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 is 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.Ingeological disposal of high-level radioactive wastes, bentonite-based buffer fills spaces between the wastecontainer and bedrockbecause it has swelling properties and low permeability. High-level radioactive wastes generate decayheat. It is inferred that the swelling characteristics of bentonite declineby the decayheat of wastes.

The author assessed the swellingdeformationof one kind of sodium bentonite thathad undergonesome 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 appearspossible that bentonite receives decay heat of high-level radioactive wastejust as other soils do (see Fig. 1). In a disposal pit, as Fig. 1 shows, the swelling characteristics of bentonite might declinebecause of exposure to decay heat from wastes. It is therefore necessary to investigate thermal effectson swelling pressure and swelling deformation characteristics of somebentonitesfor buffer material development.

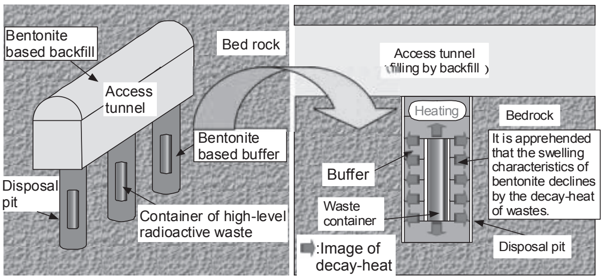


Figure 1. High-level radioactive waste disposal facility and impact on bentonite base buffers during decay-heating of wastes.

To elucidatethis problem, this study investigated swelling pressure and swelling deformation of bentonites that had undergone some heatexposure in the laboratory. Moreover, this study assessedmechanisms of thermal influences to bentonite-swelling by examiningthe 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 bentonitesof 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 studyartificial barrier materials against radioactive waste. Bentonite C, called Kunibond, is produced at the Dobuyama Mine in Miyagi prefecture, Japan. This calcium-type bentonite hasnearly 80% montmorillonite content.

This study produced some bentonites that had undergone different thermal exposure by oven drying to investigate the thermal history influence onbentonite swelling characteristics. The heating conditions for producing the heated bentonites were 60, 90, 110, and 130°C.Heating periods were28, 120, and 365 days.Those heating temperatureswere used to simulate decayheating of HLWsbased on analytical results of maximum temperatures in bentonite-basedbuffers, whichare65–165°C (Japan Nuclear Cycle Development Institute, 2000) andon results of previous research (Japan Nuclear Cycle Development Institute, 2000) showing that montmorillonite will not be altered to illite at less than 130°C.

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 werekeptat 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 conductedswelling characteristic experiments of twokinds. The swelling pressure test measured the bentonites' swelling pressure as water was supplied to the confined bentonite specimen. The swelling deformation test measured the relationbetween the axial swelling deformation and the time from the start of water supply. Test proceduresweredescribed in an earlierarticle (Komine et al., 2009).

Figure 3 portrays the relation between maximum swelling pressure and the initial dry density of bentonites' thermal exposureduring swellingpressure tests. This figureshows that the influence of thermal exposureon swelling pressure characteristics of both bentonites is slight, provided that theheating temperature is less than 130°C and that the heating duration is less than 120 days.Regardingresultsobtained for calcium-type bentonite C depicted in Fig. 3(b), almost no influenceof thermal exposureon swelling pressure characteristics is shown for1.37–1.53 Mg/m^{3}initial dry density. However, a slightinfluenceof thermalexposure is apparentat 1.22–1.27 Mg/m^3.

