

Sorption of neptunium on kaolinite under environmental conditions

A. Jermolajev, J. R. Kasbohm, S. Amayri, T. Reich

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

The sorption behavior of neptunium(V) on the reference clay mineral kaolinite (KGa-1b, Source Clay Repository, Washington County, Georgia, USA) has been studied at an initial neptunium concentration of 7×10^{-12} M. The isotope Np-239 (half live 2.355 days), produced at the research reactor TRIGA Mainz, was employed due to its low detection limit in γ -spectroscopy. We investigated the sorption behavior of Np(V) as a function of pH (6.0 – 11.0), ionic strength (0.01 – 1.0 M NaClO₄), absence and presence of ambient CO₂, and time (20 minutes – 60 hours).

Experimental

The batch experiments were done at 25 ± 3 °C in 15 or 50 mL polypropylene centrifuge tubes. The solid to liquid ratio was 4 g/L. During the preconditioning (72 h) and contact time (60 h), the samples were agitated by overhead rotation at 16 rpm. The phases were separated by centrifugation at 4025 g for 7 minutes.

The CO₂-free experiments were done in argon atmosphere in a glove box ($O_2 \leq 20$ ppm).

Results

As can be seen from Fig. 1, the presence of ambient CO₂ strongly affects the sorption of Np(V) onto kaolinite. The decrease of the sorption in the pH range 9 – 11 is probably due to a competing reaction on Np in solution, i.e., the formation of Np(V) carbonato complexes [1]. At pH 9 Np(V) sorption increases with increasing ionic strength both in the presence and absence of ambient CO₂ (Table 1). However, the relative increase is larger in the CO₂ equilibrated system, indicating differences in the sorption mechanism between the two systems.

Figure 2 shows that after approx. 100 min the amount of Np sorbed or desorbed is constant. At pH 9.0 80 % of 7×10^{-12} M Np(V) is sorbed onto kaolinite and 20 % remains in solution. For the desorption experiment, the sorption sample was centrifuged and 0.1 M NaClO₄ solution at pH 9.0 (equilibrated with ambient CO₂) was added to the kaolinite. 20 % of the sorbed Np was desorbed and 80 % remained at the kaolinite surface (Fig. 2). This experiment shows that the interaction of 7×10^{-12} M Np(V) at the kaolinite-water interface is fast and reversible, at least at pH 9.0.

Acknowledgements

This work was supported by BMWA (Bundesministerium für Wirtschaft und Arbeit) under contract No. 02E9653. J.R. Kasbohm

from the University of Arkansas thanks the DAAD for its support through the RISE program.

References

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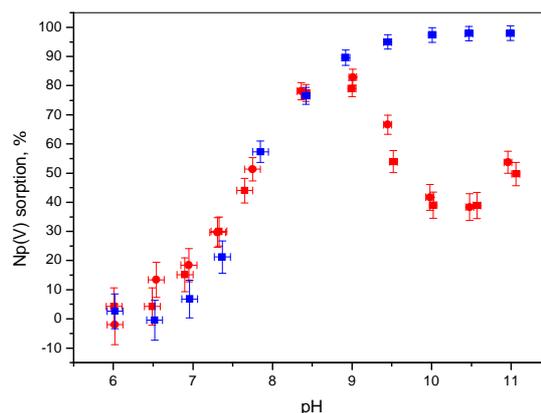


Fig. 1. The pH dependence of the sorption of Np(V) onto kaolinite in the presence (red points) and in the absence (blue points) of ambient CO₂.

Table 1. The dependence of the sorption of Np(V) on ionic strength in the presence and absence of ambient CO₂ at pH 9.0.

<i>I</i> , M	CO ₂ -free	ambient CO ₂
	Np(V) sorption %	
0.01	88.7(6)	72(1)
0.05	88.4(7)	80(1)
0.10	89.8(6)	85.0(8)
0.50	94.2(3)	87.7(7)
1.00	95.8(2)	86.5(8)

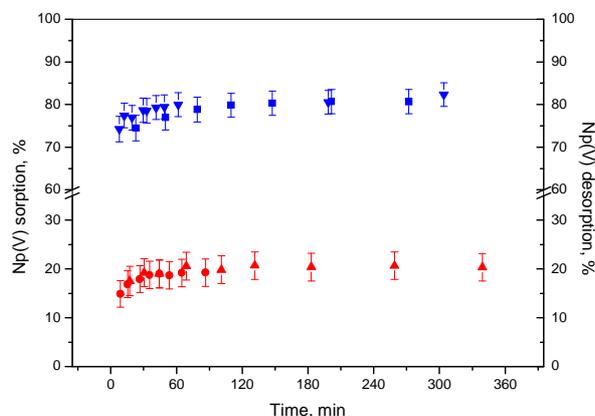


Fig. 2. Time dependence of Np(V) sorption (blue points) and desorption (red points) onto kaolinite in the presence of ambient CO₂ at pH 9.0.