Influences of Temperature on Permeability Changes of Flow-Paths Altered by Highly Alkaline Ca-rich Groundwater

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INTRODUCTION

Fractures in host rock around the geological repository are considered as main flow-paths for the migration of radionuclides. Such fractures would be altered by the alkaline components (Ca, Na, K) leaching from cementitious materials.

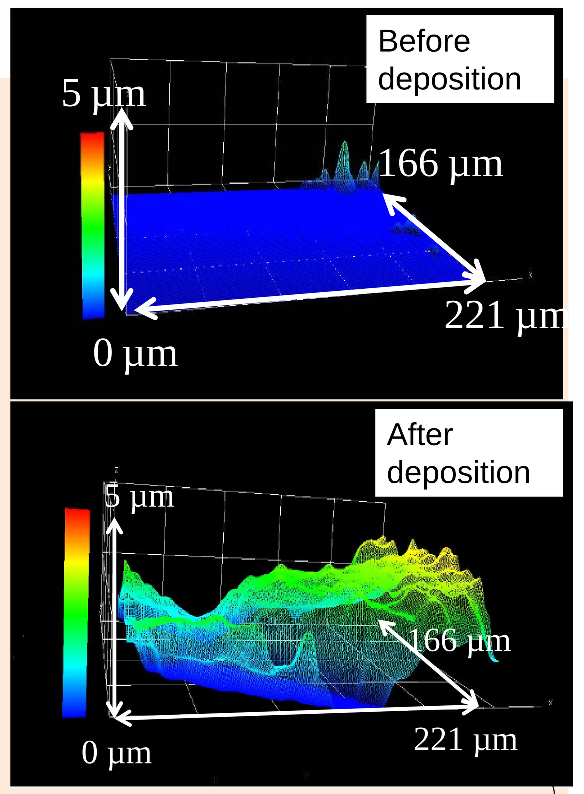
→ Dissolution of silicate minerals & Deposition of secondary minerals

✓**Calcium-silicate-hydrate (CSH-gel) may clog the flow-paths.**

✓**Temperature changes greatly affect the chemical reactions.**

<Authors' previous study (Kurata et al., 2015)>

The formation of CSH-gel and the clogging of micro flow-path under the condition of Ca-rich and high pH were showed by flow experiments (298 K).

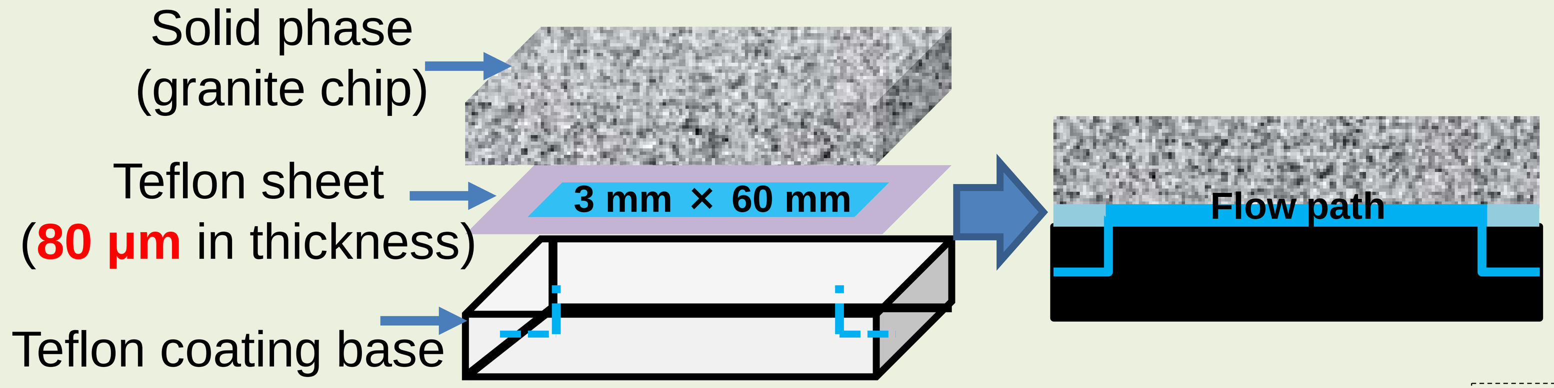


Surface of granite chip observed by digital micro scope (Kurata, 2015)

