Radionuclide interaction and transport – Update of laboratory experiments

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Objectives

- To determine the release and stability of bentonite colloids in different groundwater conditions
- To determine radionuclide sorption on MX-80 bentonite colloids and montmorillonite
- To study colloid/radionuclide and host rock interaction in dynamic conditions
- To apply new methods to study radionuclide sorption

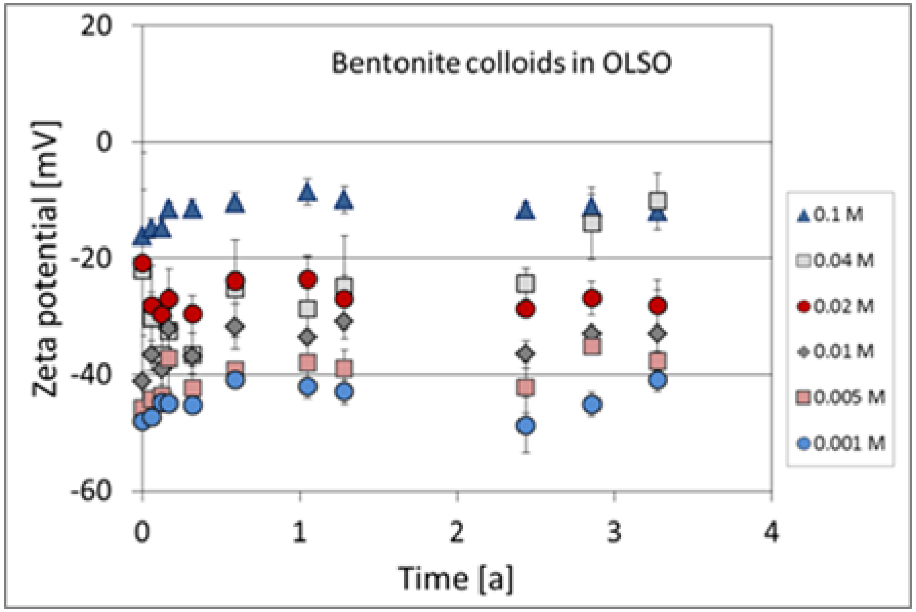
Materials and methods

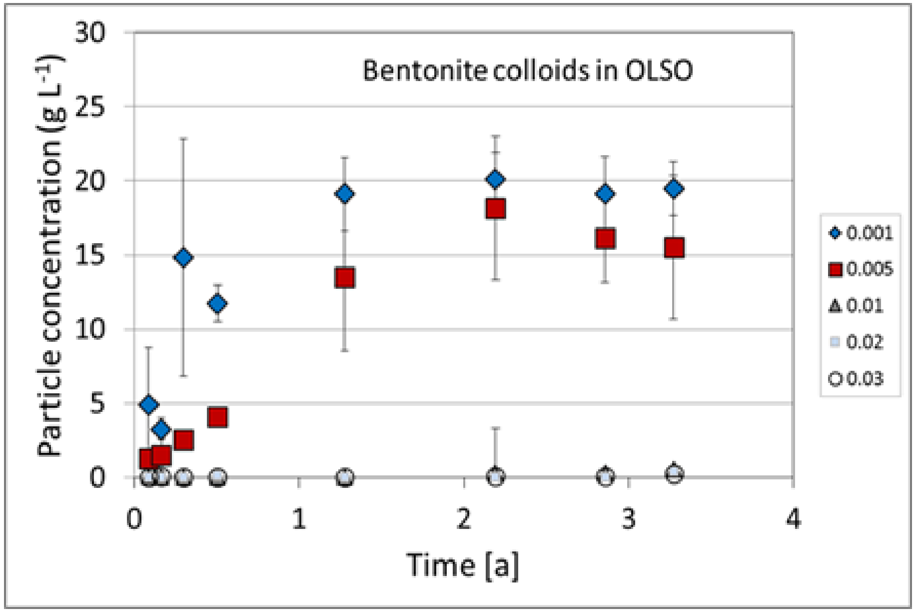
- MX-80 Volclay bentonite powder and pellets, Na-montmorillonite provided by B+Tech
- Reference groundwater: OLSO (I = 0.517 M) and Allard (I = 4.2 mM)
- NaCl and $CaCl_2$ solutions (I = 1 mM - 0.1 M)
- Photon correlation spectroscopy (PCS): particle size, concentration and zeta potential
- ICP-MS: Colloid concentration: Al determination
- Small angle x-ray scattering SAXS: Colloid and gel phase characterization
- In-situ ATR FT-IR, EXAFS

Bentonite erosion and stability of colloids

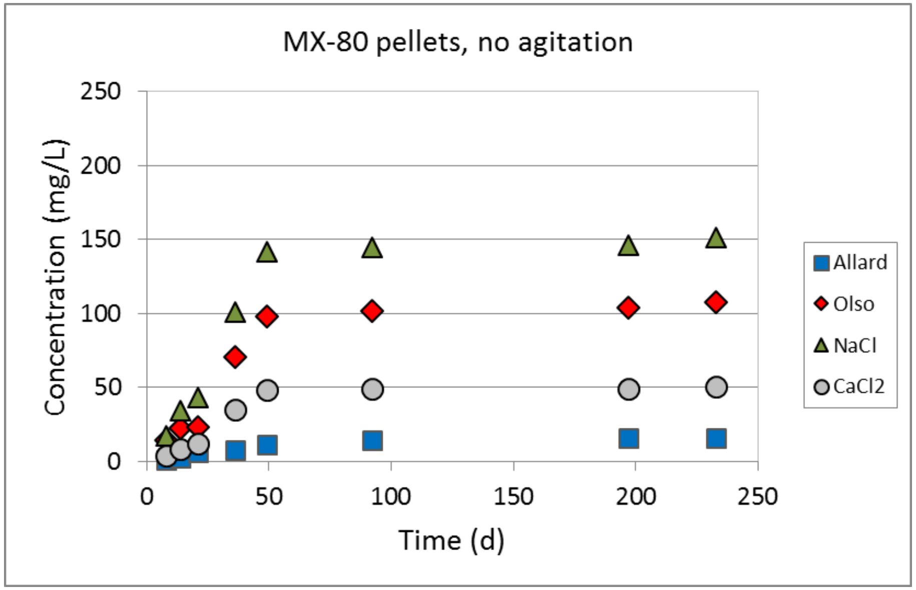
- The release and stability of colloids have been followed from earlier made MX-80 bentonite powder samples.
- Diluted OLSO reference groundwater, NaCl and $CaCl_2$ solutions (I = 0.001 - 0.1 M)
- Particle size distribution and zeta potential
  - Photon correlation spectroscopy (PCS)
- Colloid concentration
  - A standard series made from MX-80 bentonite applying the PCS measurement count rate
  - Al determination (ICP-MS)

