U.S. DEPARTMENT OF ENERGY | Office of NUCLEAR ENERGY

Spent Fuel and Waste Science and Technology (SFWST)

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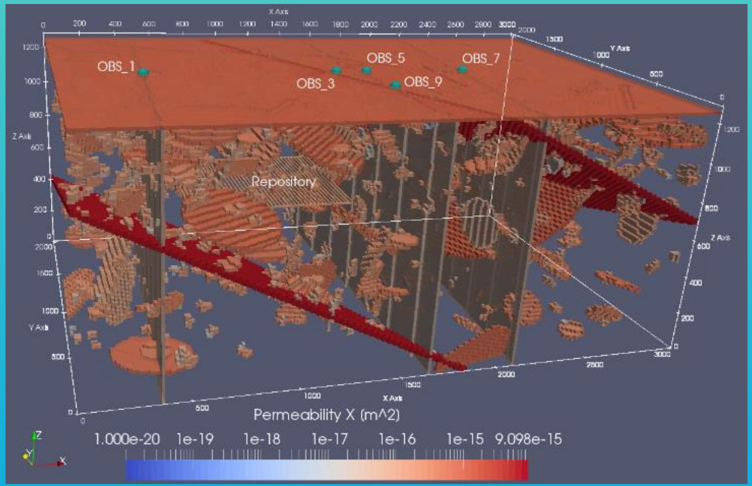

Buffer Erosion, Coagulation/Flocculation and Clogging

U.S. Nuclear Waste Technical Review Board
Summer 2024 Fact Finding Meeting
April 23, 2024
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SAND2024-07144O

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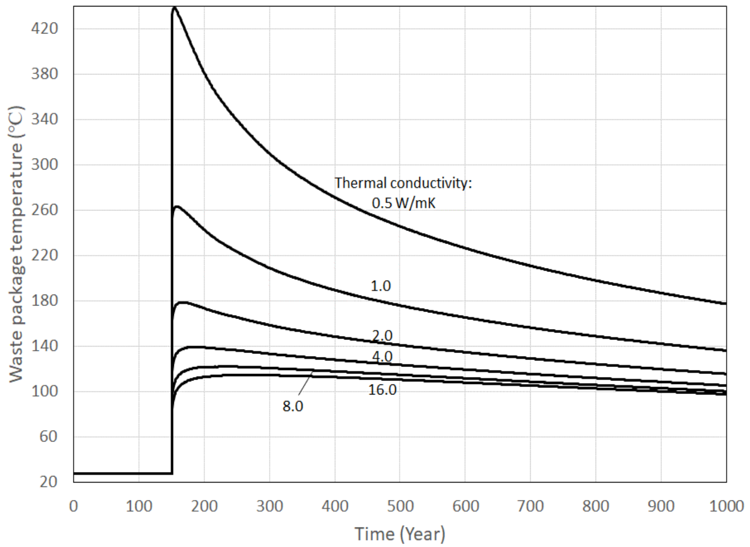
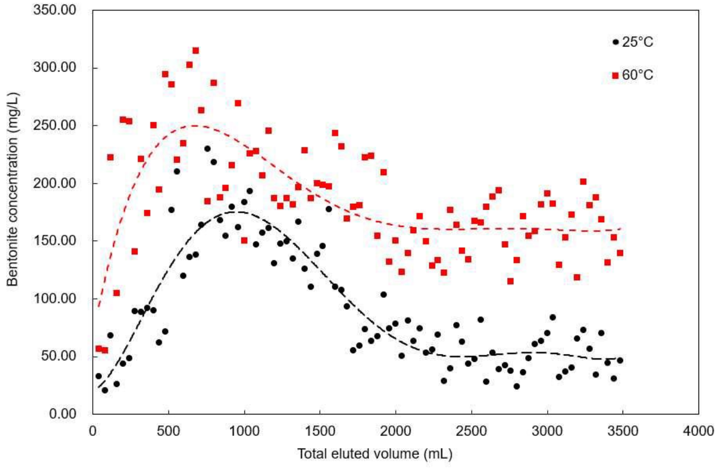
Buffer Stability, Erosion, and Clogging: Temperature Effects

SFWST focus areas:

- Thermal stability and limit of buffer materials
  - Understanding smectite-to-illite transformation
- Buffer materials for extreme environments
  - Saponite as an alternative buffer material
- Buffer erosion and fracture clogging
  - Microfluidic cell experiments
  - Bentonite swelling, extrusion and fracture clogging

Note: EURAD-HITEC aims to develop and document improved THM understanding of clay based materials (host rock and buffer) exposed to elevated temperatures (>100°C) for extended durations.