Objective

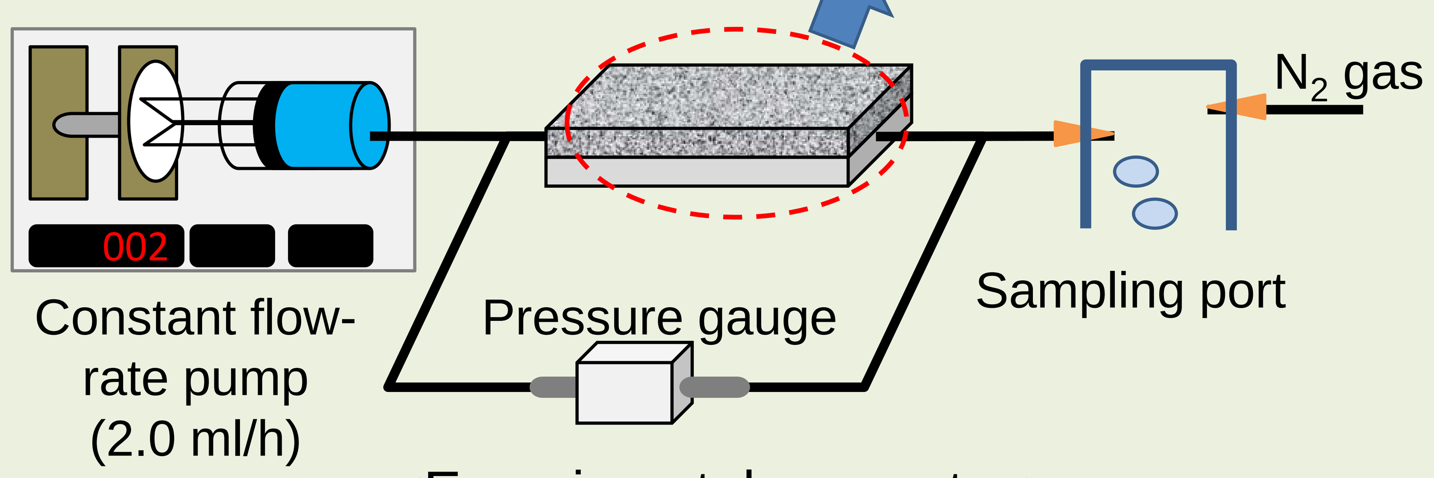
This study examined the influence of temperature on **the clogging effect** (**the permeability changes**) in micro flow-paths with the deposition of CSH-gel.

EXPERIMENTAL



<Micro flow-cell>

The permeability of overall micro flow-cell was estimated by measuring **the pressure difference** between inlet and outlet.



<Experimental apparatus>

- Flow rate: 2.0 ml/h.
- Solution: 8.5 mM $Ca(OH)_2$ adjusted to **pH 12.2 – 12.5** with NaOH.
- Temperature: 278, 296, 313 K. (submerging the micro flow-cell into a thermostat water bath)
- Sampling port was filled with N_2 gas.
- Eluted Ca and Si were measured by ICP-ASE.

Theoretical

<Measurement of Permeability>

Permeability can be estimated from *Δp*.

Permeability (m²) ***k* = *b*²/12** (Oron, 1988)

$$v = \frac{k}{\mu}\frac{p_{in} - p_{out}}{x_1}, \quad (\Delta p \equiv p_{in} - p_{out}),$$

$$Q = wbv$$

$$\therefore b = \left(\frac{12\mu Q x_1}{w\Delta p}\right)^{\frac{1}{3}}$$

v: Darcy's fluid flow velocity (m/s)
μ: fluid viscosity (Pa·s)
p: pressure (Pa)
x_1: length of micro flow-cell (m)
w: width of micro flow-cell (m)
b: aperture of flow-path (m)
Q: volume flow rate (m³/s)

<Evaluation of Deposition rate of CSH-gel>

Deposition rate constant can be evaluated from the time dependency of the permeability change.

