EFFECTS OF BENTONITE COLLOIDS ON THE RADIONUCLIDE MIGRATION IN GRANITIC ROCK

Pirkko Hölttä, Outi Elo, Valtteri Suorsa, Elina Honkaniemi, Suvi Niemiaho and Eini Puhakka

Laboratory of Radiochemistry, Department of Chemistry
University of Helsinki, Finland

INTRODUCTION

The bentonite erosion resulting in the formation of colloids may have a direct impact on the overall performance of the bentonite buffer used in the repository for spent nuclear fuel (SNF).

If colloids are sufficiently stable and mobile, irreversible sorption on colloids may increase radionuclide transport in the geosphere.

The objective was to study radionuclide sorption on bentonite colloids, colloid stability and mobility and the effect of colloids on radionuclide migration.

EXPERIMENTS

Materials and methods:

- MX-80 Volclay bentonite (76 %) and Nanocor PGN Montmorillonite (98 %).
- Reference groundwater Allard (I = 4.2 mM) or diluted OLSO (I = 0.517 M; 1–100 mM)
- Colloid suspension: ultrasonic separation and centrifugation (12000 rpm/20 min), 270 – 530 nm, 2.4 - 5.9 g/L

Bentonite erosion and colloid stability:

- 1 g powder or 2 pellets + 50 mL solution with and without slow agitation
- The stability of colloids was determined using Photon correlation spectroscopy and the dynamic electrophoretic mobility (Malvern Zetasizer Nano ZS)

Batch sorption experiments:

- Colloid dispersion + 90 mL solution + Sr-85 or Eu-152 tracer
  → 4.7 mL aliquot after 2 h, 1, 2 and 7 days
  → Ultracentrifugation (90000 rpm/60 min)
  → Radioactivity measurement
- In a clove box under CO_2 free conditions

Column experiments:

- Kuru Grey granite (KGG) Drill core column (28 cm, 4.4 cm, 0.5 mm)
- KGG and Sievi altered tonalite Crushed rock columns (15 and 30 cm, i.d. 1.5 cm).