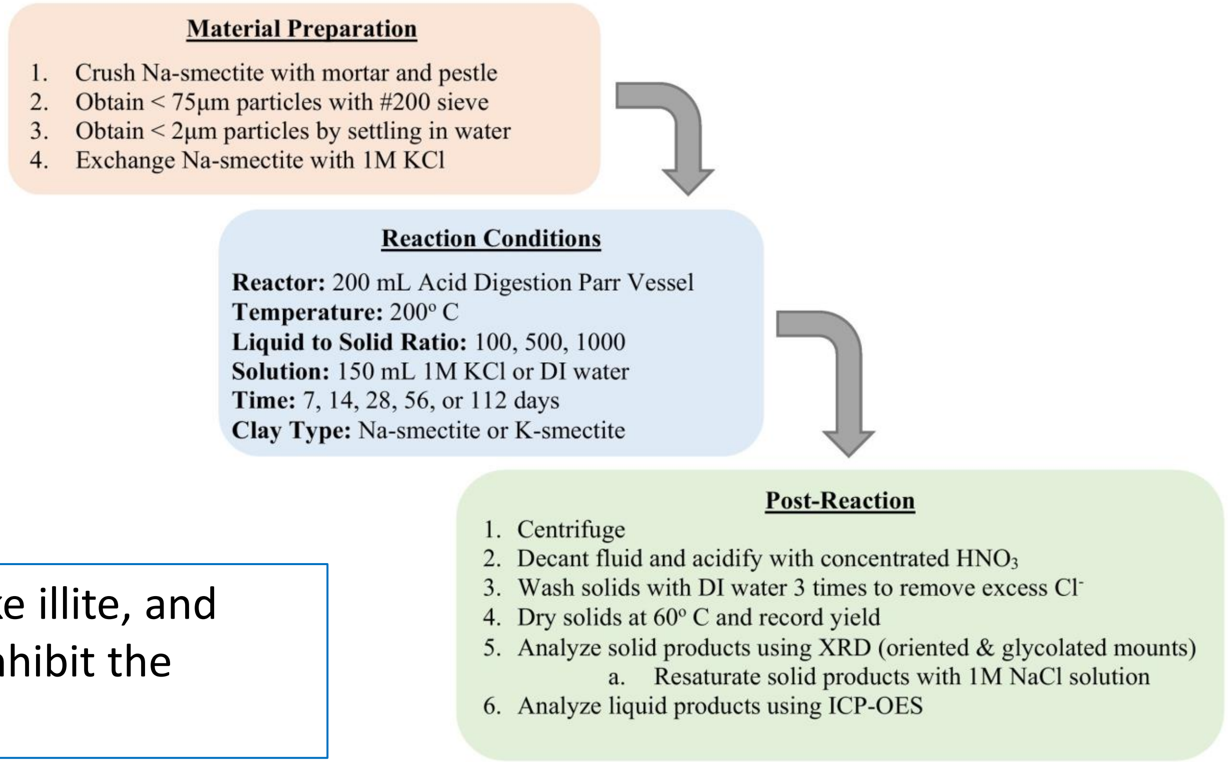


Sedighi et al. (2023)

Understanding Smectite-to-Illite Transformation

Potential controlling factors:

- Temperature
- Water/solid ratio (SiO_2 concentration)
- Duration
- Cations (Na, K)
- Solution chemistry (DI, KCl solution)

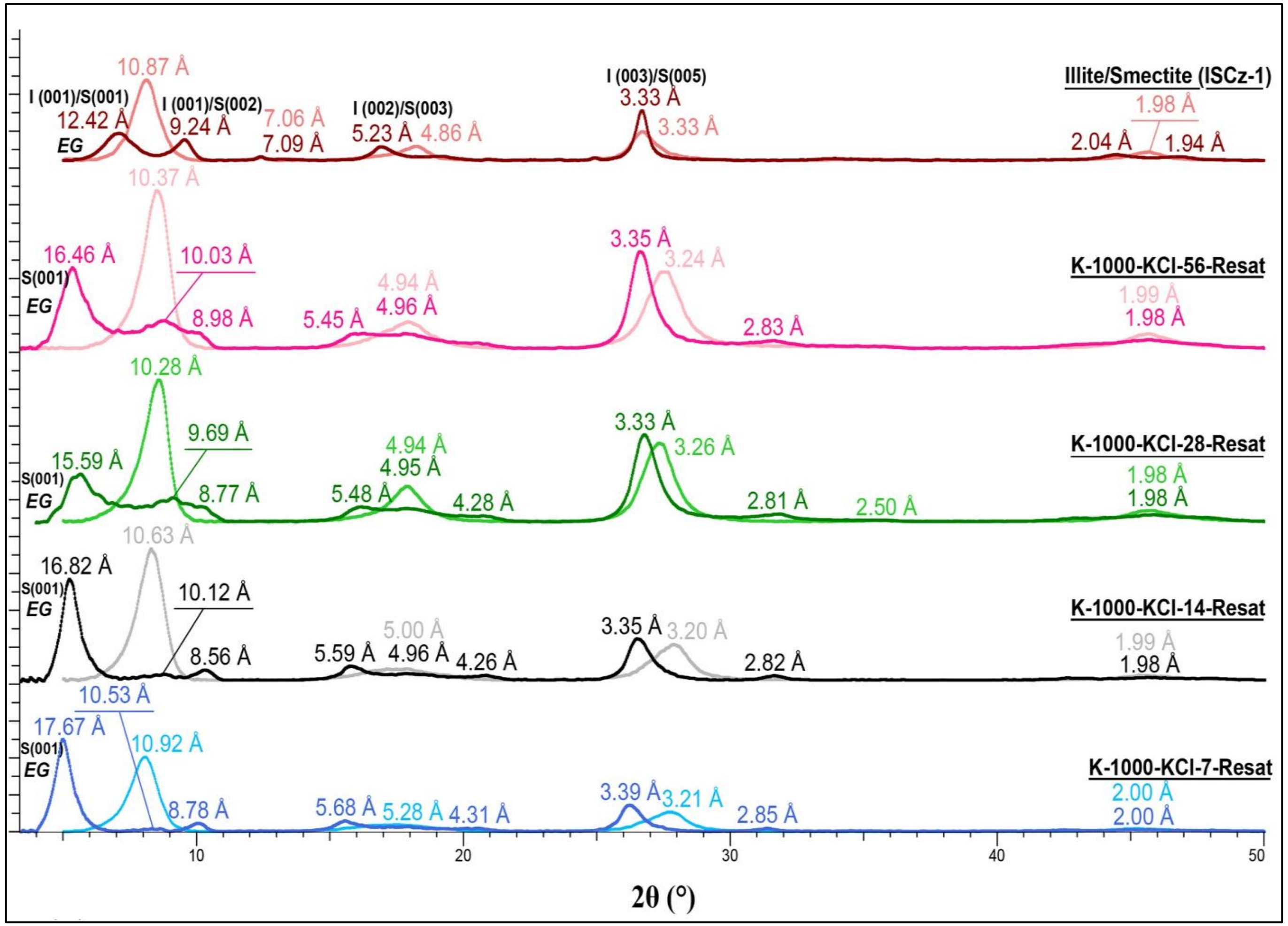


Approach: (1) Create optimal conditions to make illite, and then (2) find out which factors can effectively inhibit the transformation.