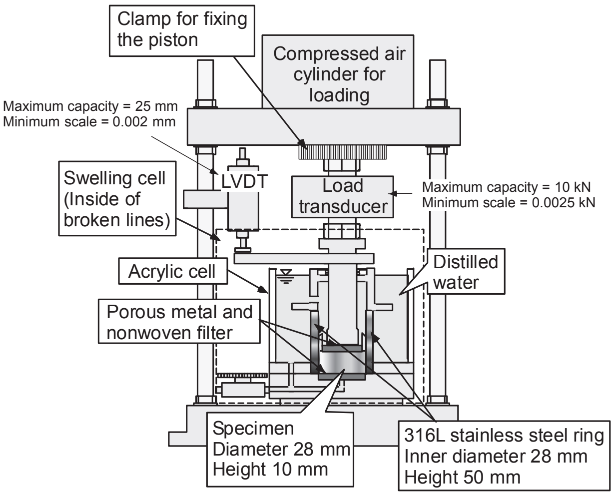
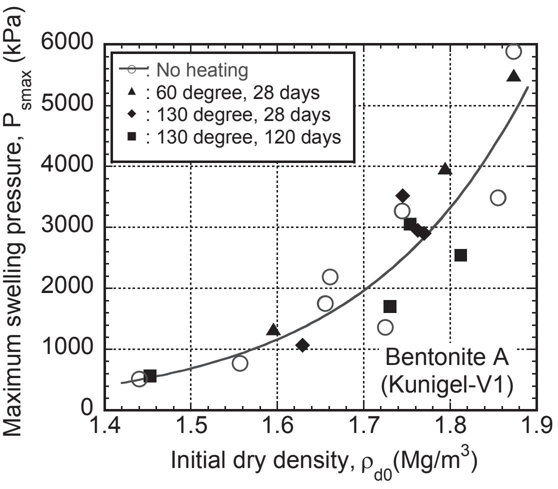
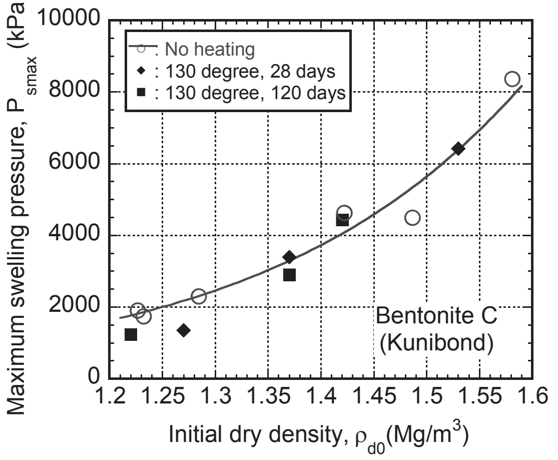


Figure 2. Experimental apparatus.



(a) Bentonite A

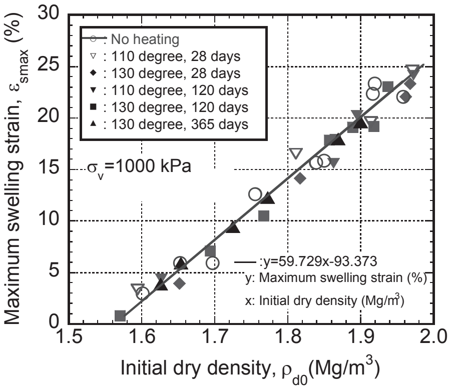


(b) Bentonite C

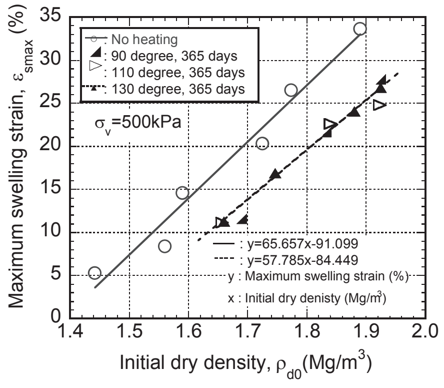
"No heating" in the legend denotes experimentally obtained results for bentonite with no thermal exposure. The temperature in the legend is the heating temperature by dryingoven. The days in the legend show the heating duration.

Figure 3. Relationbetween maximum swelling pressure and initial dry density of bentonites A and B withthermal exposure.

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 figureshow that maximum swelling strain increases linearly as theinitial dry density increases. For the sodium-type bentonite A, the swelling deformation property of bentonite after thermal exposure is almost unchangedunder 1000 kPa vertical stress. However, the swelling deformation property of bentonite isgreatlyreduced for heating at 90–130°C fora 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 onthe vertical stress condition. The influences of thermal exposureon swellingdeformation decrease for high vertical stresssuch as 1000 kPa. Thisdiscussion shows agreement with previously presenteddiscussionindicating that swelling pressure characteristics of bentonite Ashowalmost no change according to thermal exposure, as depicted in Fig. 3(a) and previously reportedexperimentally obtained results (Komine and Ogata, 1998) showing thatswelling 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 stressof 1000 kPa and 500 kPa.

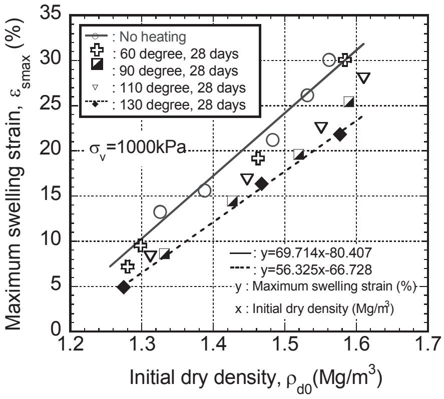
characteristics of sodium-type bentonite are strongly reduced by thermal exposure at 9.8–10.0 kPa vertical stress.

