

Thermal influences on swelling pressure and swelling deformation of bentonites and investigation of its factors

Effets thermiques sur la pression et les déformations de gonflement des bentonites et facteurs d'influence

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ABSTRACT: Because buffers for disposal of high-level radioactive wastes must have high swelling characteristics for sealing wastes, bentonite is currently designated for that use. High-level radioactive wastes generate decay-heat, and it has been inferred that the swelling characteristics of bentonite decline because of the decay-heat of wastes. This study investigated swelling pressure and swelling deformation of bentonites during some thermal experiments conducted in the laboratory. Moreover, this study discusses the mechanism of thermal influences to bentonite-swelling by considering the above experimentally obtained results with chemical analyses such as measurement of cation concentration of water around the specimen, a methylene blue absorption test, and X-ray powder method for heated bentonites.

RÉSUMÉ : Parce que les bouchons pour le scellement de déchets radioactifs de haut niveau doivent avoir de fortes caractéristiques de gonflement pour fixer les containers, la bentonite est utilisée actuellement pour cet usage. Les déchets radioactifs de haut niveau produisent une chaleur décroissante, et ceci entraîne une baisse des caractéristiques de gonflement de la bentonite. Ce travail étudie la pression de gonflement et les caractéristiques de déformabilité des bentonites par des expérimentations thermiques menées en laboratoire. De plus, on discute le mécanisme des effets thermiques sur le gonflement de la bentonite obtenus ci-dessus par une analyse chimique incluant la concentration en cations, l'absorption du bleu de méthylène ou la diffraction de rayons X sur des poudres de bentonite chauffée.

KEYWORDS: Bentonite, Swelling, Radioactive waste disposal, Clay minerals, Chemical properties, X-ray diffraction analysis

1 INTRODUCTION

Because buffer materials for disposal of high-level radioactive wastes (HLWs) must have high swelling characteristics and very low permeability to seal the waste, bentonite is currently designated for that use. In geological disposal of high-level radioactive wastes, bentonite-based buffer fills spaces between the waste container and bedrock because it has swelling properties and low permeability. High-level radioactive wastes generate decay heat. It is inferred that the swelling characteristics of bentonite decline by the decay heat of wastes.

The author assessed the swelling deformation of one kind of sodium bentonite that had undergone some thermal exposure in a laboratory in an earlier study (Komine and Ogata, 1998) as preliminary research. Some researchers reported changes of soil behavior during heating (Akagi, 1994; Oscarson and Dixon, 1989). It appears possible that bentonite receives decay heat of high-level radioactive waste just as other soils do (see Fig. 1). In a disposal pit, as Fig. 1 shows, the swelling characteristics of bentonite might decline because of exposure to decay heat from wastes. It is therefore necessary to investigate thermal effect on

swelling pressure and swelling deformation characteristics of some bentonites for buffer material development.

To elucidate this problem, this study investigated swelling pressure and swelling deformation of bentonites that had undergone some heat exposure in the laboratory. Moreover, this study assessed mechanisms of thermal influences to bentonite-swelling by examining the experimentally obtained results using chemical analyses for the heated bentonites such as measuring cation concentrations of water around the specimen, methylene blue absorption tests, and X-ray powder method.

2 BENTONITE USED FOR THIS STUDY AND THERMAL EXPOSURE CONDITIONS

Commercial bentonites of two kinds (Table 1) were used. Bentonite A, called Kunigel-V1, was produced at the Tsukinuno Mine in Yamagata prefecture, Japan. This sodium-type bentonite contains nearly 57% montmorillonite. It is used frequently in Japan to study artificial barrier materials against radioactive waste. Bentonite C, called Kunibond, is produced at the Dobuyama Mine in Miyagi prefecture, Japan. This calcium-type bentonite has nearly 80% montmorillonite content.

