

Sorption of neptunium on kaolinite under environmental conditions

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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.

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References

[1] V. Neck, W. Runde, J.I. Kim., Journal of Alloys and Compounds **225**, 295 (1995).

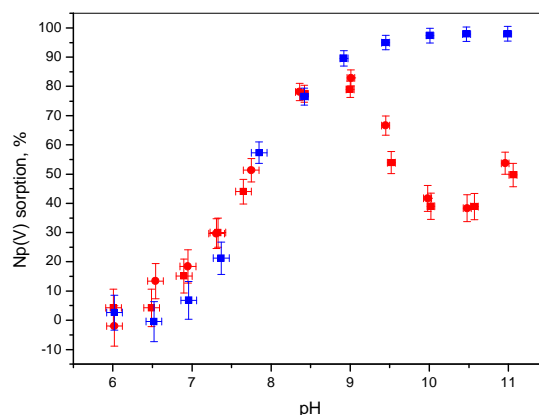


Fig. 1. The pH dependence of the sorption of Np(V) onto kaolinite in the presence (red points) and in the absence (blue points) of ambient CO₂.

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)

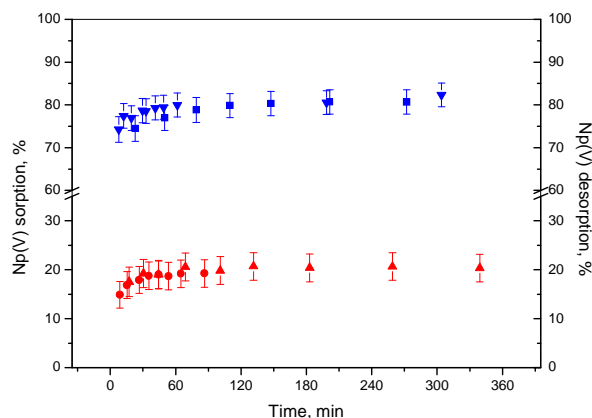


Fig. 2. Time dependence of Np(V) sorption (blue points) and desorption (red points) onto kaolinite in the presence of ambient CO₂ at pH 9.0.