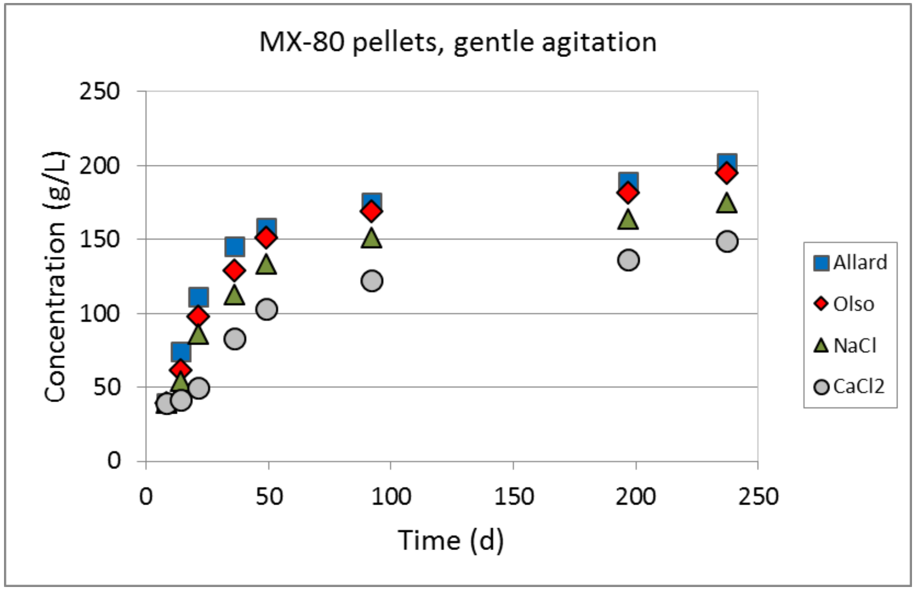
Formation and stability of bentonite colloids





Formation of bentonite colloids





Estimated particle concentration of colloids formed from bentonite pellets in 4.2 mM Allard and 1 mM OLSO, NaCl and $CaCl_2$ solutions without agitation with gentle agitation.

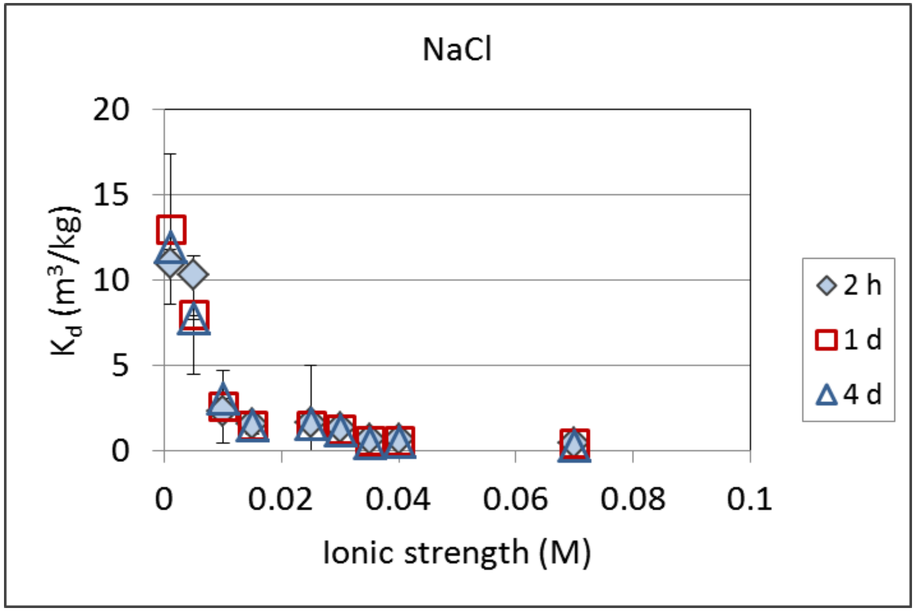
Radionuclide sorption

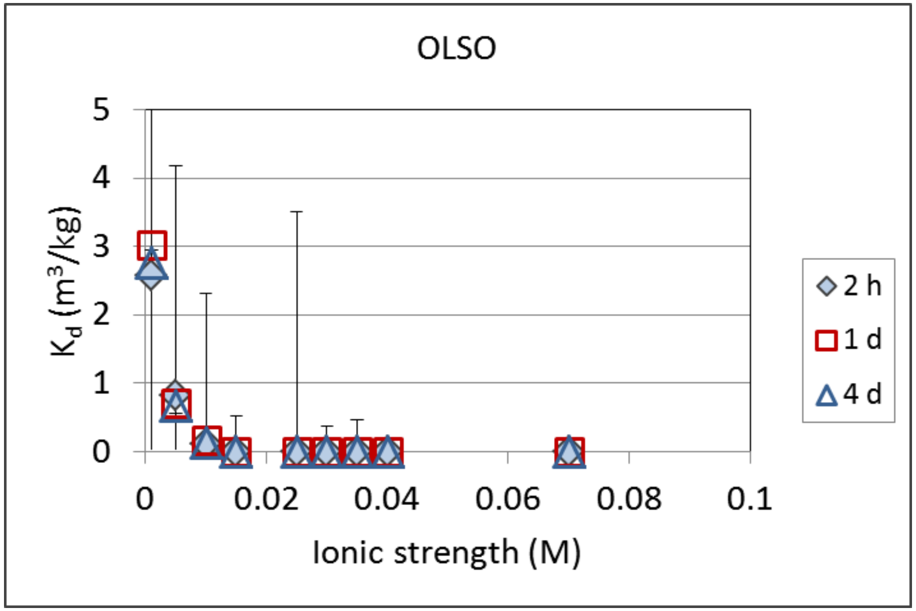
- MX-80 bentonite colloid suspension
  - 4.9 g/L or 2.4 g/L
  - colloid size 270 – 530 nm
- Allard, diluted OLSO, NaCl or $CaCl_2$
- I = 0.001–0.1 M, pH 2-11
- 2 mL colloid suspension + 90 mL solution + ^{85}Sr or ^{152}Eu → 4.7 mL aliquot after 2 h, 1 d, 4 d and 7 d → Ultracentrifugation (90000 rpm/ 60 min) → Radioactivity and particle size measurements
- Ionic strength samples in an ambient air
- pH samples in a clove box under CO_2 free conditions