This study produced some bentonites that had undergone different thermal exposure by oven drying to investigate the thermal history influence on bentonite swelling characteristics. The heating conditions for producing the heated bentonites were 60, 90, 110, and 130°C. Heating periods were 28, 120, and 365 days. Those heating temperatures were used to simulate decay heating of HLWs based on analytical results of maximum temperatures in bentonite-based buffers, which are 65–165°C (Japan Nuclear Cycle Development Institute, 2000) and on results of previous research (Japan Nuclear Cycle Development Institute, 2000) showing that montmorillonite will not be altered to illite at less than 130°C.

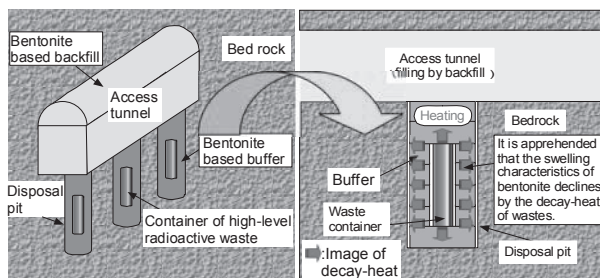


Figure 1. High-level radioactive waste disposal facility and impact on bentonite base buffers during decay-heating of wastes.

Table 1. Fundamental properties of bentonites A and C

Bentonite	A	C
Type	Sodium	Calcium
Density of soil particle(Mg/m ³)	2.79	2.71
Liquid Limit (%)	458.1	128.7
Plastic Limit (%)	23.7	38.4
Plasticity index	434.4	90.3
Montmorillonite content (%)	57	84
Cation Exchange Capacity (meq/g)	1.166	0.795
Exchange Capacity of Na ⁺ (meq/g)	0.631	0.119
Exchange Capacity of Ca ²⁺ (meq/g)	0.464	0.585
Exchange Capacity of K ⁺ (meq/g)	0.030	0.019
Exchange Capacity of Mg ²⁺ (meq/g)	0.041	0.072

After the bentonite heating described above, these samples were kept at a constant temperature (22 ± 3°C) and constant humidity (70% relative humidity) until the water contents of samples stopped changing.

3 SWELLING PRESSURE AND SWELLING DEFORMATION OF BENTONITES AFTERSOMETHERMAL EXPOSURE

This study used the experimental apparatus presented in Fig. 2. The maximum capacity and the minimum scale of the load transducer were 10 kN and 0.0025 kN, respectively. The maximum capacity and the minimum resolution of the linear variable displacement transducer (LVDT) were, respectively, 25 mm and 0.002 mm. This study conducted swelling characteristic experiments of two kinds. The swelling pressure test measured the bentonites' swelling pressure as water was supplied to the confined bentonite specimen. The swelling deformation test measured the relation between the axial swelling deformation and the time from the start of water supply. Test procedures were described in an earlier article (Komine et al., 2009).

Figure 3 portrays the relation between maximum swelling pressure and the initial dry density of bentonites' thermal exposure during swelling pressure tests. This figure shows that the influence of thermal exposure on swelling pressure characteristics of both bentonites is slight, provided that the heating temperature is less than 130°C and that the heating duration is less than 120 days. Regarding results obtained for calcium-type bentonite C depicted in Fig. 3(b), almost no