Figure 5 showsthe relationbetween 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 swellingdeformation characteristics of calcium-type bentonite C aremarkedlyreduced by thermal exposure at vertical stress of 1000 kPa. The swellingdeformation characteristics of calcium-type bentonite C are strongly reduced byheating temperatures greater than 90°Cfor all heating durations. Furthermore, the reduction ratio of maximum swelling strain attributable tothermal exposureincreases along with theinitial dry density.

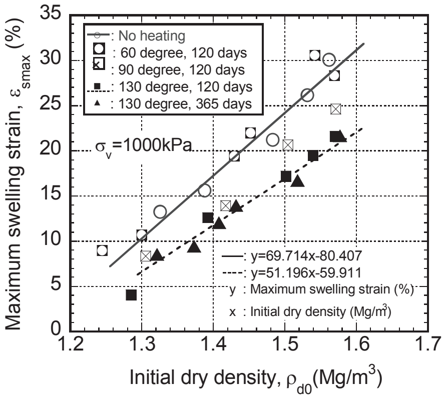
4 THERMAL INFLUENCEON BENTONITESSHOWN BYCHEMICAL ANALYSIS RESULTS

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 relationbetween sodium ion concentration and heating duration with parameters of heating temperature and vertical stress.

Results depicted in Fig. 6 show thatthe sodium-ion concentration of water around the specimen is increasedby thermal exposure for sodium-type bentonite A. Especiallyfor 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 thesame 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 thatthe sodium and calcium ionconcentrations are almost unchanged irrespective of thermal exposure. Therefore, the thermal influence on calcium-type bentonite differs from that on sodium-type bentonite.



(a) Heating duration 28 days



(b) Heating duration 120 days and 365 days

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

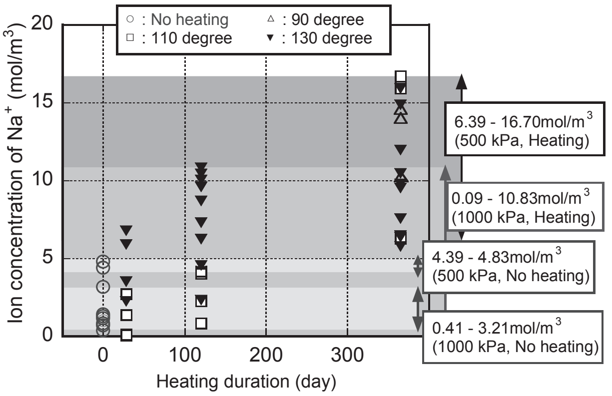


Figure 6. Results of sodium ion concentration of water around the compacted specimens of bentonite A after swellingdeformation tests.

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

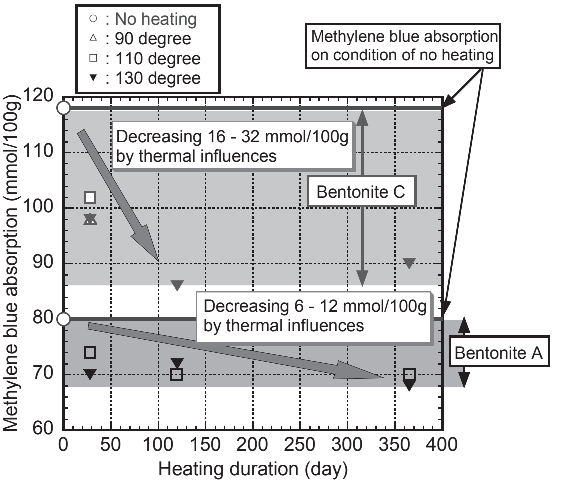


Figure 7.Methylene blue absorption for bentonites A and C with thermal exposure.

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

Figure 8 presents X-ray diffraction plots of bentonites A and C experienced thermal exposure. Results show thatthe X-ray diffraction plot of bentonite A hasalmost 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.

5 CONCLUSIONS

This study quantitatively assessedthermal exposure effects onthe swelling characteristics of sodium-type and calcium-type bentonites using swellingpressure and swellingdeformation testing of bentonites that had undergonethermal exposure. This reportdescribedmechanisms of thermal influences onswelling 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.

6 ACKNOWLEDGMENTS

This study was supported through funding by research funds of the Japanese Ministry of Education, Culture, Sports, Science and Technology and the TEPCO Research Foundation. The author alsothanks all members and students of the Geotechnical Laboratory, Ibaraki University, for their kind assistance and discussions.