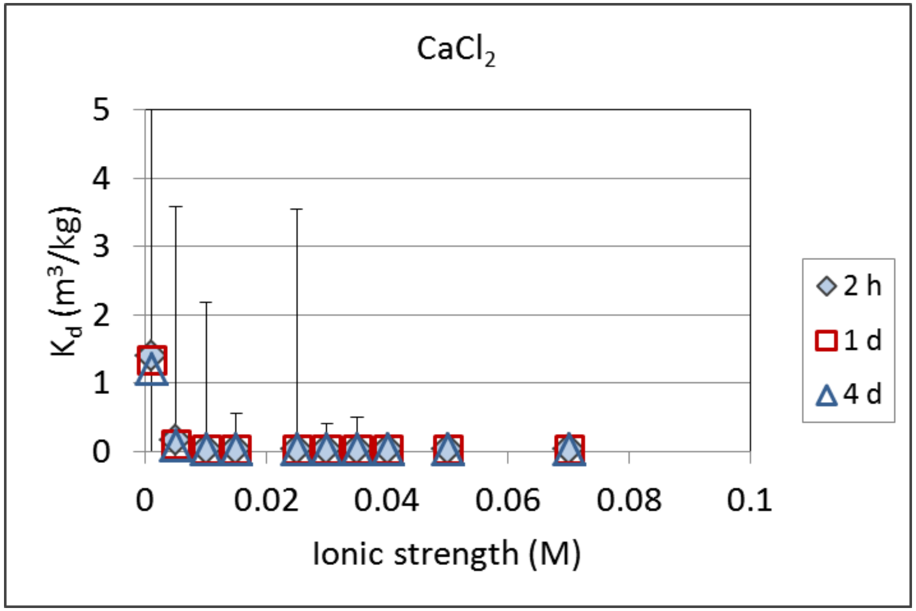
Sr-85 sorption on bentonite colloids

Nearly all of ^{85}Sr was sorbed onto bentonite colloids in 0.001 M solutions.

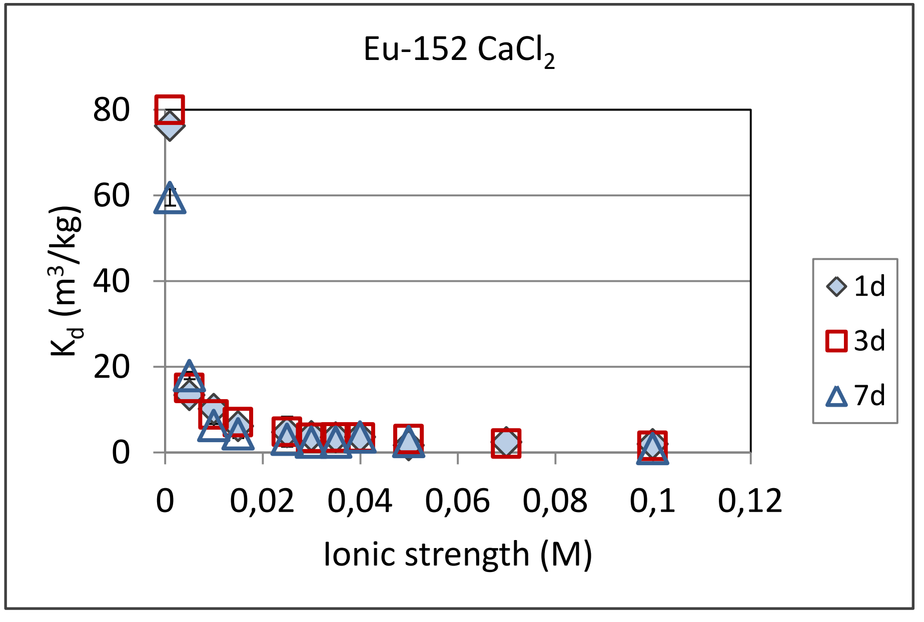
The distribution coefficient (K_d) decreased when the ionic strength increased.