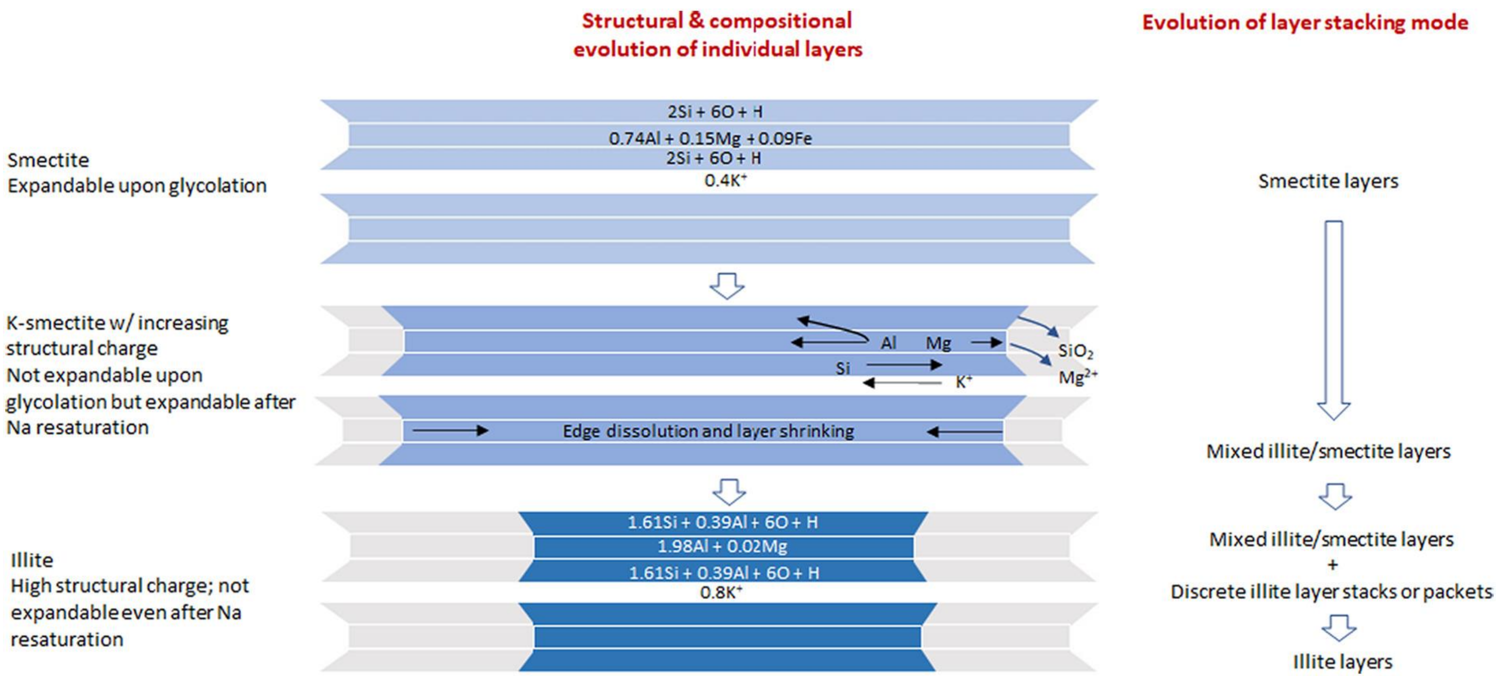
$$2Na_{0.4}(Al_{1.47}Mg_{0.29}Fe_{0.18})Si_4O_{10}(OH)_2 \text{ (Na-smectite)} + 0.85K^+ + 1.07H^+ \rightarrow$$
$$1.065K_{0.8}(Al_{1.98}Mg_{0.02})(Si_{3.22}Al_{0.78})O_{10}(OH)_2\text{(illite)} + 4.6SiO_2 + 0.36Fe(OH)_3 + 0.56Mg^{2+} + 0.8Na^+ + 0.9H_2O$$

Findings

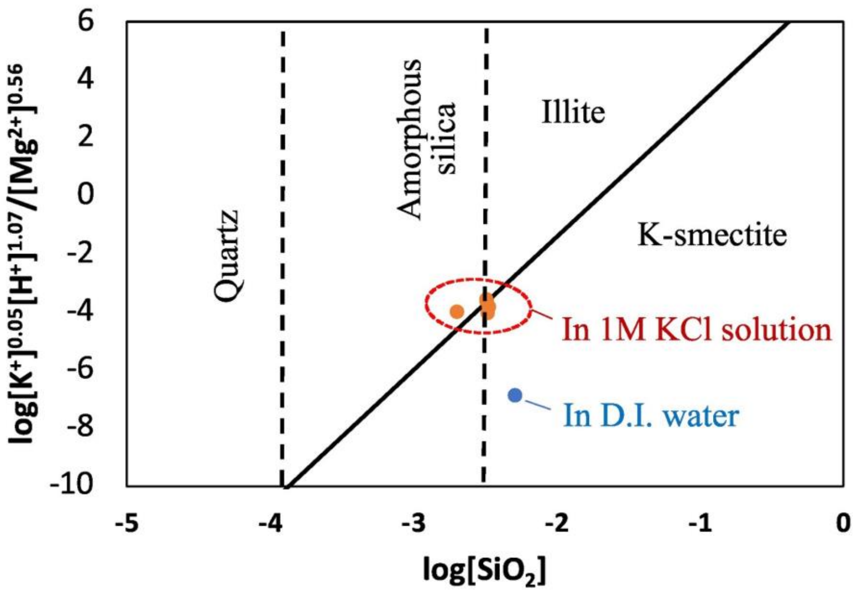
- Under optimal conditions, smectite-to-illite transformation can happen relatively fast (on a time scale of weeks).
- The transformation requires an external K^+ source. K-exchanged smectite in DI water is still deficient in K^+ to convert smectite to illite at 200 °C.
- Water/solid ratio (and thus dissolved SiO_2 concentration) is an important factor controlling the extent of the transformation.
- The required optimal conditions can hardly be realized in an actual barrier system.