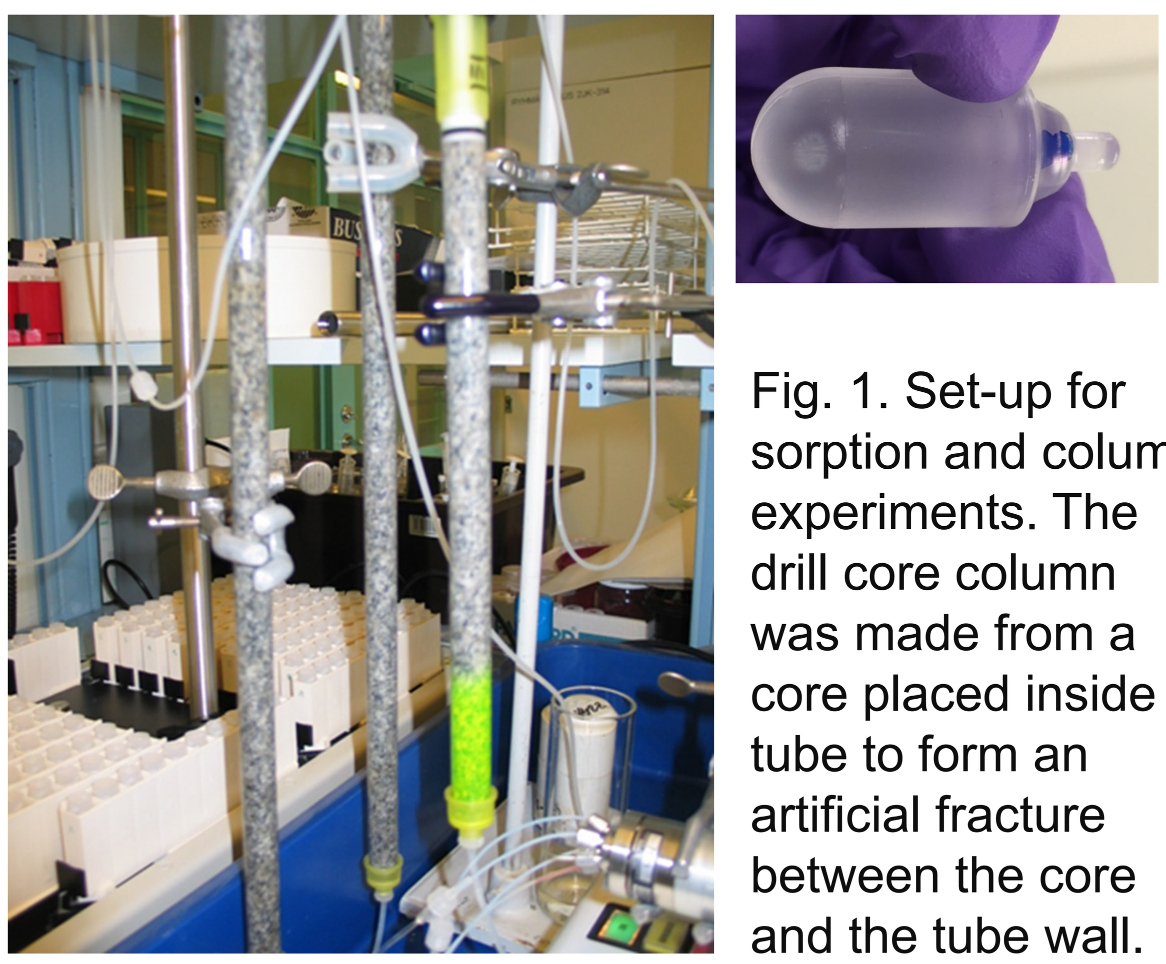

Fig. 1. Set-up for sorption and column experiments. The drill core column was made from a core placed inside a tube to form an artificial fracture between the core and the tube wall.

RESULTS

The bentonite erosion and colloid stability depended strongly on the ionic strength and the valence of the cations.

In dilute solutions (1 –5 mM), the mean particle diameter was under 400 nm and ZP lower than -30 mV, indicating stable colloids (Fig. 2). Noticeable colloid generation occurred only in the most diluted solutions and the concentration reached the level where no more colloids were released.

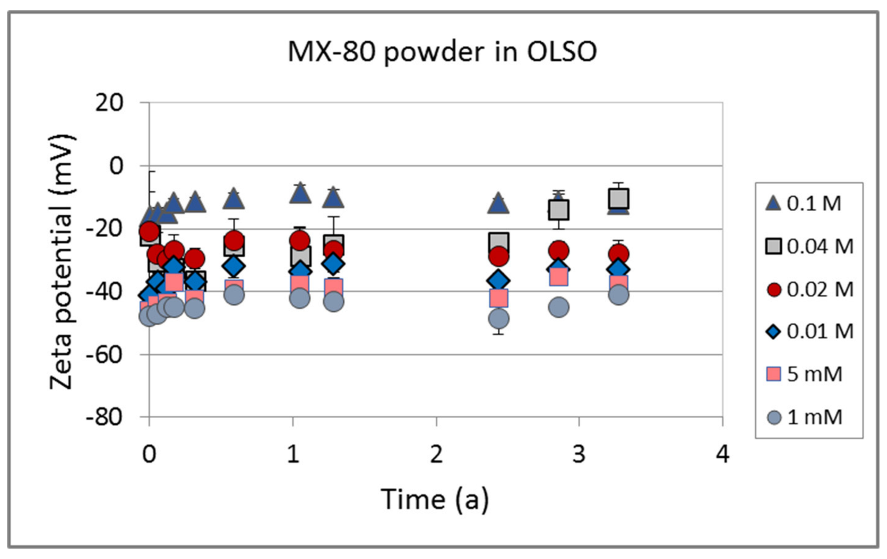

Fig. 2. Mean zeta potential of colloids formed from MX-80 bentonite in diluted OLSO water.

Sr-85 and Eu-152 sorption was highly dependent on pH, adsorption increasing with increasing pH (Fig. 3). Adsorption decreased with increasing ionic strength due to particle aggregation and lower specific surface area.

According to calculations, Eu^{3+} coordinates in the middle of Si-O ring on the tetrahedral layer of montmorillonite (Fig. 4)

60 % of Eu-152 was desorbed from Nanocor colloids and suspension (Fig. 5).