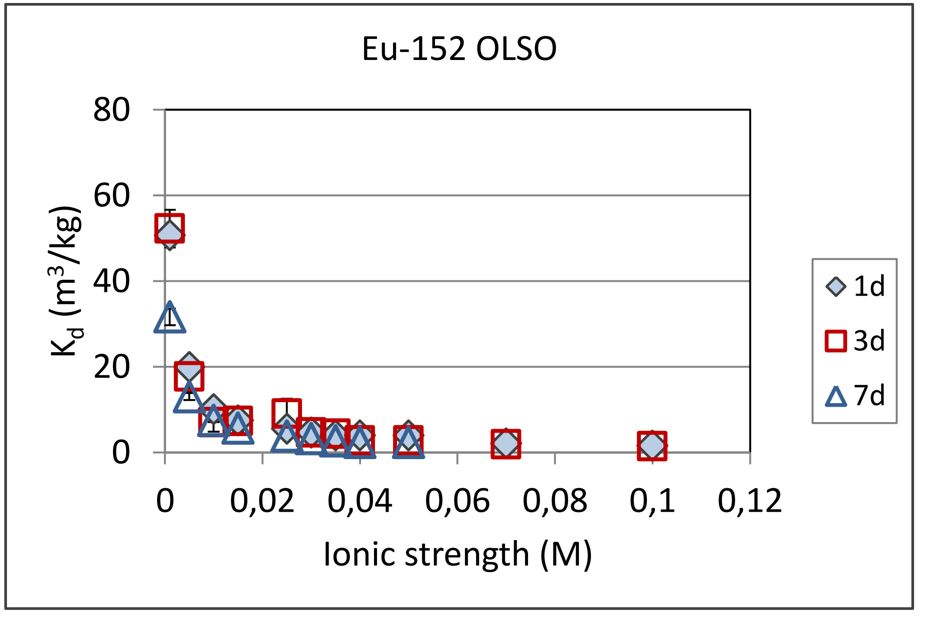




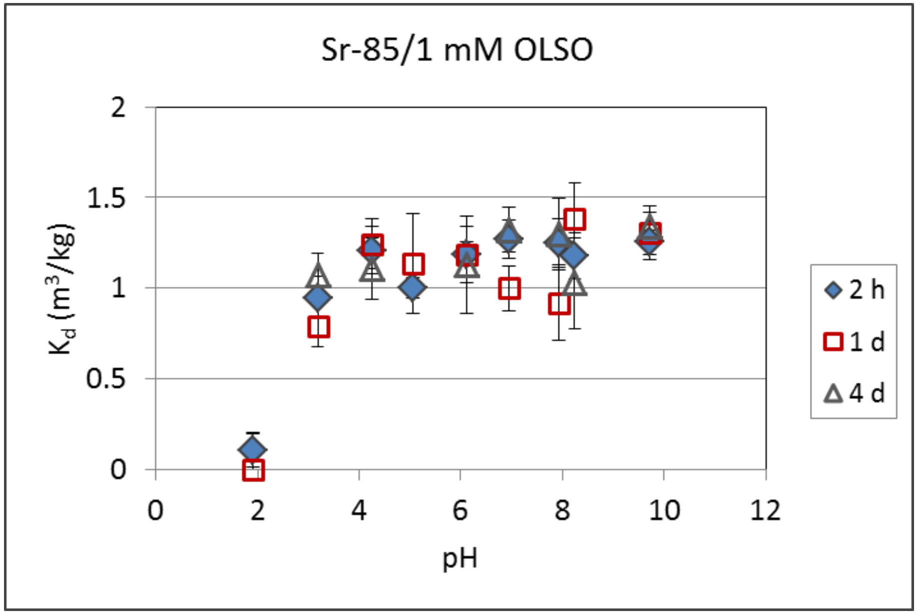


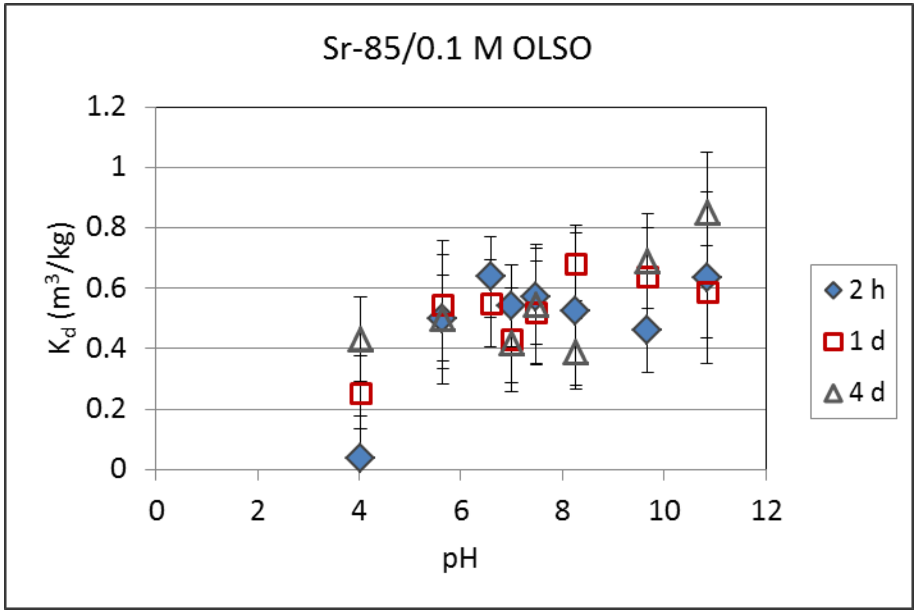
Eu-152 sorption on bentonite colloids



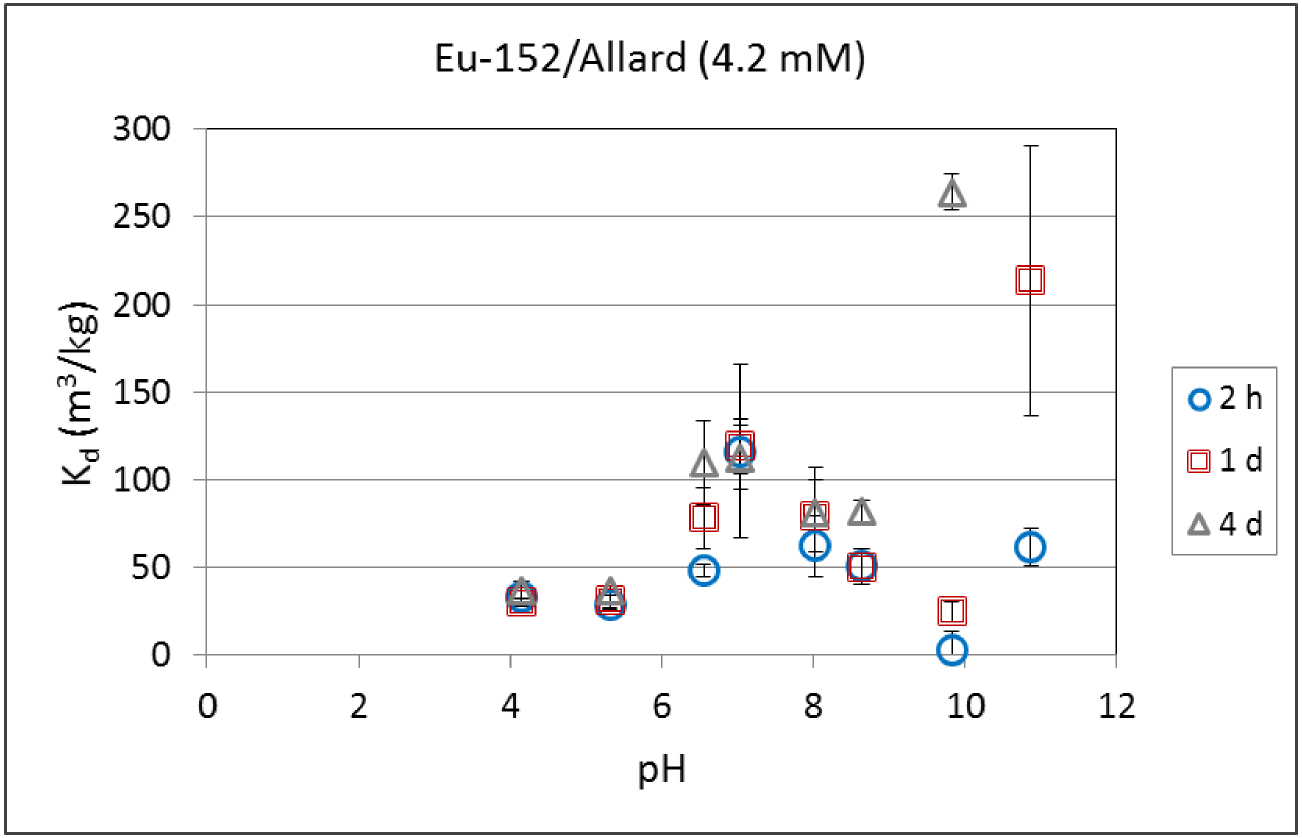


Sr-85 sorption on bentonite colloids





Eu-152 sorption on bentonite colloids



Np-237 sorption on corundum, montmorillonite and colloids

Batch experiments

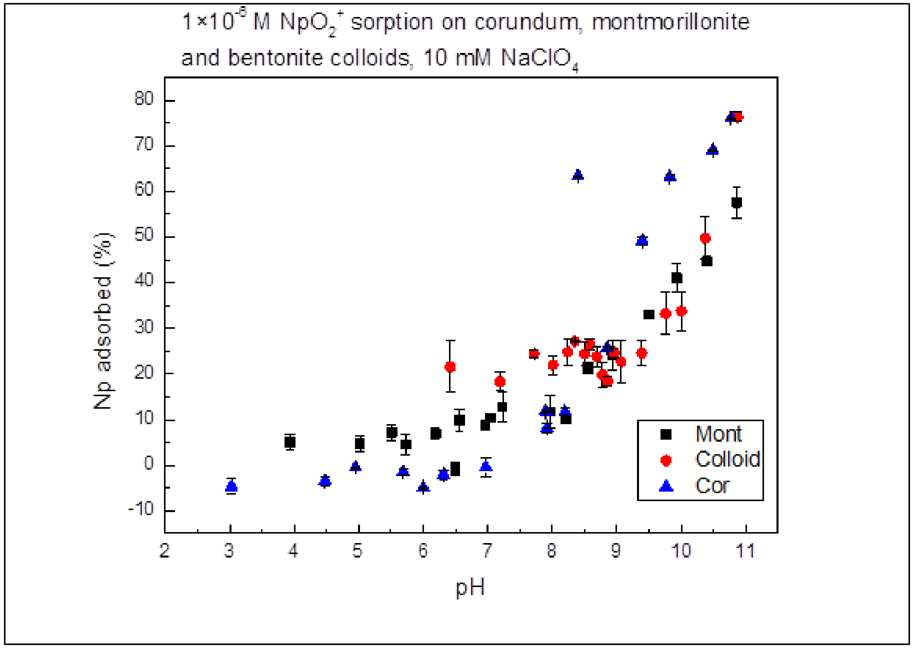