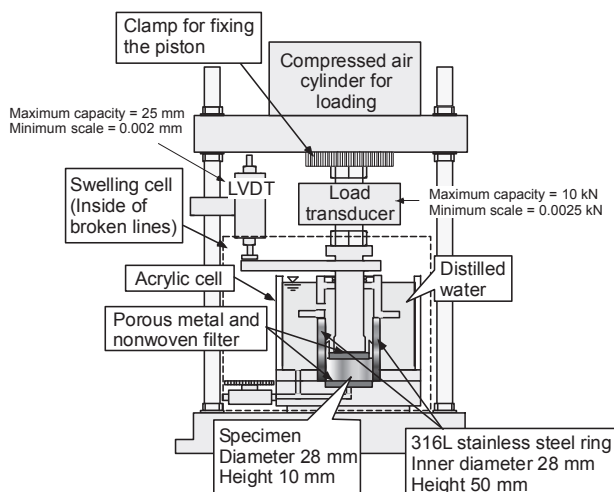
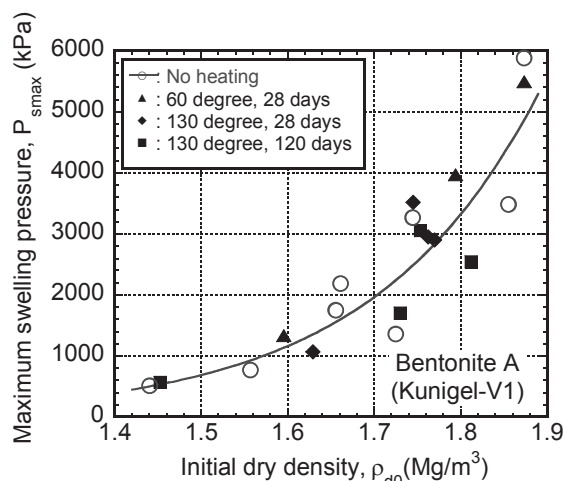
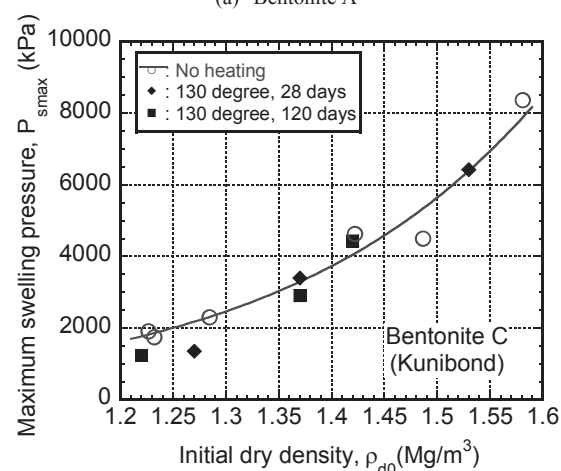


Figure 2. Experimental apparatus.



(a) Bentonite A



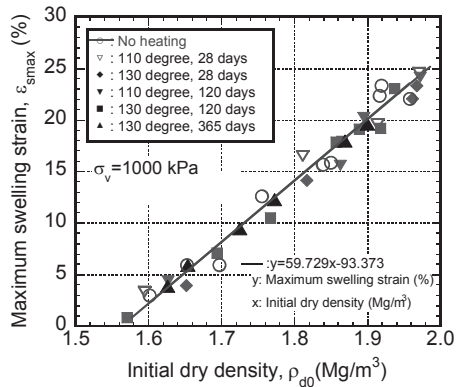
(b) Bentonite C

“No heating” in the legend denotes experimentally obtained results for bentonite with no thermal exposure. The temperature in the legend is the heating temperature by drying oven. The days in the legend show the heating duration.

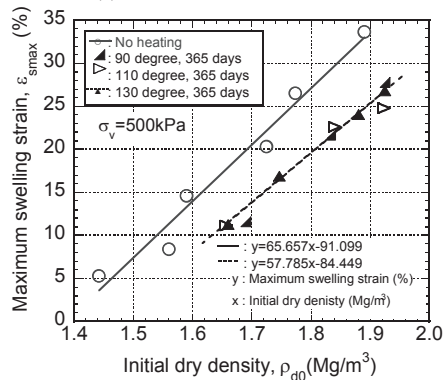
Figure 3. Relation between maximum swelling pressure and initial dry density of bentonites A and B with thermal exposure.

influence of thermal exposure on swelling pressure characteristics is shown for 1.37–1.53 Mg/m³ initial dry density. However, a slight influence of thermal exposure is apparent at 1.22–1.27 Mg/m³.