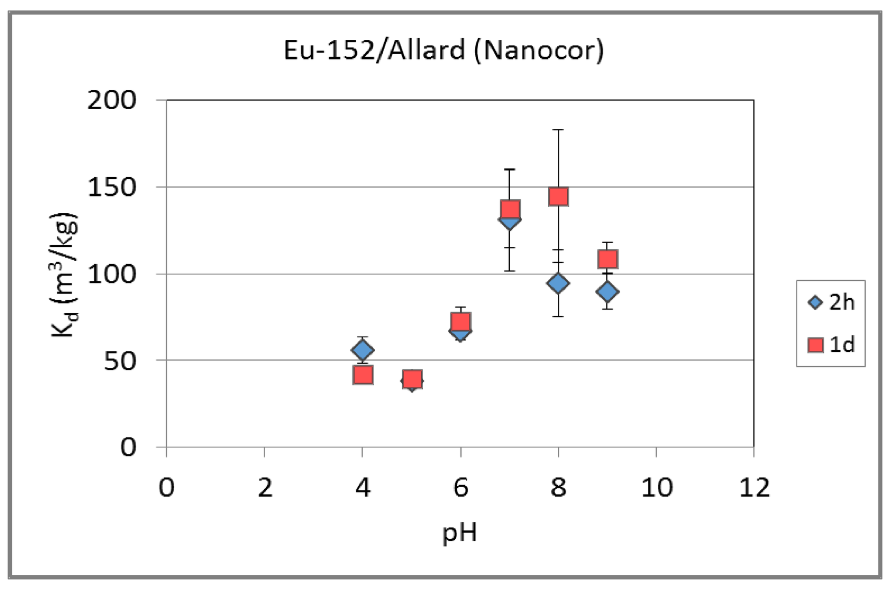

Fig. 3. K_d-values of Eu-152 for Nanocor colloids in Allard water.

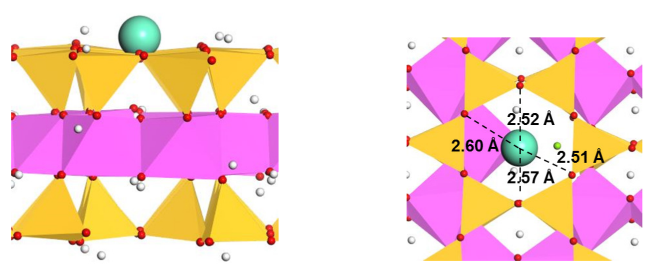

Fig. 4. Coordination of Eu^{3+} on the basal surface of montmorillonite.

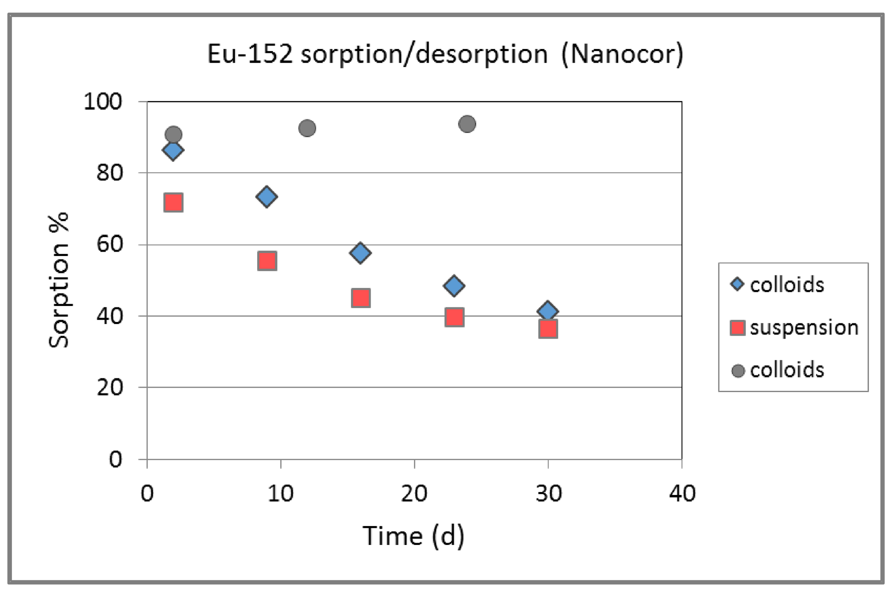

Fig. 5. Eu-152 desorption in Nanocor colloid solution and suspension in Allard water (pH 8).

The recovery of colloids (Fig. 6) was low and depended on column and rock type. Slowing down the water flow rate, the recovery was decreased.

In the presence of bentonite colloids, the faster elution of Sr-85 was obtained in Drill core column (Fig. 7).