$$\rho_M (b_0 - b) = \int_0^t k_R c dt$$

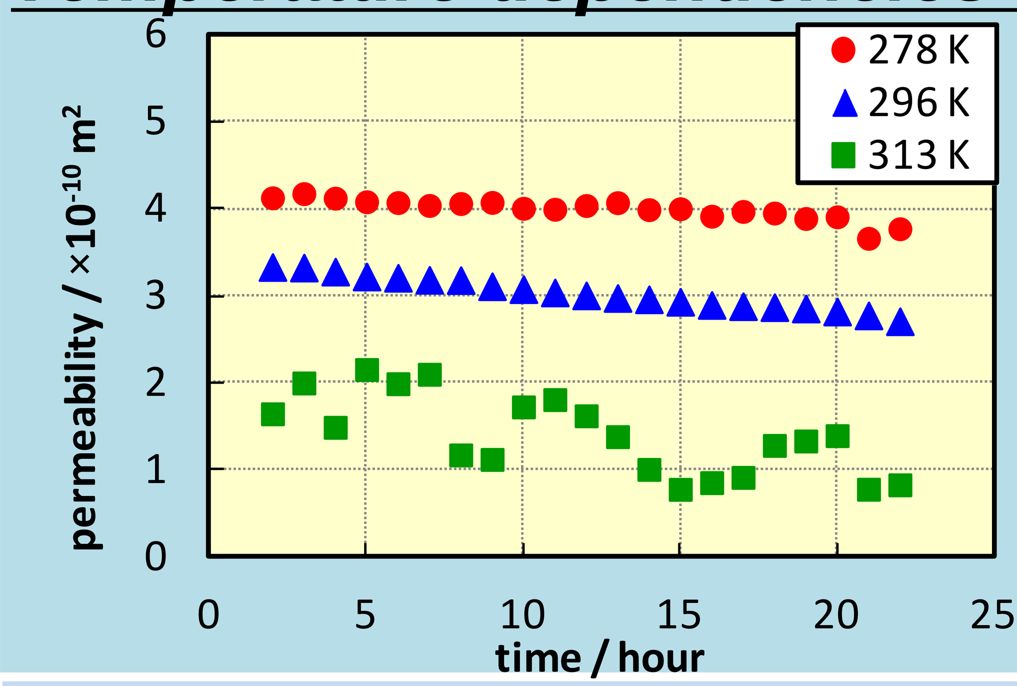
$$k_R = \frac{\rho_s (b_0 - b)}{ct}$$

$$\therefore \sqrt{K} = 1 - \frac{k_R c}{\rho_s b_0} t$$

k_R: deposition rate constant (m/s)
ρ_M: density of CSH (mol/m³)
p: pressure (Pa)
c: Ca concentration (mol/dm³) (*c* is constant.)
b_0: initial aperture of flow-path (m)
b: aperture of flow-path (m)
k_0: initial permeability (m²)
$K = k/k_0$.