- Under anoxic conditions
- 10 mM $NaClO_4$ (batch)
- 10 mM NaCl (IR, ζ-potential)
- pH edges
- Np-237(V) sorption isotherms
- effect of time and the amount of mineral concentration

In-situ ATR FT-IR measurements in HZDR

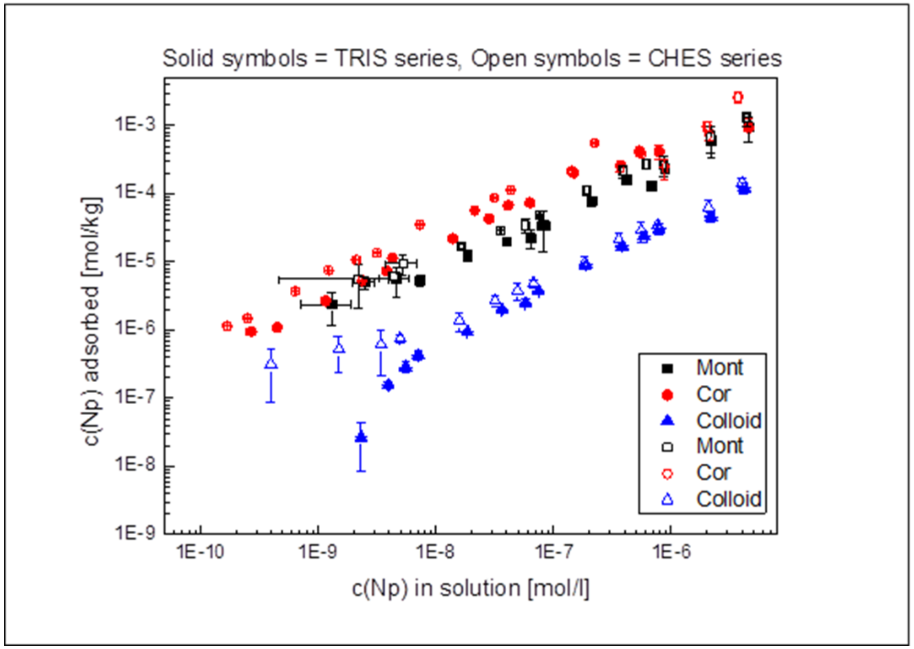
(attenuated total reflection Fourier transform infrared spectroscopy)

EXAFS

The neptunium(V) adsorption onto corundum, montmorillonite and bentonite colloids

