NEPTUNIUM(V) SORPTION ONTO MONTMORILLONITE AND BENTONITE COLLOIDS AND THE INFLUENCE OF COLLOIDS ON Np(V) TRANSPORT

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INTRODUCTION

The bentonite buffer in EBS consists mainly of montmorillonite which, like other aluminosilicates is known to retain radionuclides, thus, contributing to the retention or immobilization of them. Long-lived Np-237 (2.144×10^6 a) in the pentavalent oxidation state forms a neptunyl cation NpO_2^+, which is rather soluble, poorly sorbed and readily mobile making it highly relevant for research concerning SNF repository safety. The potential relevance of colloids for radionuclide transport is highly dependent on the formation of stable and mobile colloids in different environmental conditions and the interaction of radionuclides with the formed colloids. In this study we investigated the sorption of Np(V) on bentonite and montmorillonite colloids in batch sorption and the influence of mobile colloids on Np(V) transport in granite column experiments.

SORPTION AND DESORPTION

Sorption studies as a function of pH for montmorillonite, bentonite colloids and crushed Kuru gray (Figure 1, left) and desorption studies at pH 8, 9 and 10 for montmorillonite (Figure 1, right).

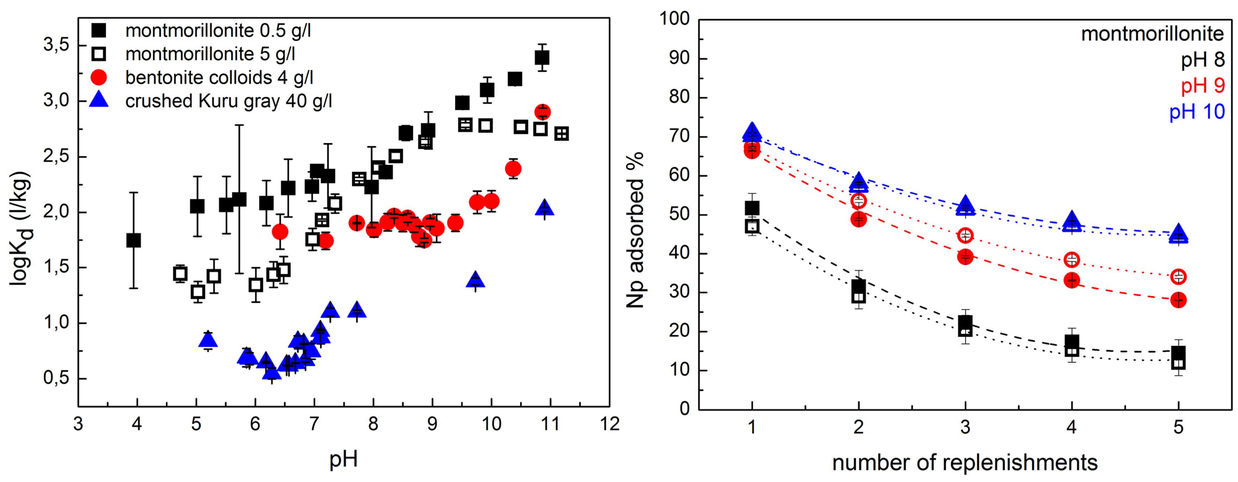

Figure 1. Np(V) sorption onto montmorillonite (black squares), bentonite colloids (red dots) and crushed Kuru gray (< 1 nm) in 10 mM $NaClO_4$ as a function of pH (c(Np) = 10^{-6} M) (left). Desorption of Np(V) from montmorillonite at pH 8 (black), 9 (red) and 10 (blue) with equilibration time of 2 – 3 days (open symbols) or 7 days (closed symbols) (right).

COLUMN EXPERIMENTS

The experiment set up of the column experiments are presented in Figure 2 (left). The flow properties of a column were determined by using a non-sorbing tracer (Cl-36) to determine the elution times in two columns at two different flow rates (Figure 2, right).