Figure 4 shows the relation between maximum swelling strain and initial dry density of sodium-type bentonite A at vertical stresses of 1000 kPa and 500 kPa. Results in this figure show that maximum swelling strain increases linearly as the initial dry density increases. For the sodium-type bentonite A, the swelling deformation property of bentonite after thermal exposure is almost unchanged under 1000 kPa vertical stress. However, the swelling deformation property of bentonite is greatly reduced for heating at 90–130°C for a 365-day heating duration under conditions of 500 kPa vertical stress, as portrayed in Fig. 4(b). Those results indicate that the thermal influence to swelling deformation characteristics of sodium-type bentonite A is dependent on the vertical stress condition. The influences of thermal exposure on swelling deformation decrease for high vertical stress such as 1000 kPa. This discussion shows agreement with previously presented discussion indicating that swelling pressure characteristics of bentonite A show almost no change according to thermal exposure, as depicted in Fig. 3(a) and previously reported experimentally obtained results (Komine and Ogata, 1998) showing that swelling deformation



(a) Vertical stress = 1000 kPa



(b) Vertical stress = 500 kPa

Figure 4. Maximum swelling strain and initial dry density of sodium-type bentonite A with vertical stress of 1000 kPa and 500 kPa.

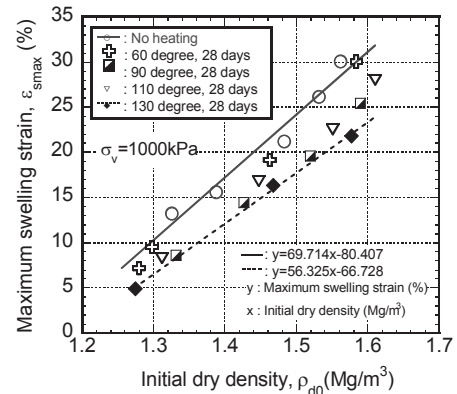
characteristics of sodium-type bentonite are strongly reduced by thermal exposure at 9.8–10.0 kPa vertical stress.

Figure 5 shows the relation between maximum swelling strain and initial dry density of calcium-type bentonite C at vertical stress of 1000 kPa. By comparing results presented in Fig. 5 with those in Fig. 4, the swelling deformation characteristics of calcium-type bentonite C are markedly reduced by thermal exposure at vertical stress of 1000 kPa. The swelling deformation characteristics of calcium-type bentonite C are strongly reduced by heating temperatures greater than 90°C for all heating durations. Furthermore, the reduction ratio of maximum swelling strain attributable to thermal exposure increases along with the initial dry density.

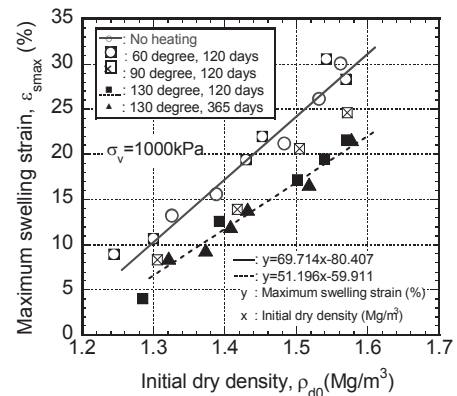
4 THERMAL INFLUENCE ON BENTONITE SHOWN BY CHEMICAL ANALYSIS RESULTS

Figure 6 presents sodium ion concentrations of water measured around the compacted specimen of sodium-type bentonite A after swelling deformation tests. This figure shows the relation between sodium ion concentration and heating duration with parameters of heating temperature and vertical stress.