Mechanism and Stability Field



Layer-by-layer transformation

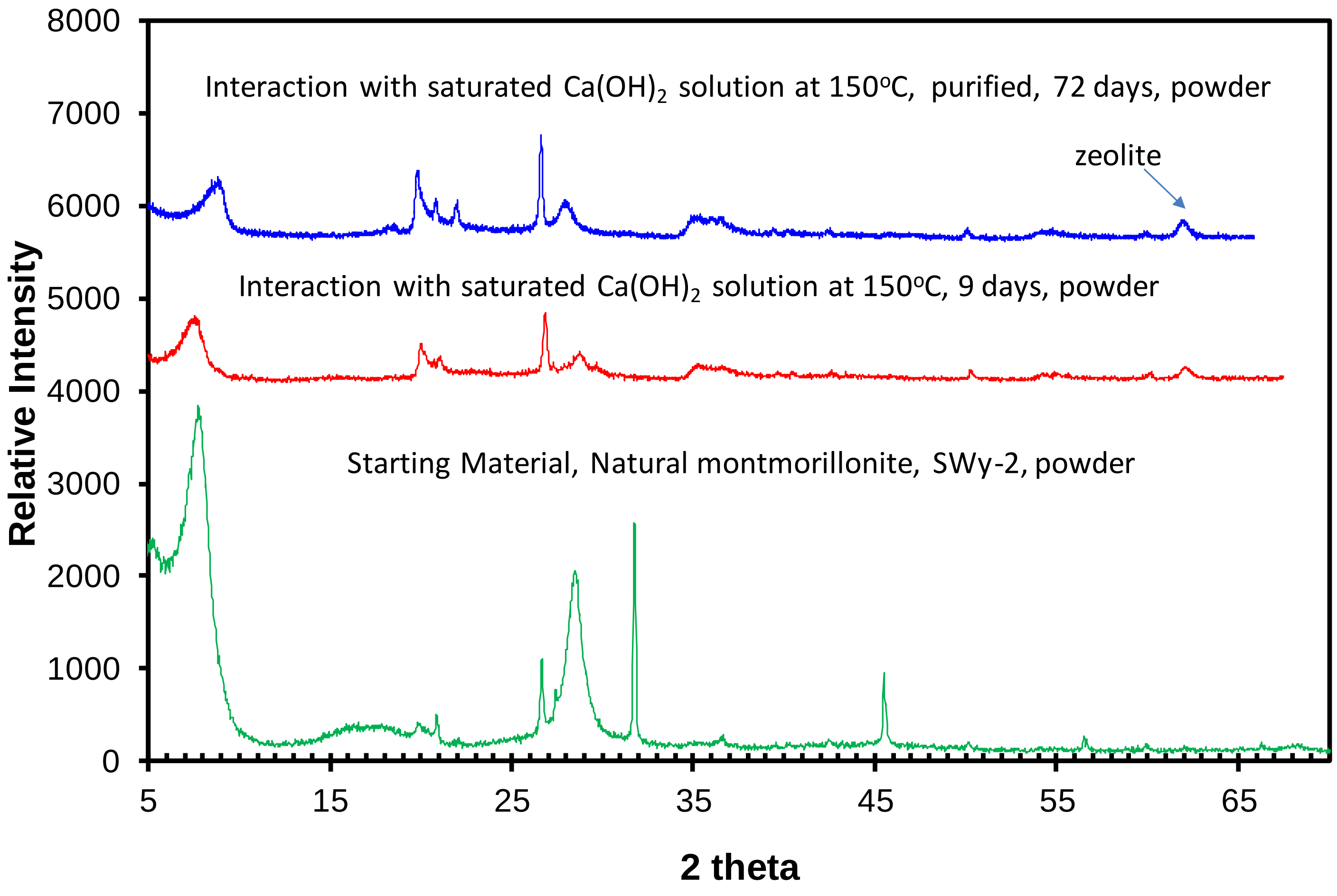


Chemical additives to inhibit smectite-to-illite transformation:

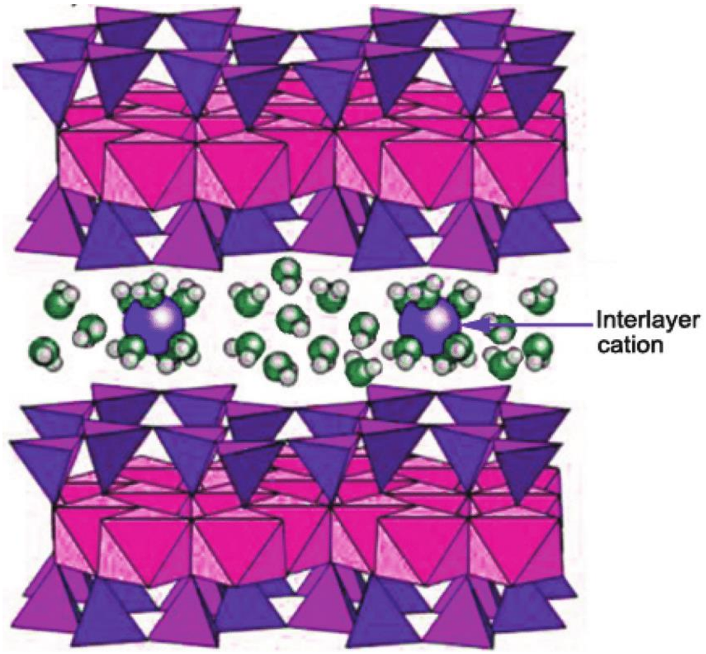
$Mg(OH)_2$ (brucite) → $Mg^{2+} + OH^-$

SiO_2 (am) → SiO_2 (aq)