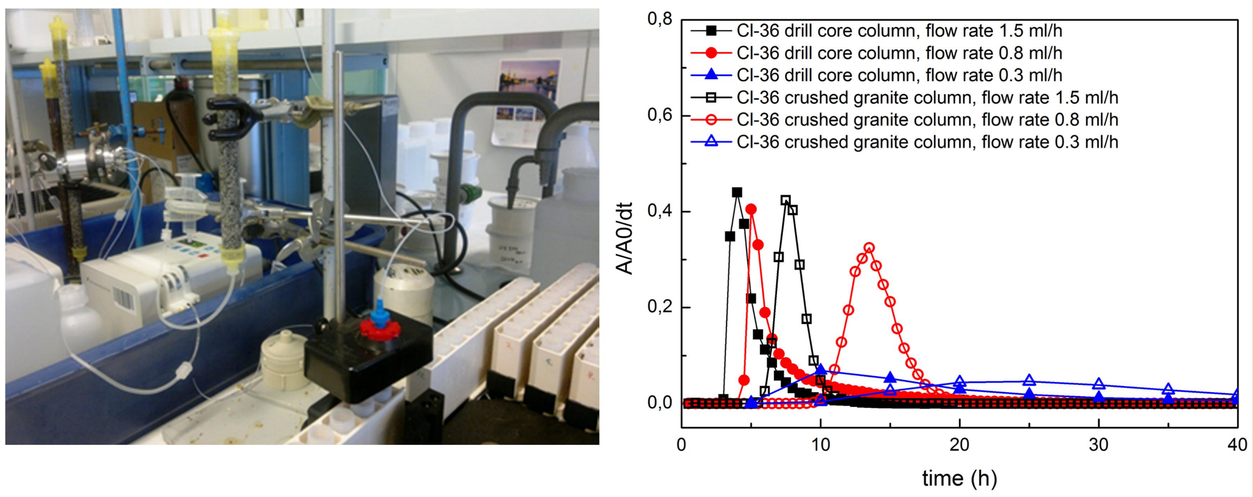

Figure 2. The experimental set-up for column experiments (left). The measured breakthrough curves of Cl-36 through the drill core column (black squares) and the crushed granite column (red dots) with flow rates of 1.5 ml/h, 0.8 ml/h and 0.3 ml/h (right).

To study the effect of colloids on Np-237 migration column experiments in the presence and absence of bentonite colloids were conducted. Np(V) break through curves without colloids (open symbols) and with bentonite colloids (solid symbols) are presented in Figure 3. The particle size and presence of colloids were analysed by photon correlation spectroscopy (Figure 4).