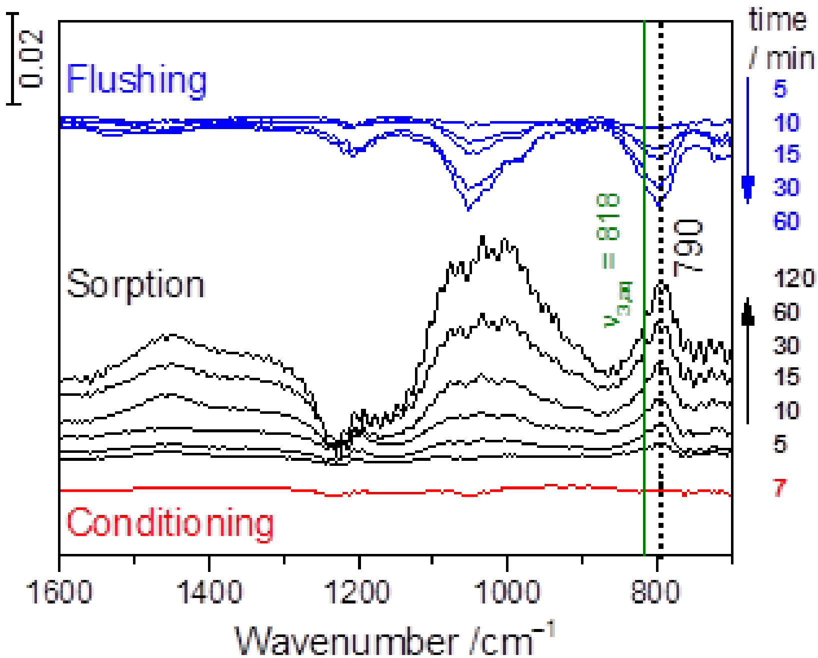


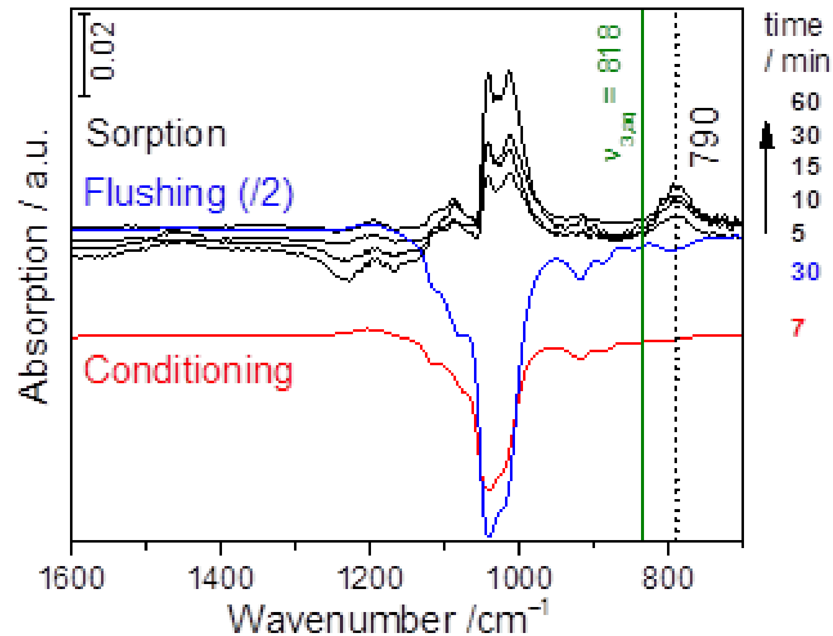
pH - edge



The sorption isotherms in TRIS and CHES buffer solution

ATR FT-IR measurements of Np(V) sorption





Sorption of 50 µM Np(V) in 0,01 M NaCl in D_2O on corundum at pD 9,6, flow rate 0,1 mL/min.

Sorption of 50 µM Np(V) in 0,01 M NaCl in D_2O on montmorillonite at pD 9,6, flow rate 0,1 mL/min.

Column experiments

<u>Crushed rock columns:</u>

Kuru grey granite and Sievi strongly altered tonalite

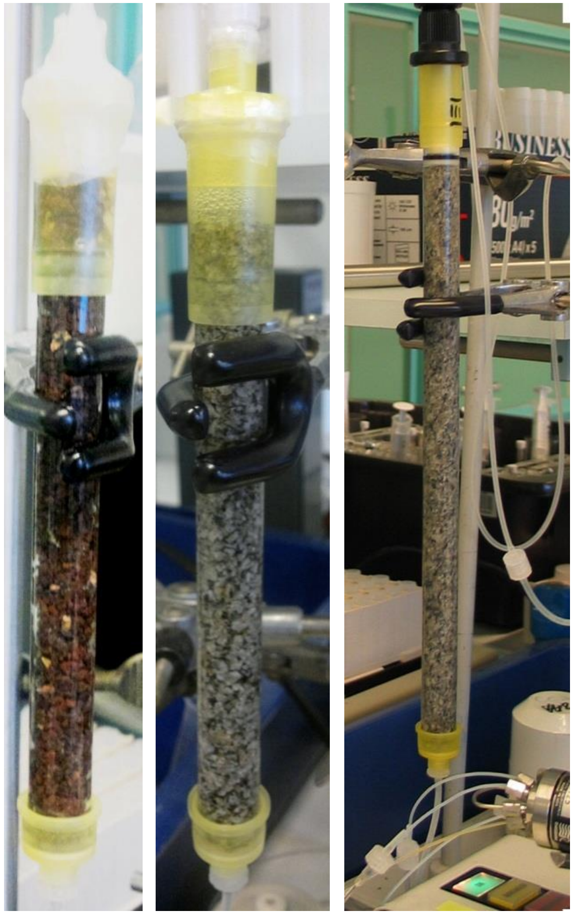
L = 15 cm or 30 cm, i.d. = 1.5 cm

<u>Drill core column</u>:

Kuru grey granite, core placed inside a tube

Flow channel in a 0.5 mm gap between the core and the tube

L = 68.5 cm, w = 4.4 cm

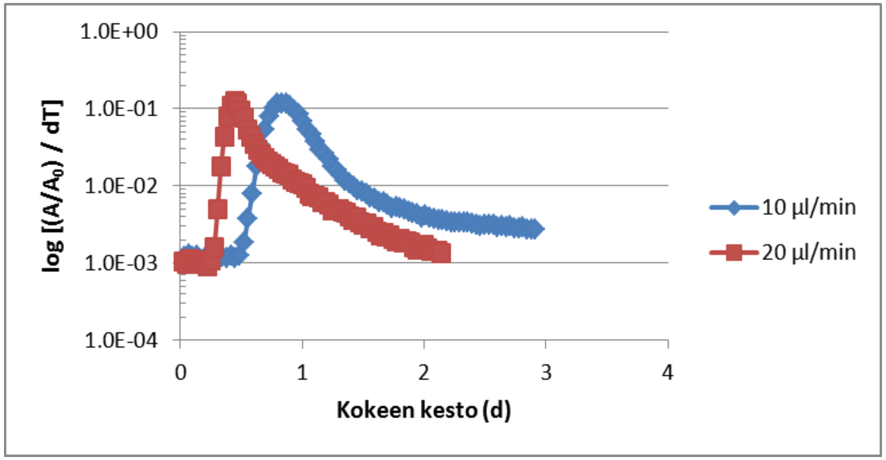


Column experiments

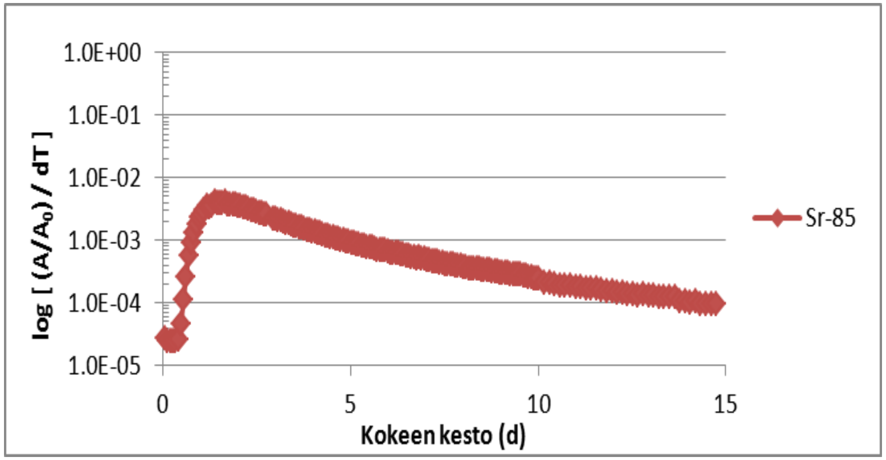
- The hydraulic properties determined using non-sorbing tracers (^{36}Cl, ^{125}I) without colloids
- Bentonite colloid suspension
- Allard reference ground water
- ^{85}Sr and ^{152}Eu transport with and without colloids
- Colloid migration experiments

Kuru grey granite crushed rock column (30 cm)

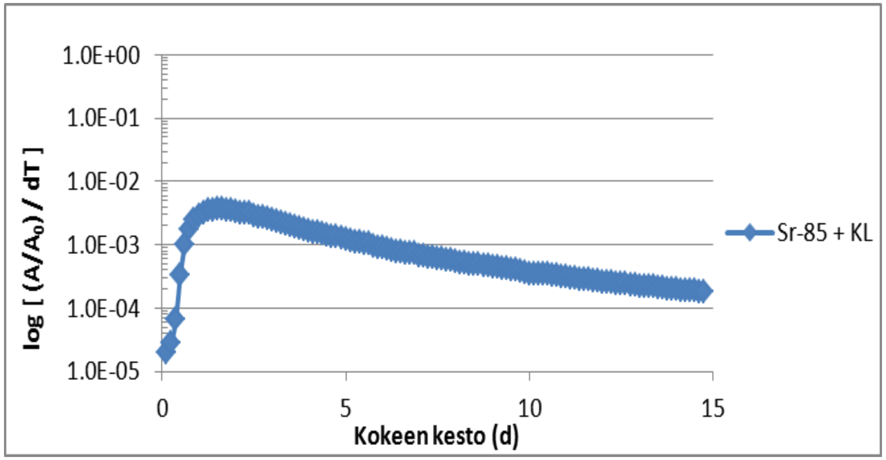
Breakthrough curves of ^{125}I



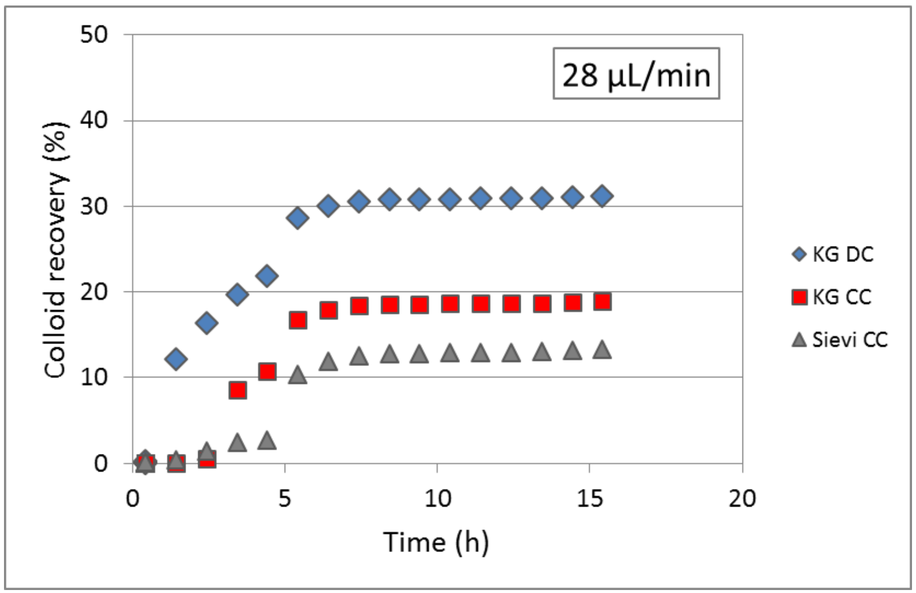
Breakthrough curve of ^{85}Sr without bentonite colloids