Stability of Montmorillonite

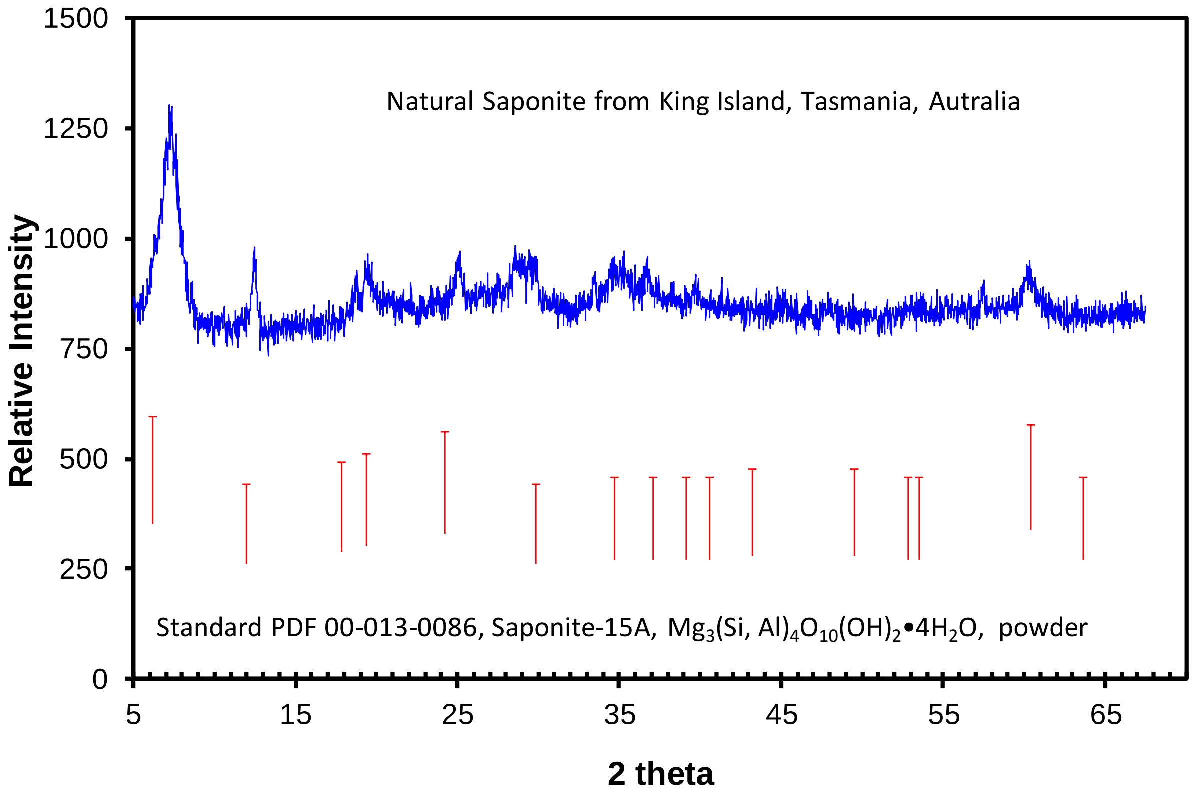


Saponite: Trioctahedral Mineral of Smectite Group

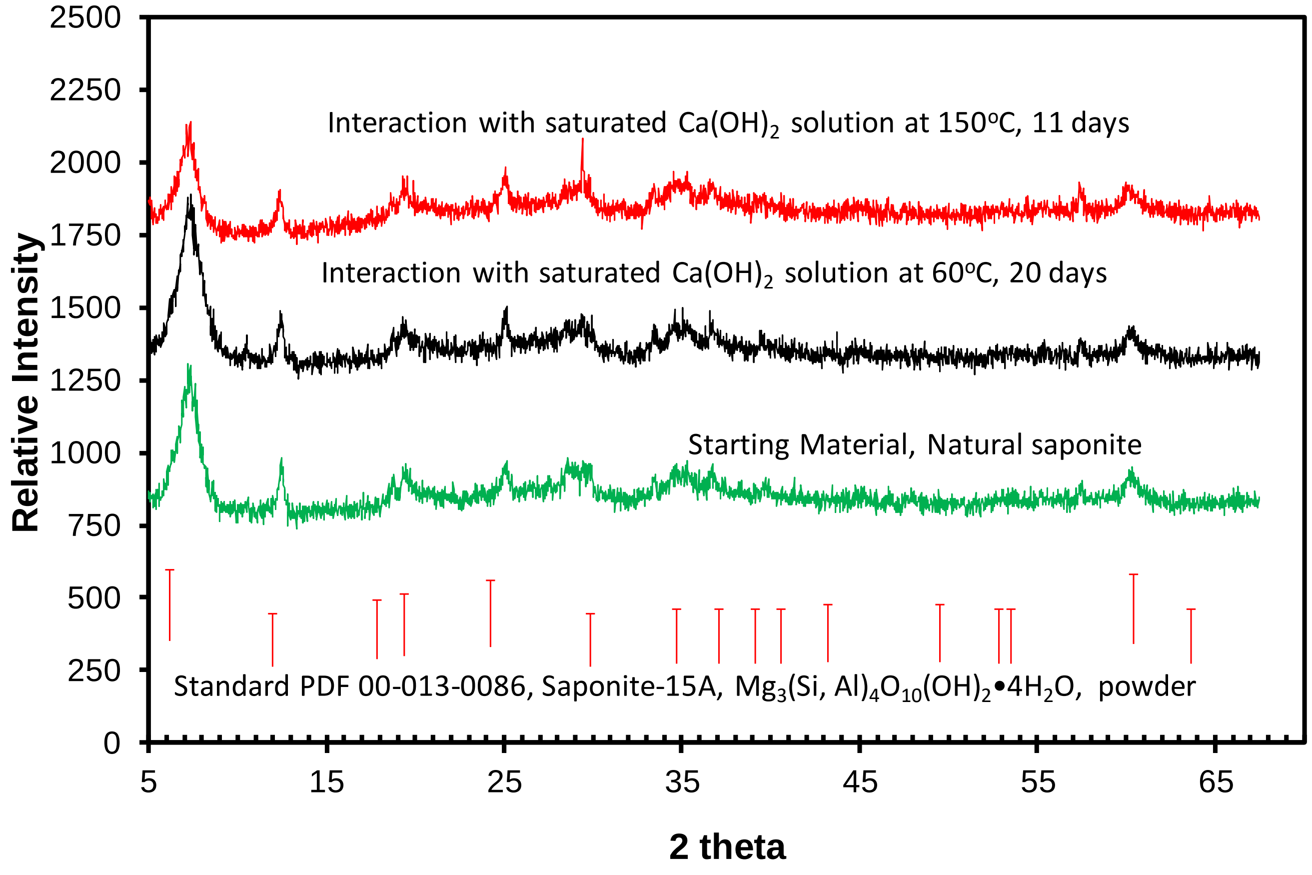


$Na_{0.95}Mg_{5.90}[(Si_{7.07}Al_{0.99})O_{20}](OH)_4$

Mitra et al. (2013)



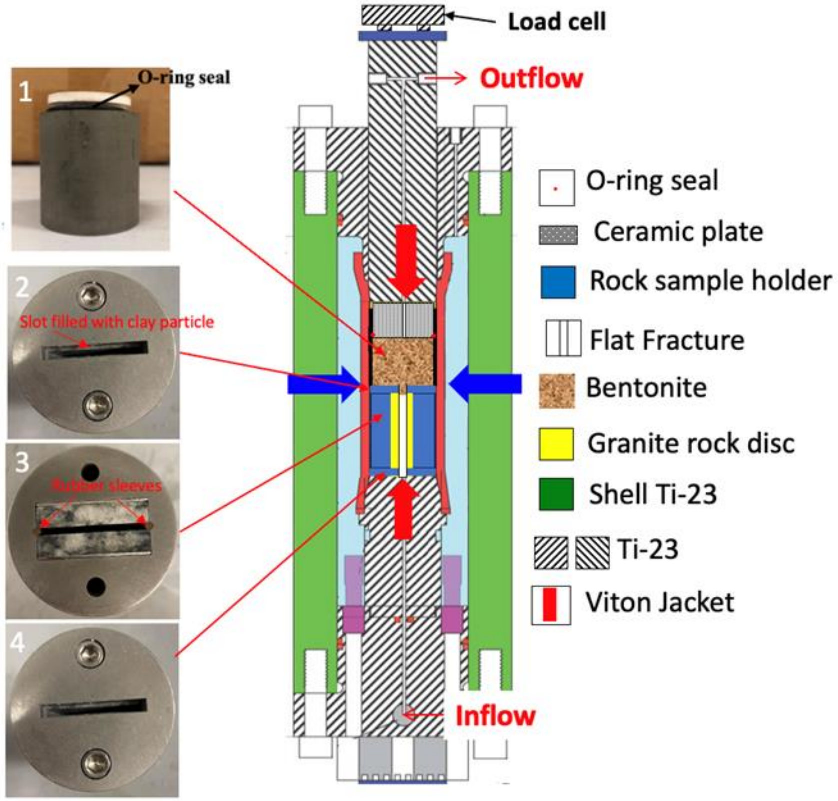
Saponite as a Buffer Material for Harsh Disposal Environments

