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Measurements of Thermodynamic Data of Water in Na-Bentonite in the Standard Condition by Relative Humidity Method

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- Buffer material (compacted bentonite) composing engineered barrier in the geological disposal of a HLW develops swelling stress by penetration of groundwater from the surrounding rock mass.

- In the past studies, we measured the activities of water and the Gibbs free energies of water in Na-montmorillonite which is the major component of Na-bentonite used as a buffer material, and proposed a thermodynamic model to analyze the swelling stress of bentonite based on thermodynamic theory [eg., Sato 2007, 2008a, 2008b].

- However, the thermodynamic data of water in bentonite are limited. In this study, we measured the thermodynamic data of water in Na-bentonite and Na-montmorillonite by measuring relative humidity (RH) and temperature. Based on the thermodynamic data of water and the thermodynamic model developed so far, we calculated the swelling stress of bentonite and compared with measured data.
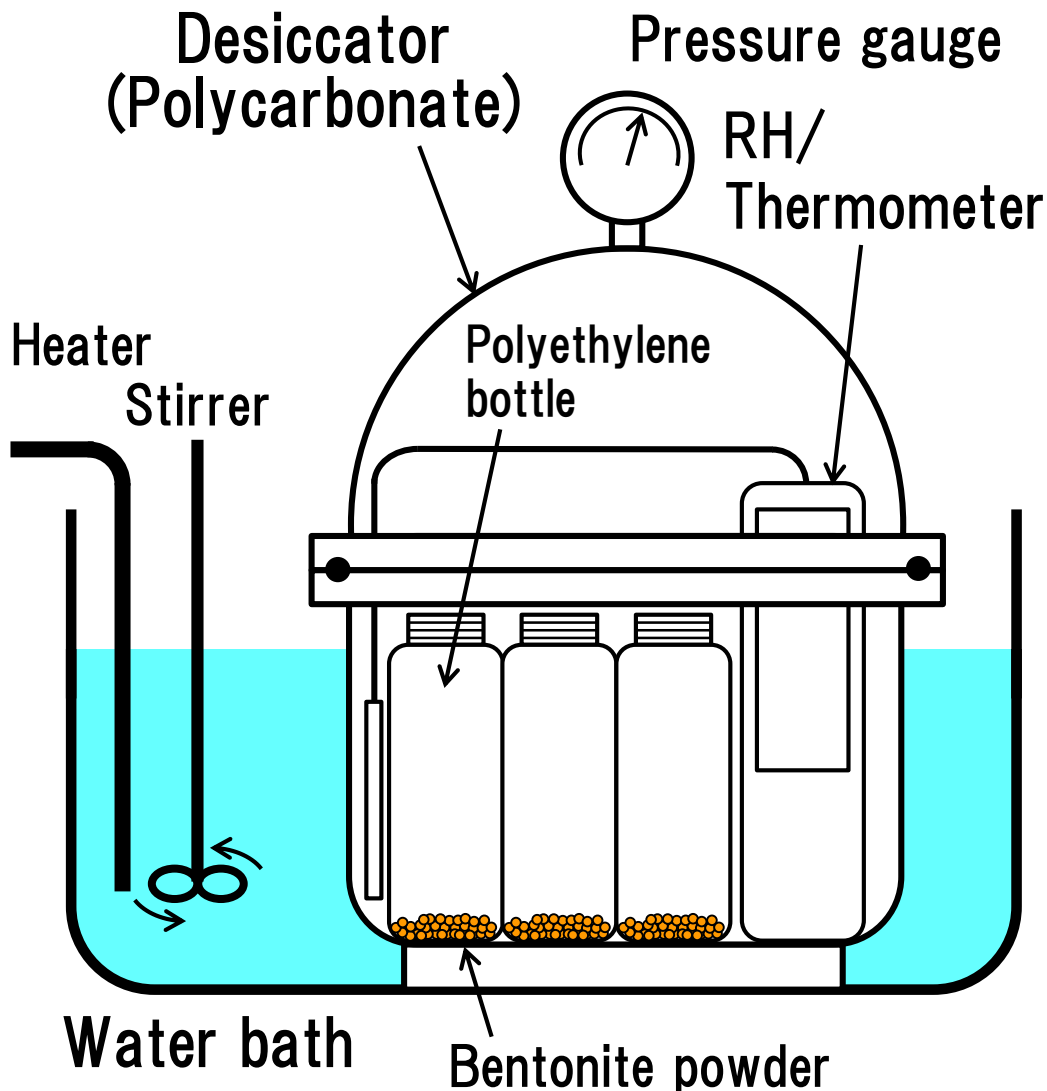
[1] H. Sato：ICONE15-10207 (2007).
[2] H. Sato：Phys. and Chem. of the Earth 33, pp.S538-S543 (2008a).
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Desiccator
(Polycarbonate)