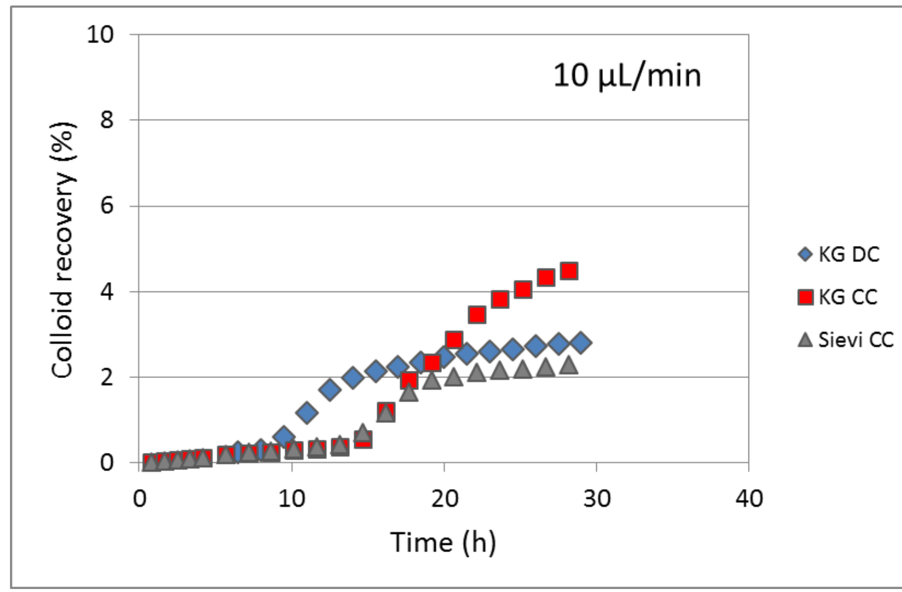


Breakthrough curve of ^{85}Sr with bentonite colloids



Colloid mobility





Kuru grey granite drill core column (30 cm) (blue), crushed rock columns (15 cm): Kuru grey (red) and Sievi altered tonalite (grey), flow rate 28 and 10 ul/min, particle size 230 nm

Conclusions

The stability of bentonite colloids depended strongly on the ionic strength and the valence of the cation.

The colloid dispersion has remained stable in low salinity solutions so far for near four years.

The colloid formation was significantly increased with the slow agitation.

pH and ionic strength have a great influence on the chemical form of the radionuclides, especially actinides and thus their adsorption behavior.

Colloid recovery in column experiments was low.

Migration of colloids was affected by water flow rate, column type and colloid size.

Promising results from Np(V) sorption experiments,

Future work

- The solid liquid–ratio to obtain an optimum mineral concentration needed
- The kinetic experiments to optimize the equilibration time for the batch experiments
- The Zeta potential of the system as a function of pH with and without a radionuclide to provide information about the adsorption mechanisms (outer-sphere or inner-sphere complexation)
- Desorption experiments
- The sorption of neptunium(V) on montmorillonite (Nanocor) and bentonite colloids
- The effect of bentonite colloids on neptunium(V) transport in the column experiments

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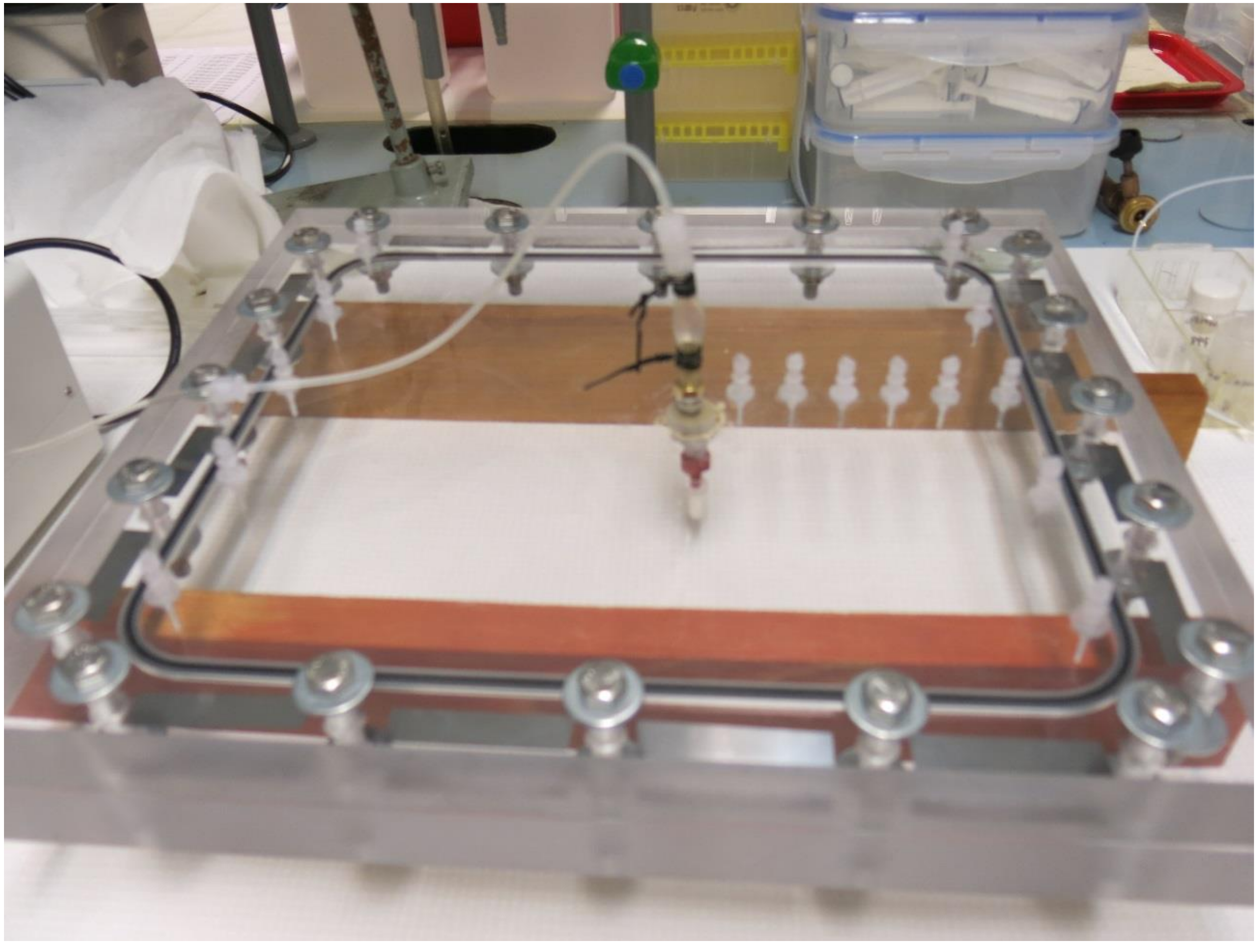
Thank you!



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