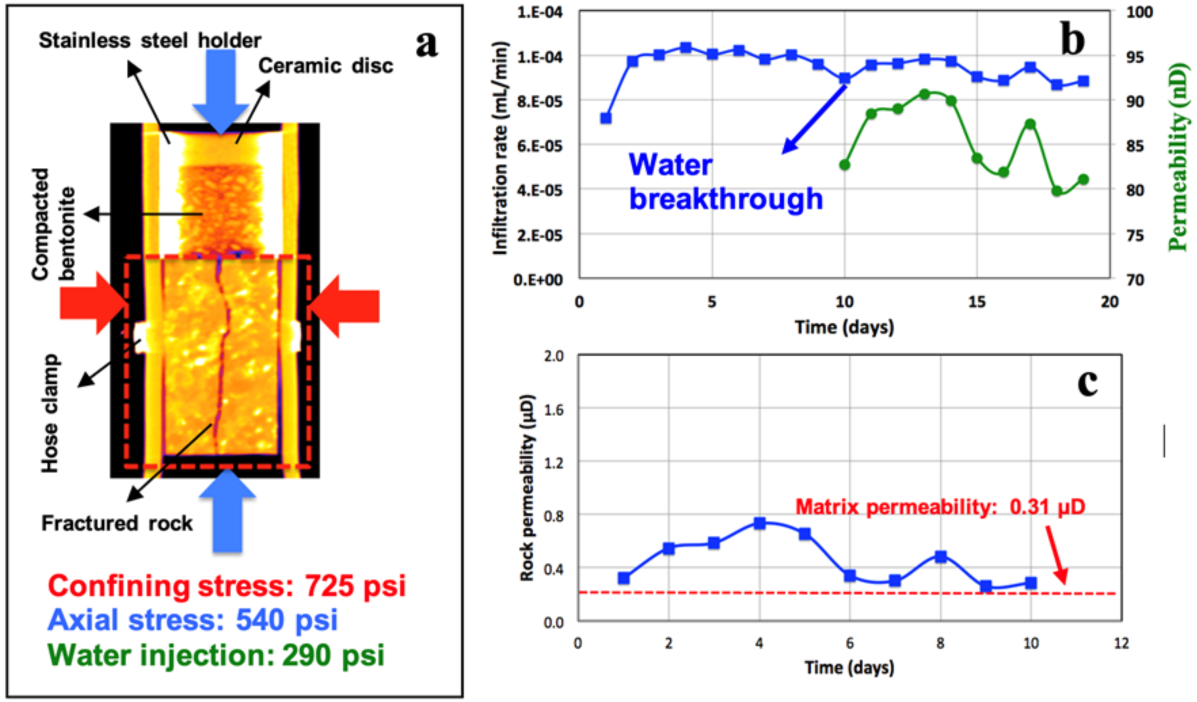


Samples	Density kg/m^3	Dry density kg/m^3	Hydraulic conductivity (K), m/s	Swelling pressure (p_s), kPa
MX-80	1800	1310	E-10	200
MX-80	2000	1175	2E-13	4700
GMZ	1788	1233	E-11	530
Saponite	1800	1175	4E-12	1300
Mixed-layer FIM	1800	1392	4E-11	280
Mixed-layer FIM	2000	1175	2E-11	1000
Mixed-layer Holmehus	1800	1310	2E-11	600
Mixed-layer Holmehus	2000	1175	8E-12	2000

Yang et al. (2014)