Results depicted in Fig. 6 show that the sodium-ion concentration of water around the specimen is increased by thermal exposure for sodium-type bentonite A. Especially for higher heating temperatures and longer heating durations, the sodium-ion concentration of water around the specimen is higher. Moreover, Fig. 6 depicts that the sodium-ion concentration in water around the specimen at 500 kPa of vertical stress is greater than that at 1000 kPa of vertical stress. The measured results of calcium ion concentration are the same results of sodium ion concentration. Those results show that exchangeable cations such as Na^+ , Ca^{2+} are apt to elute to surrounding water around the bentonite specimen for the sodium-type bentonite A. In contrast, Table 2 presents results for calcium-type bentonite C which show that the sodium and calcium ion concentrations are almost unchanged irrespective of



(a) Heating duration 28 days



(b) Heating duration 120 days and 365 days

Figure 5. Relation between maximum swelling strain and initial dry density of calcium-type bentonite C at vertical stress of 1000 kPa.

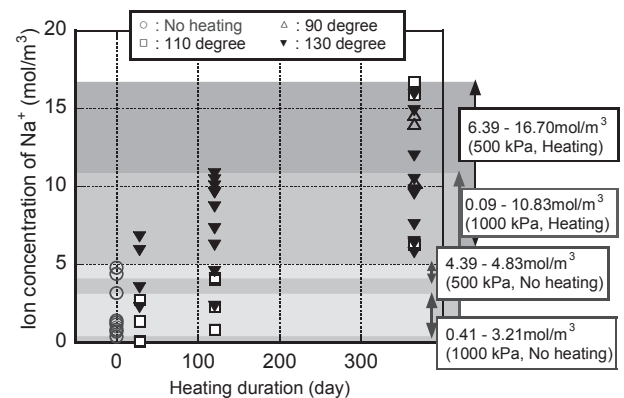


Figure 6. Results of sodium ion concentration of water around the compacted specimens of bentonite A after swelling deformation tests.

Table 2. Results of sodium and calcium ions concentrations of water around calcium-type bentonite C after swelling pressure and swelling deformation tests.

Experiment	Heating temperature (°C)	Heating duration (day)	Na^+ ion concentration (mol/m ³)	Ca^{2+} ion concentration (mol/m ³)
Swelling pressure test	No heating		0.33–1.65	0.01–0.16
	130	28–120	0.20–0.46	0.00–0.14
Swelling deformation test at 1000 kPa vertical stress	No heating		0.21–0.59	0.05–0.48
	90–130	28	0.03–1.27	0.01–0.45
	90–130	120	0.06–0.84	0.17–0.27
	130	365	0.50–0.78	0.12–0.25

thermal exposure. Therefore, the thermal influence on calcium-type bentonite differs from that on sodium-type bentonite.

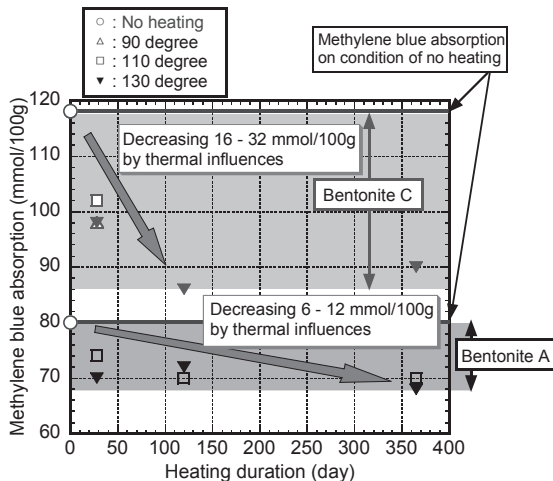


Figure 7. Methylene blue absorption for bentonites A and C with thermal exposure.

Figure 7 portrays the methylene blue absorption for bentonites A and C that had undergone thermal exposure. Results show that the absorbing capacities of methylene blue for bentonites of both kinds are reduced by thermal exposure.

Figure 8 presents X-ray diffraction plots of bentonites A and C experienced thermal exposure. Results show that the X-ray diffraction plot of bentonite A has almost no change by thermal exposure. However, the plot for bentonite C shows a marked change because of thermal exposure. Therefore, the mechanism of thermal effects on sodium- and calcium-type bentonites can be understood as shown in Fig. 9.