Pressure gauge

RH/
Thermometer

Heater

Stirrer

Polyethylene
bottle

Water bath

Bentonite powder

Concept of measurement of water
vapor pressure by RH method

Procedure & Condition

① Dry Na-bentonite powder (Kunigel-V1 & Kunipia-F (Kunimine Industries Co. Ltd.), 105-110℃, 24h～, 3.00g/ sample, n=3))

② Adsorb vapor to bentonite (RH=100%, lower than -99kPa, ～9 months, periodically measure the weight of water)

③ Measure RH & temperature (-101.3kPa, 25℃, measure every 24h)

④ Take out sample (bentonite powder) & measure the weight of water

⑤ Draw a vacuum to lower water content

⑥ Repeat procedures ③～⑤ vs. water content (～100%)
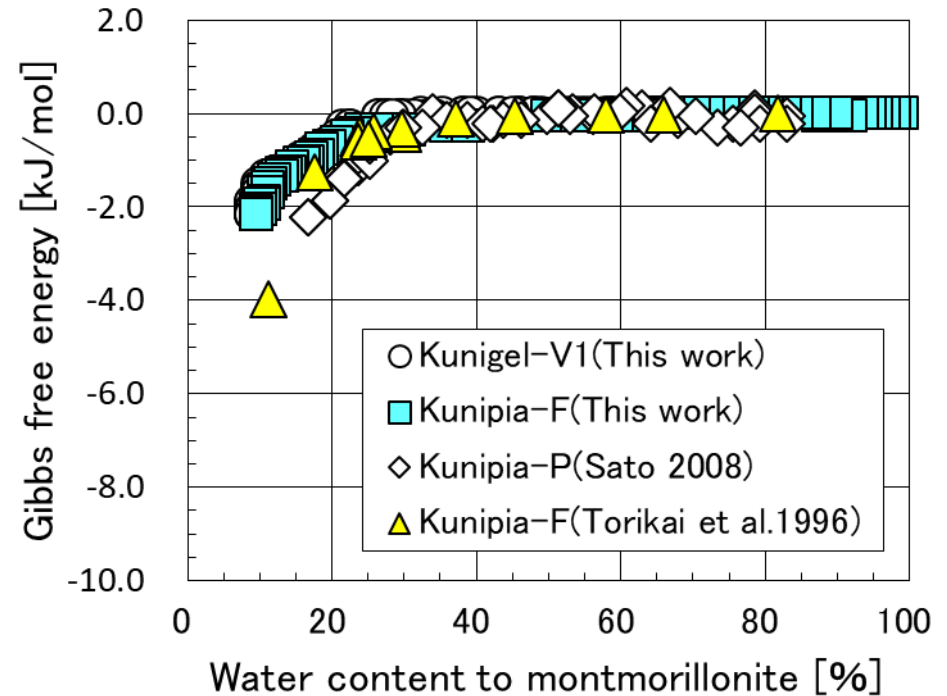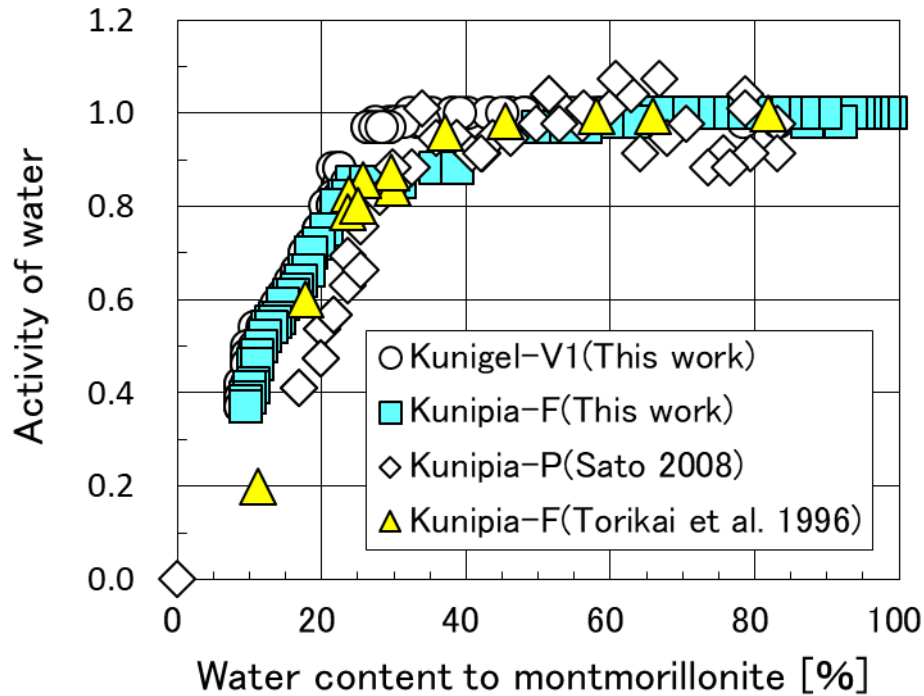
Activity of water