Bentonite Swelling, Extraction, and Fracture Clogging

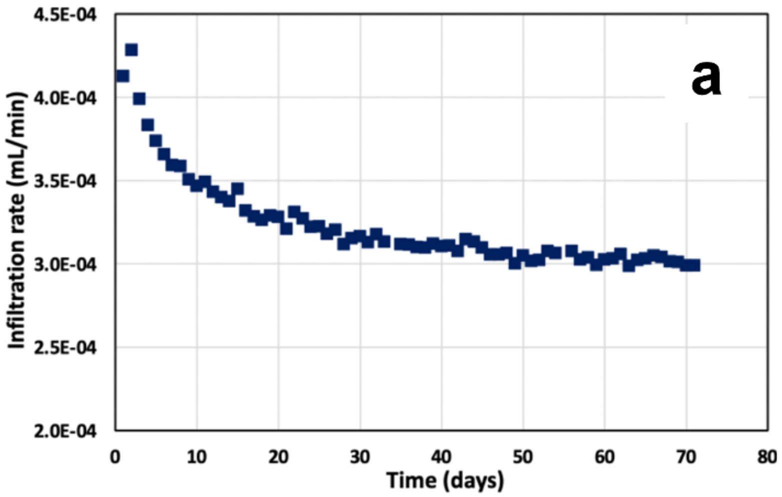


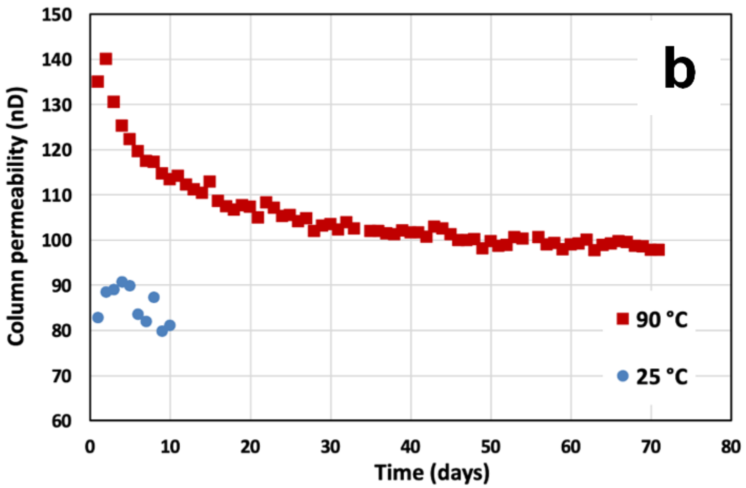


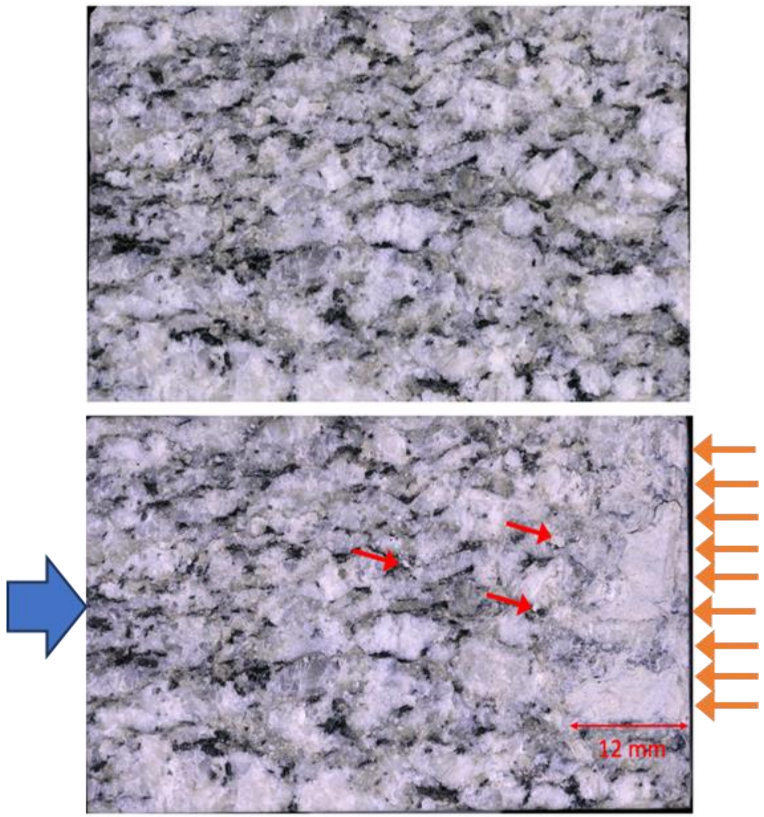
Permeability of the fractured sample without an adjacent bentonite layer: 156 mD, close rock matrix permeability

5 order reduction in fracture permeability by bentonite extrusion and clogging

Bentonite Swelling, Extraction, and Fracture Clogging (cont.)