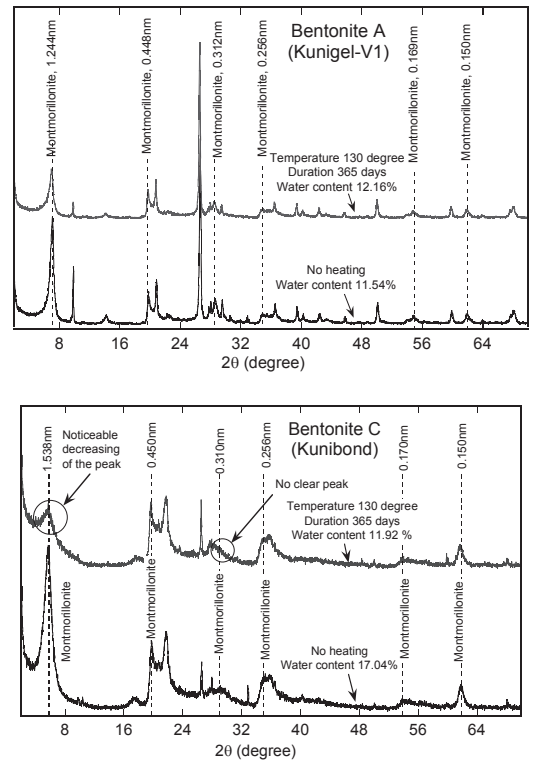


Figure 8. X-ray diffraction plots of Bentonite A and C with thermal exposure.

5 CONCLUSIONS

This study quantitatively assessed thermal exposure effects on the swelling characteristics of sodium-type and calcium-type bentonites using swelling pressure and swelling deformation testing of bentonites that had undergone thermal exposure. This report described mechanisms of thermal influences on swelling of these heated bentonites by consideration of the experimentally obtained results with measurements of cation concentrations of water surrounding the specimens, with methylene blue absorption tests, and X-ray powder method.

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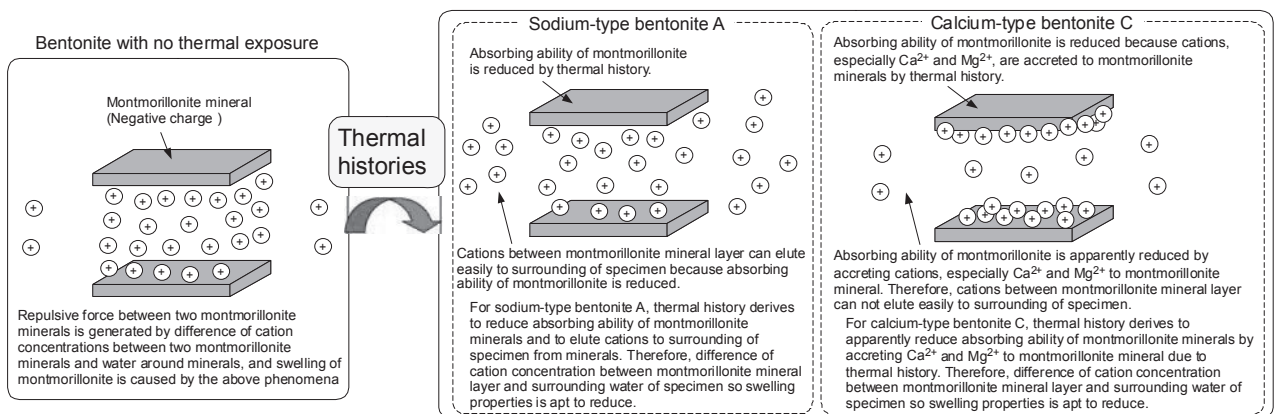


Figure 9. Mechanism of thermal influences to bentonite-swelling