RESULTS and DISCUSSION

Temperature dependencies of the permeability change

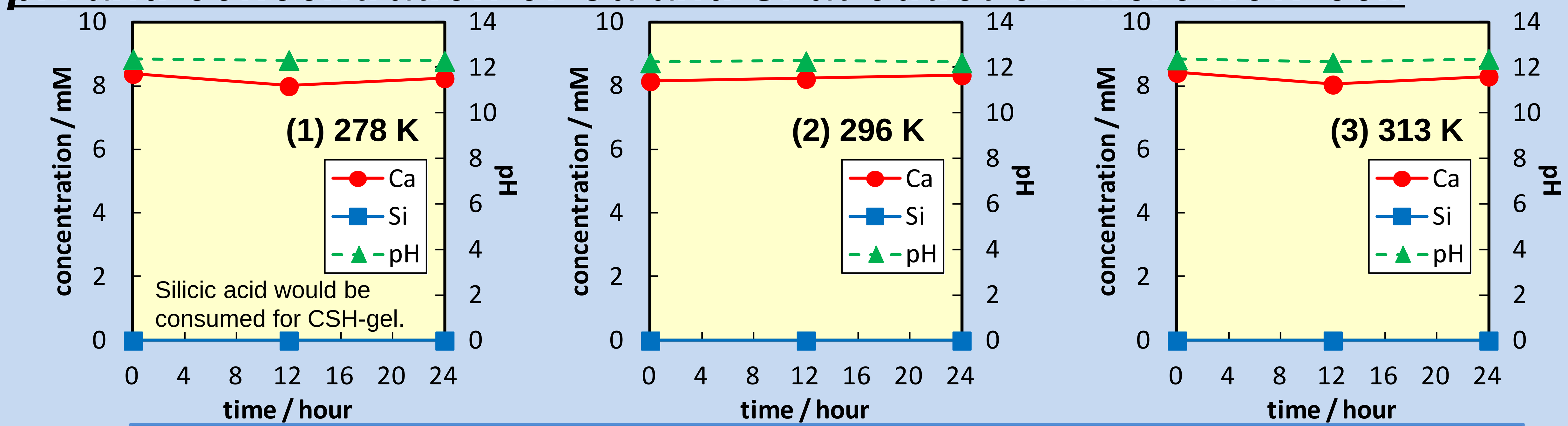


Permeability became lower with the increase in temperature.

✓Deposition rates of CSH-gel became larger with the increase in temperature.
- Formation of CSH-gel becomes faster.
- Supply rate of silicic acid from granite increases.

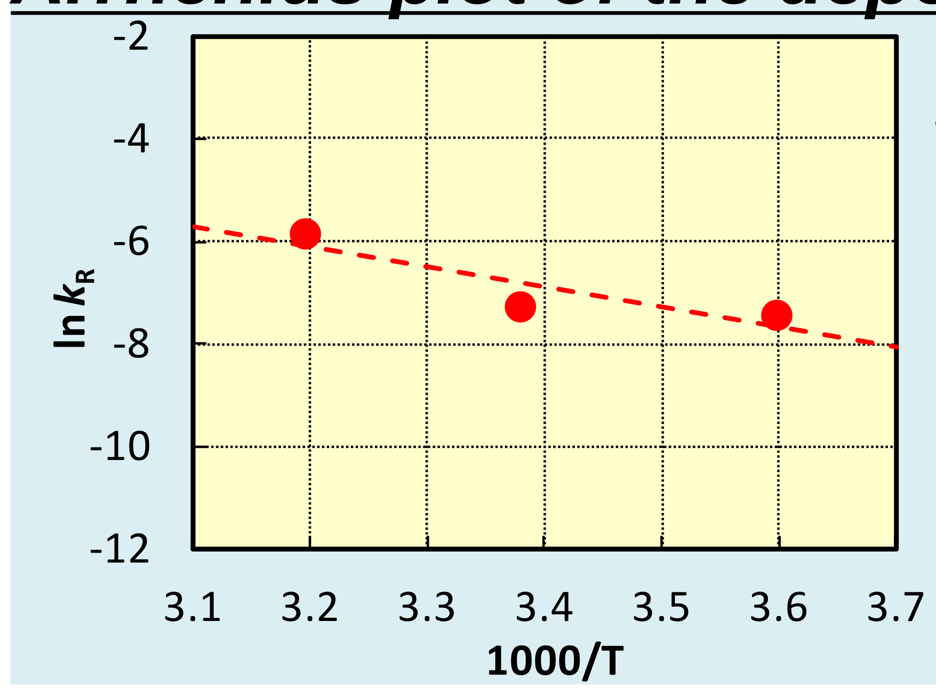
Flow paths around the repository would undergo the clogging with CSH-gel in a shorter time-period under a condition of higher temperature.

pH and concentration of Ca and Si at outlet of micro flow-cell



The concentration of Ca ions and pH at the outlet were kept the condition of the injected solution.

Arrhenius plot of the deposition rates of CSH-gel



Deposition rate constants became larger with increment of temperature. Apparent activation energy was **32.0 kJ/mol**.

✓Activation energy is smaller than that of quartz (45.5 kJ/mol).

✓Deposition rate decreased due to the slightly outflow from the micro flow-cell (Kurata,2015).

Deposition of CSH-gel was controlled by…

- Supply of silicic acid from quartz in granite.
- Migration of suspended CSH-gel (mass transfer).

CONCLUSIONS

Main results in this study are as follow:

✓Permeability through micro flow-cell became lower with the increase in temperature.
✓CSH-gel formation accelerated with the increase in the supply rate of silicic acid.
✓Deposition rate constants became larger with the increase in temperature. (although the suspension of CSH-gel might flow out of the micro flow system)

The clogging with CSH-gel in fracture of granite would be accelerated under a relatively higher temperature condition such as in the geological repository.

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