$$a^o_{H2O} = \frac{P^\beta_{H2O}}{P^o_{H2O}} = \frac{RH}{100} \qquad (1)$$

Relative partial molar Gibbs free energy

$$dG^o_{H2O} = RT \ln\left(\frac{RH}{100}\right) \qquad (2)$$

a^o_{H2O}: activity of water

dG^o_{H2O}: relative partial molar Gibbs free energy

P^β_{H2O}: vapor pressure of water in the β phase at 25℃

P^o_{H2O}: saturated vapor pressure of pure water at 25℃ (3.168kPa)

RH: relative humidity at 25℃ (%)
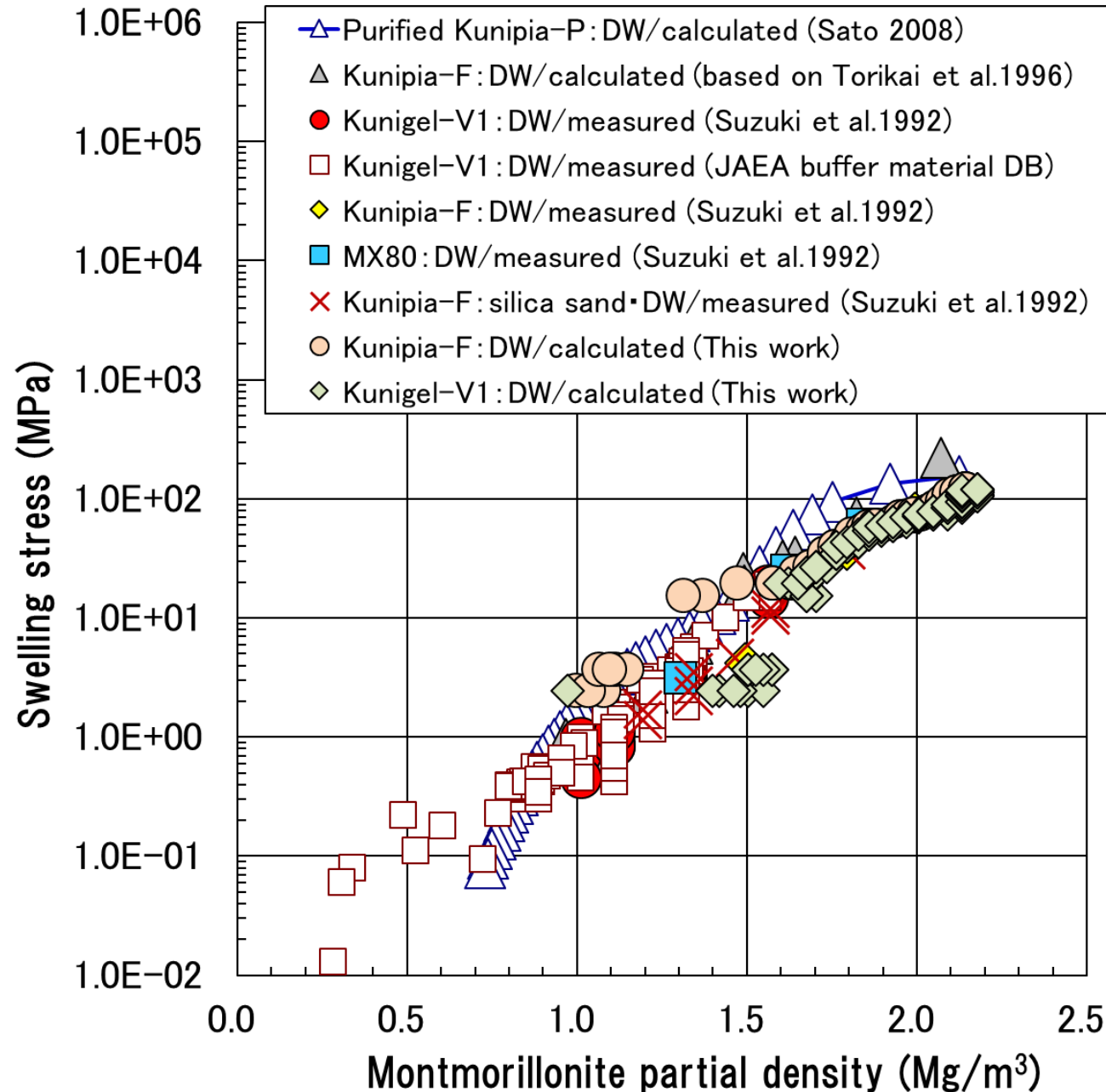
R: gas constant (8.314J/mol/K)

T: absolute temperature (273.15+t (℃), K)

Legend:
- Purified Kunipia-P: DW/calculated (Sato 2008)
- Kunipia-F: DW/calculated (based on Torikai et al.1996)
- Kunigel-V1: DW/measured (Suzuki et al.1992)
- Kunigel-V1: DW/measured (JAEA buffer material DB)
- Kunipia-F: DW/measured (Suzuki et al.1992)
- MX80: DW/measured (Suzuki et al.1992)
- Kunipia-F: silica sand・DW/measured (Suzuki et al.1992)
- Kunipia-F: DW/calculated (This work)
- Kunigel-V1: DW/calculated (This work)

Y-axis: Swelling stress (MPa)
X-axis: Montmorillonite partial density (Mg/m³)

Thermodynamic Model for Swelling Stress [Sato 2007, 2008a, 2008b]

$$dP_{ext} = \frac{-dG^o_{H2O}}{V_w}$$

$$= -\frac{RT}{V_w} \ln\left(\frac{RH}{100}\right) \quad (3)$$

DW: distilled water system

dP_{ext}: swelling stress (Pa)

dG^o_{H2O}: relative partial molar Gibbs free energy (J/mol)

V_w: specific volume of pure water at 25℃ (18.0686cm³/mol)

RH: relative humidity at 25℃ (%)

R: gas constant (8.314J/mol/K)

T: absolute temperature (273.15+t (℃), K)

Conclusions

- We measured the thermodynamic data of water (activities of water (a_{H2O}^o) & relative partial molar Gibbs free energies (dG_{H2O}^o)) in Na-bentonite and Na-montmorillonite vs. water content, and calculated the swelling stresses of bentonite based on the thermodynamic data and model.

- The calculated results of swelling stress of bentonite were in good agreement with measured data. Therefore, thermodynamic model is useful to calculate the swelling stress of bentonite.

Acknowledgements

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2. H. Sato: Thermodynamic Understanding on Swelling Pressure of Bentonite Buffer, Proc. of the International Conference on Nuclear Engineering (ICONE15), ICONE15-10207, 7 pages (in pdf format) (2007).

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