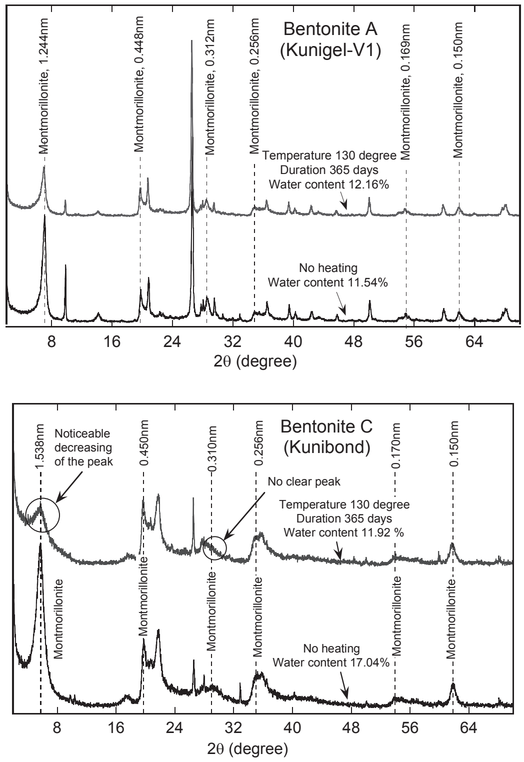


Figure 8. X-ray diffraction plots of Bentonite A and C with thermal exposure.

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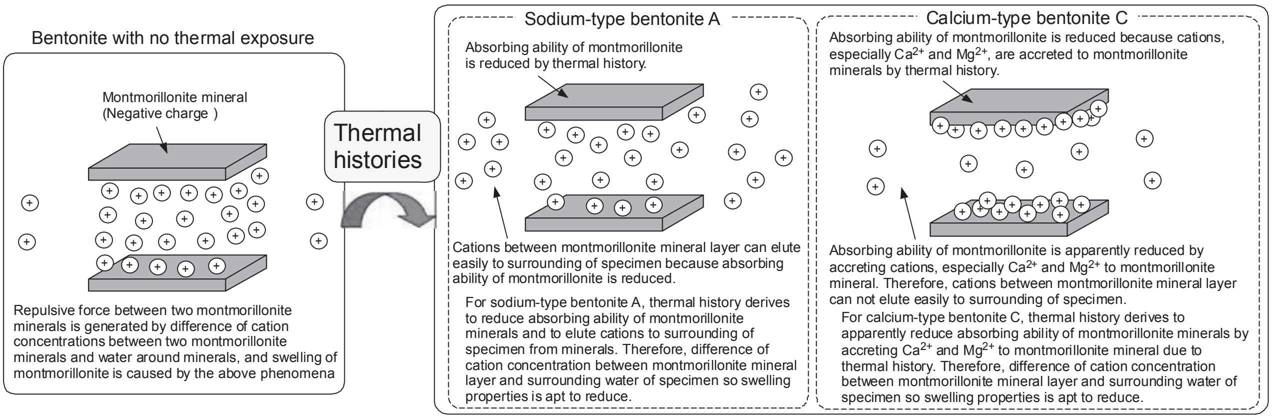

Repulsive force between two montmorillonite minerals is generated by difference of cation concentrations between two montmorillonite minerals and water around minerals, and swelling of montmorillonite is caused by the above phenomena

Absorbing ability of montmorillonite is reduced by thermal history.

Cations between montmorillonite mineral layer can elute easily to surrounding of specimen because absorbing ability of montmorillonite is reduced.

For sodium-type bentonite A, thermal history derives to reduce absorbing ability of montmorillonite minerals and to elute cations to surrounding of specimen from minerals. Therefore, difference of cation concentration between montmorillonite mineral layer and surrounding water of specimen so swelling properties is apt to reduce.

Absorbing ability of montmorillonite is reduced because cations, especially Ca^{2+} and Mg^{2+}, are accreted to montmorillonite minerals by thermal history.

Absorbing ability of montmorillonite is apparently reduced by accreting cations, especially Ca^{2+} and Mg^{2+} to montmorillonite mineral. Therefore, cations between montmorillonite mineral layer can not elute easily to surrounding of specimen.

For calcium-type bentonite C, thermal history derives to apparently reduce absorbing ability of montmorillonite minerals by accreting Ca^{2+} and Mg^{2+} to montmorillonite mineral due to thermal history. Therefore, difference of cation concentration between montmorillonite mineral layer and surrounding water of specimen so swelling properties is apt to reduce.

Figure 9. Mechanism of thermal influences to bentonite-swelling