No elution of Eu-152 was detected in two weeks without or with colloids owing to the injected short pulse resulting in very low colloid concentration available.

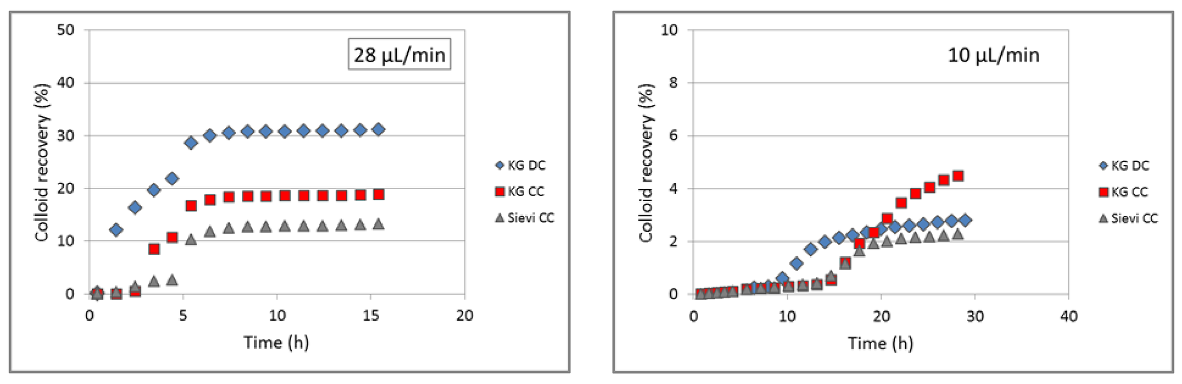

Fig. 6. Colloid recovery in KGG Drill core column (blue), KGG (red) and Sievi (grey) Crushed rock columns (15 cm). Average particle size 230 nm.

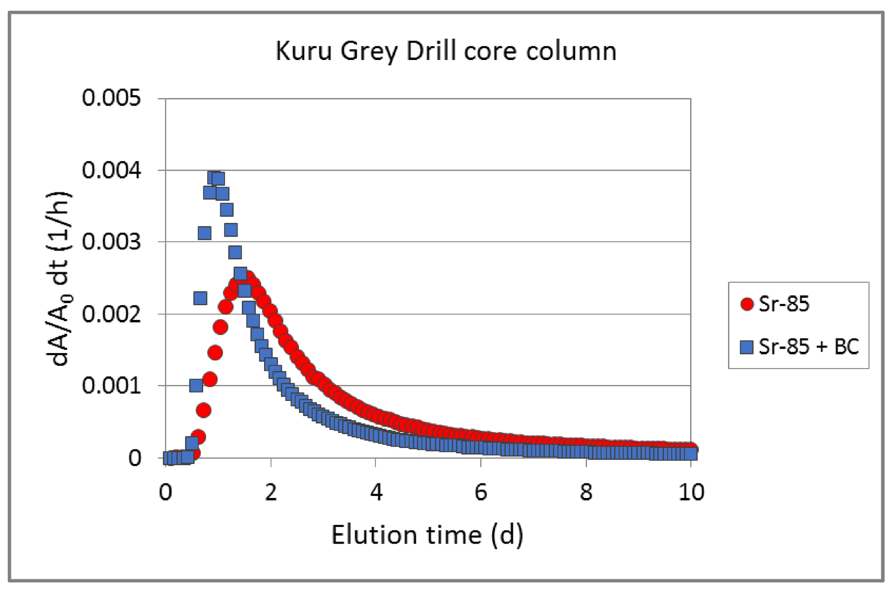

Fig. 7. The elution of Sr-85 through KGG Drill core column in the absence (red) and presence (blue) of MX-80 bentonite colloids. Allard water flow rate was 10 µL/min.

CONCLUSIONS

Geochemical conditions have a significant influence on bentonite colloid stability and the radionuclide sorption.

Europium sorption on colloids was reversible, however, fully reversibility was not obtained.

Mobility of colloids was affected by the flow rate, colloid size, column material and type.

Laboratory scale experiments showed that colloids had an effect on radionuclide transport. However, the water flow rates were orders of magnitudes faster than the groundwater flow.

ACKNOWLEDGEMENT

The 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).

NRC9, Helsinki, Finland, August 29 – September 2, 2016