amount of

Np sorbed onto the mineral surface in the airequilibrated system.



Fig. 2 Sorption of 8.0×10^{-6} M Np(V) as function of pH at two electrolyte concentrations.

Fig. 3 demonstrates the influence of ionic strength on the sorption of 7.0×10^{-12} M Np(V). The batch experiment was done at ionic strength of 0.01 and 0.1 M NaClO₄. The uptake of Np(V) reached its maximum sorption at pH 8.5 with 90% sorption. In the air-equilibrated system, the sorption of Np(V) was independent from ionic strength, indicating inner-sphere sorption.



Fig. 3 Sorption of 7.0×10^{-12} M Np(V) as function of pH at two electrolyte concentrations.

Acknowledgments

This work was supported by the BMWi grant No. 02E10166

References

- [1] Tochiyama, O. et al., Radiochimi. Acta. 73, 191 (1996).
- [2] Li, W. J. et al., J. Colloid Interface Sci. 267, 25 (2003).
- [3] Turner, D. R. et al., Clays Clay Miner. 46, 256 (1998).
- [4] Bertetti, F. P. et al., In Adsorption of Metals by Geomedia Ed. Jenne E. A., Ed. Academic Press, San Diego, California, 132 (1998).