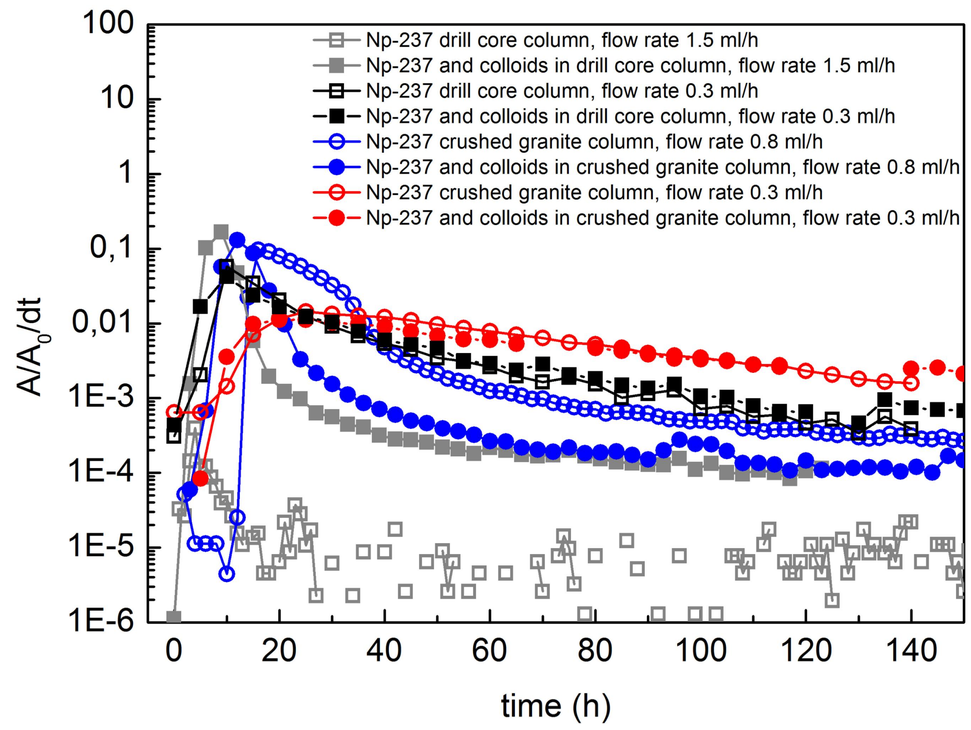

Figure 3. The measured breakthrough curves of Np-237 through drill core column (squares) and crushed granite column (dots) with flow rates of 1.5 ml/ h (gray symbols), 0.8 ml/h (blue symbols) and 0.3 ml/h (black symbols for drill core and red symbols for crushed granite column), in 10 mM $NaClO_4$. Open symbols in the absence of colloids and solid symbols in the presence of colloids.

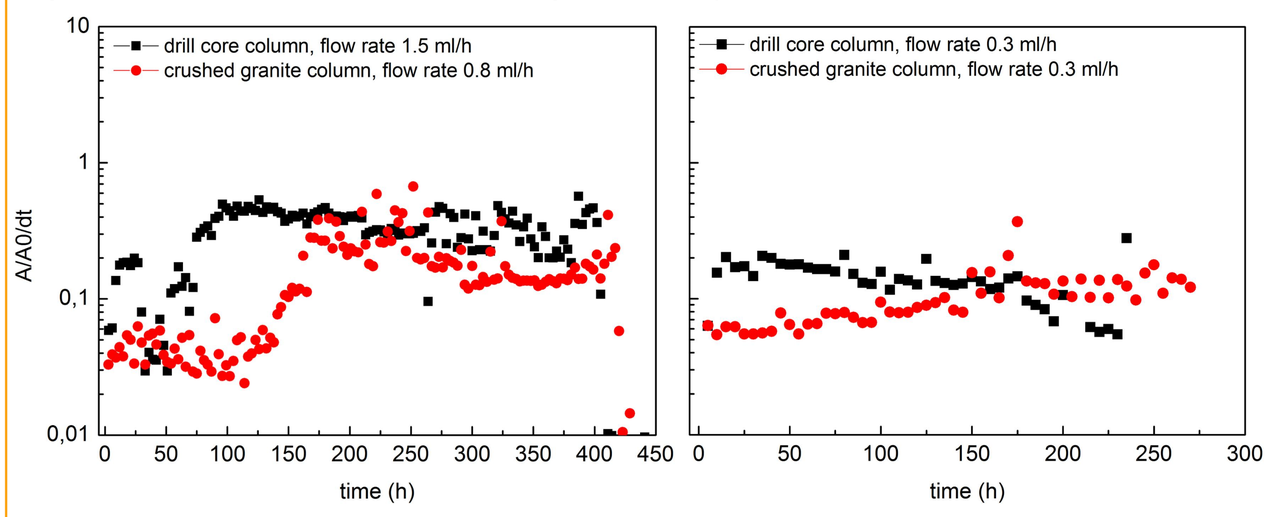

Figure 4. Colloids eluted through the drill core column with flow rate of 1.5 ml/h (black squares) and through the crushed granite column with flow rate of 0.8 ml/h (red dots). Np-237 was injected in the drill core column after 24.5 h and in the crushed granite column after 31.5 h (left figure). Colloids eluted through the drill core and the crushed granite columns with flow rate of 0.3 ml/h. Np-237 was injected in both columns after 70 h. Colloid solution was changed to 10 mM $NaClO_4$ after 192.5 h in the drill core column.

CONCLUSIONS

- At environmentally relevant pH 8 Np(V) sorption onto bentonite colloids is rather low in comparison with montmorillonite
- Np-237 sorption on montmorillonite is not irreversible and desorption from montmorillonite decreases with increasing pH
- In the presence of bentonite colloids with flow rates of 1.5 and 0.8 ml/h Np-237 eluted faster through both columns. However, with the lower flow rate of 0.3 ml/h the presence of colloids had no influence on Np-237 migration.
- The slight effect of matrix diffusion on Np-237 can be observed in the crushed granite column experiments, whereas at the presence of bentonite colloids (flow rate 0.8 ml/h) matrix diffusion cannot be observed.

This research has received funding from the European Atomic Energy community's Seventh Framework Programme (FP7/2007-2011) under grant agreement n° 295487 and Finnish Research Programme on Nuclear Waste Management (KYT 2018).

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