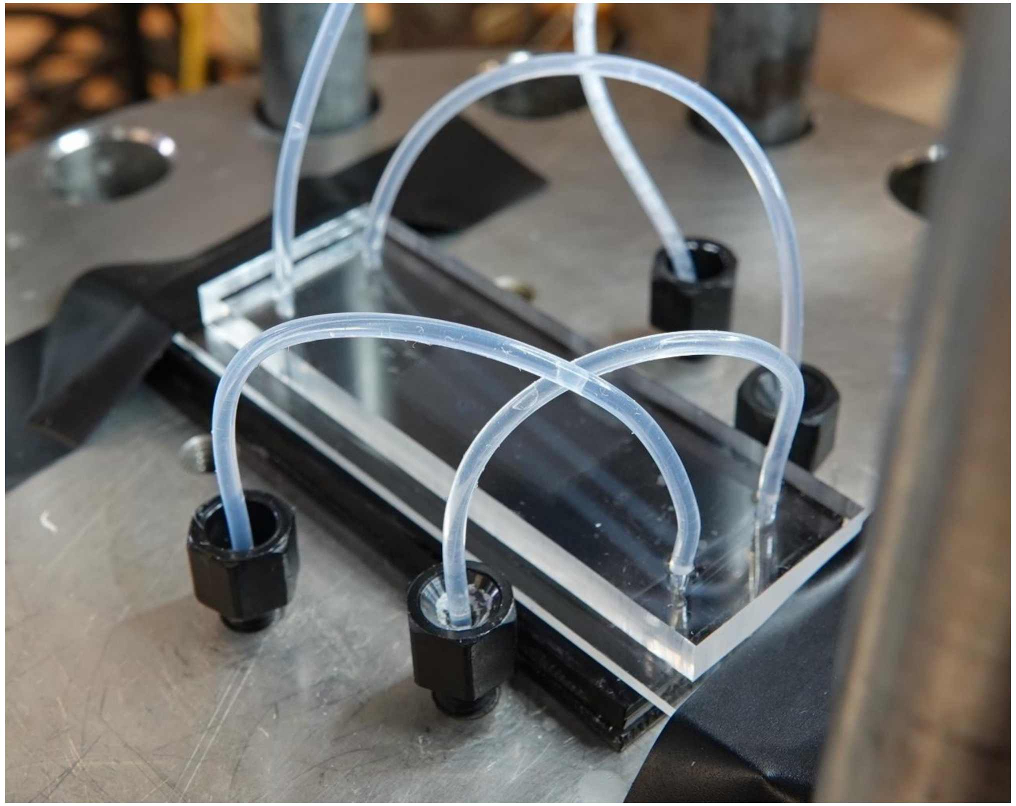


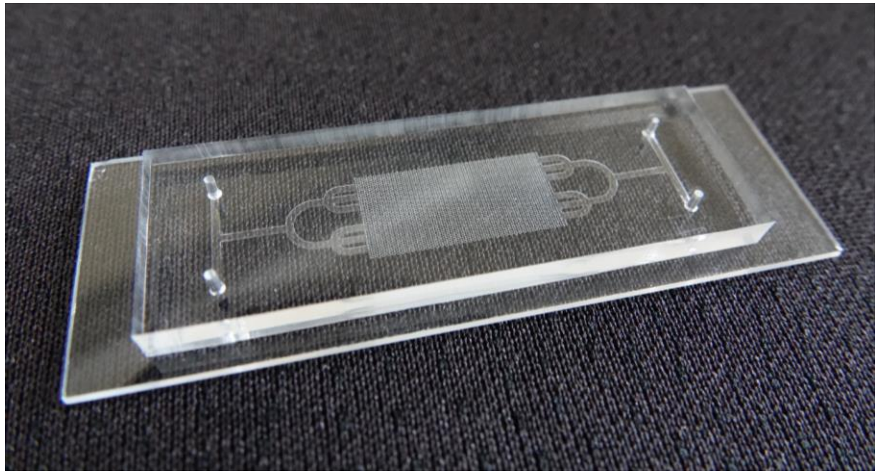


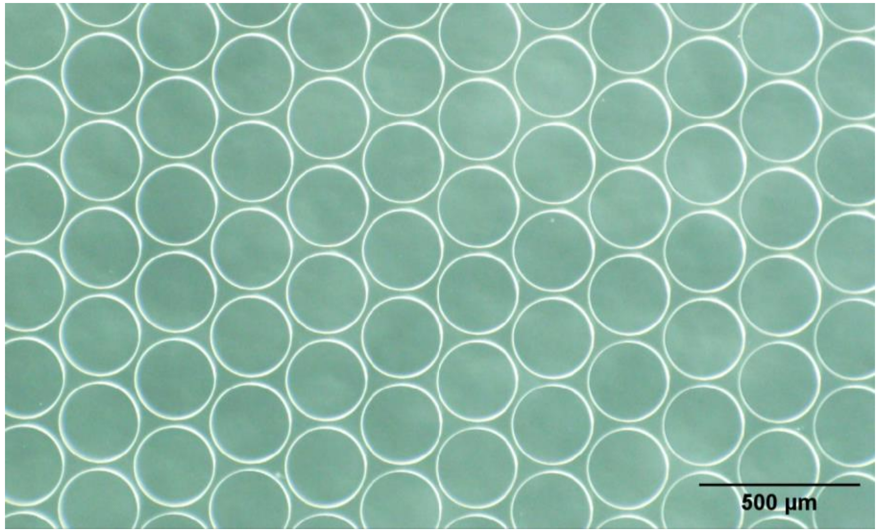


Microfluidic Cell

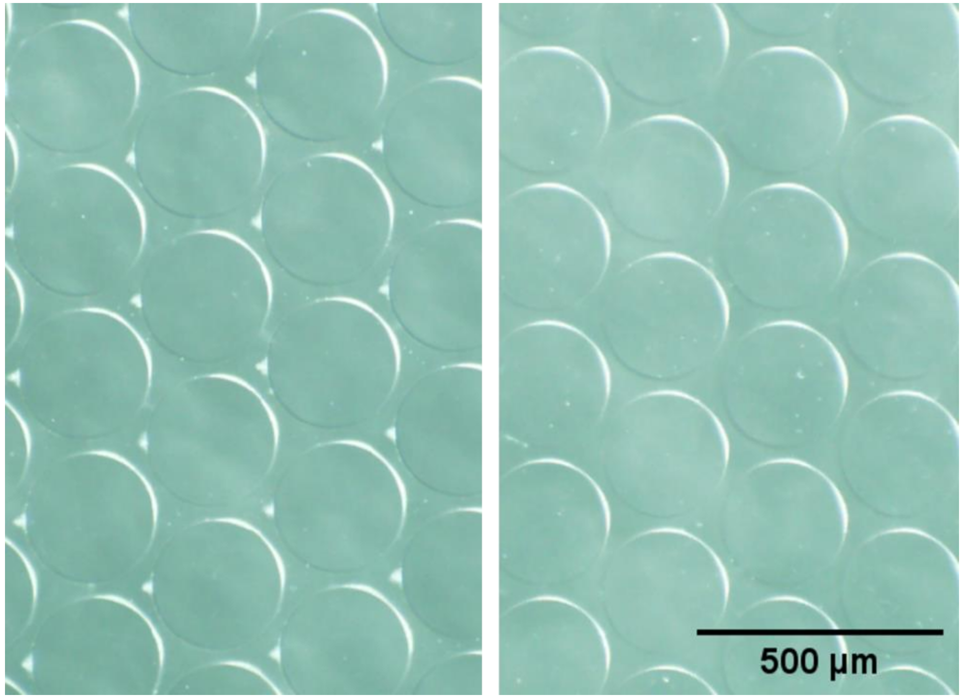
Microfluidic cell for studying bentonite accumulation and clogging in fractures



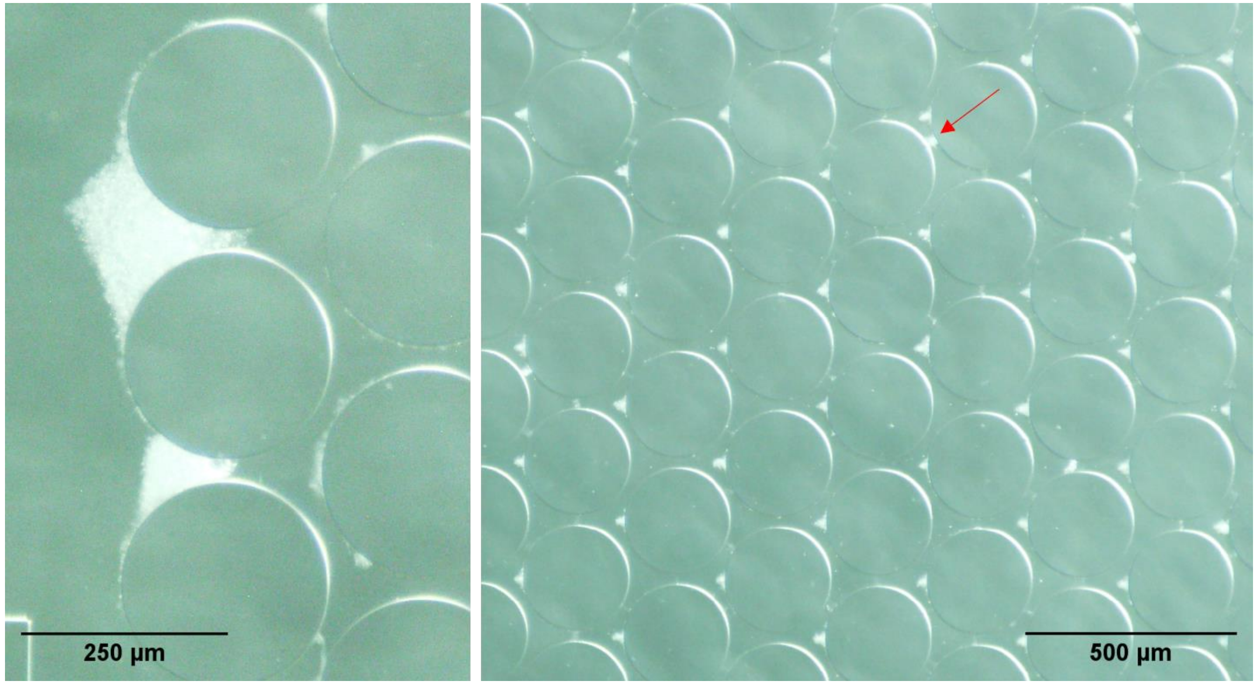




Preliminary Results



Left: The solution containing bentonite with NaCl.
Right: The solution of only bentonite.



Left: A magnified view of the large bentonite clogs in the inlet of a model after flowing the bentonite in 1 M NaCl suspension. Right: Smaller clogs occurring further away from the inlet after 30 minutes of flowing the bentonite-salt solution.