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EXPERIMENT REGARDING MONTMORILLONITE CONTENT AND STRENGTH PROPERTIES IN BENTONITE UNDER THE INFLUENCE OF HEAT

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ABSTRACT: At present, underground geological disposal at depths greater than 300 m is considered a viable disposal option for high-level radioactive waste generated from the reprocessing of spent fuel used in nuclear power plants. According to the Japan Nuclear Cycle Development Institute [1] and Agency for Natural Resources and Energy [2], in geological disposal, bentonite is employed as the primary component of buffer material to fill gaps between waste and geologic rock. However, specification details have yet to be completely determined. It is anticipated that the buffer material will be exposed to high temperature due to the heat generated by the vitrified solid. This study aims to understand the mechanical properties of bentonite buffer material by employing uniaxial compression tests with temperatures between 30°C to 90°C applied to bentonite specimens having received a temperature history treatment of 200°C applied over six months. Results show that the maximum compressive strength of the sample given the temperature history is lower than that of the sample without the temperature history. Furthermore, the montmorillonite content of the sample given a temperature history of 200°C also decreased. The reason for the decrease in strength seems to be that the compaction energy during fabrication decreased due to the decrease in the montmorillonite content. This is because the specimens were fabricated so that the dry density was constant. It seems that the maximum compressive stress decreased with increasing temperature regardless of temperature history. The reason for this seems to be microcracks, which appear to decrease rigidity and, therefore, compressive strength.

Keywords: Bentonite, Silica sand, Uniaxial compressive strength, Montmorillonite content

1. INTRODUCTION

The spent nuclear fuel generated by nuclear power generation produces high-level radioactive waste when it is reprocessed. As a disposal method, geological disposal to a depth of 300m or below is often employed. Geological disposal is a disposal method that isolates and confines radioactive materials from the human environment for an extremely long period of time. It is a multi-barrier system that combines an artificial barrier made of vitrified solid, carbon steel over pack, and bentonite cushioning material with a natural barrier made of stable rock, deep underground. The cushioning material filled between the waste and the bedrock has water-impermeable properties, self-sealing properties, various sorption delay properties, thermal conductivity, chemical buffering properties, over pack supporting properties, and stress buffering properties, among others. These properties are expected to be maintained for long periods of time. Naturally occurring clay is drawing attention as a material that can relatively satisfactorily satisfy these functions. Among these natural clay materials, compressed bentonite has the following characteristics.

(1) Ability to control water movement due to low water permeability.
(2) A mechanism for expanding with the infiltration of water to fill voids in the compressed bentonite and cracks in the adjacent rock mass.
(3) Cation exchange capacity to adsorb cation nuclides.

Therefore, bentonite is considered to be the most effective cushioning material. Bentonite is a clay whose main component is the smectite. According to Onigata [3], smectite is a generic name for montmorillonite, beidellite, nontronite, saponite, hectorite, sauconite, and stevensite clay minerals. Among them, montmorillonite is a representative clay mineral. The smectite contained in bentonite, which has been verified as a cushioning material in Japan, is also montmorillonite. The crystals of clay minerals are composed of overlapping silicic acid tetrahedral sheets and alumina octahedral sheets. Bentonite has remarkable swelling and water-absorbing properties, very low water permeability, and high cation exchange capacity. However, details about the mechanical properties of bentonite cushioning materials considering the temperature history due to nuclide decay heat have not been fully grasped. Therefore, in this study, uniaxial compression tests were performed on specimens containing bentonite and silica sand (either with an applied temperature history or without an applied temperature history), and the results were compared. From this, the strength characteristics of bentonite cushioning material with temperature are better understood.

2. EXPERIMENTAL OUTLINE

In this study, changes in the strength characteristics are clarified by applying change in temperature to bentonite-silica sand mixed samples that have been given a temperature history. In the experiment, a cylindrical specimen with a diameter of 35 mm and a height of 80 mm was prepared. The temperature history was applied to powdered bentonite. In an actual disposal facility, bentonite compacted at high pressure is considered the best material from the viewpoint of improving the performance and workability of the cushioning material. Therefore, a method of applying temperature history by placing the compacted bentonite specimen into the drying furnace is considered valid. However, in the above method, a temperature gradient is likely generated inside the test piece, creating a nonuniform temperature history condition. Therefore, Hideo, Ryoya, Kazuya, and Satoshi, [4] with reference to the experiment, adopted a method of giving a temperature history to powdered bentonite. After a certain period of time, a uniaxial compression test was performed with the specimen kept in the water tank. The montmorillonite content was measured by performing a methylene blue adsorption measurement test on the sample after destruction.

3. EXPERIMENT METHOD

3.1 Specimen Preparation

The procedure is shown below.

(1) Bentonite and silica sand No. 8 were mixed at a ratio of 7: 3.
(2) The sample was placed in a stainless-steel vat and placed in a temperature-controlled oven for 6 months to provide a temperature history. The temperature history was applied to the bentonite sample while the vat was opened, and the drying furnace was kept at atmospheric pressure. In this study, the temperature measured in the drying furnace was the heating temperature of bentonite.
(3) The sample was taken out and the water content was adjusted using a hand mixer and a sprayer. The water content was measured by the microwave oven method (500w, 15 minutes).
(4) The mass at which the dry density was 1.6 Mg/m^3 was determined and weighed with an electronic balance.
(5) The measured sample was divided into 5 parts, put into a mold, and compacted with a tamping rod.
(6) The top of the mold was covered and compressed with a hydraulic jack for 10 minutes. The molding pressure was set to 40 MPa.
(7) After 10 minutes, the height of the specimen and the diameter of the upper and lower specimen were measured with a caliper. The mass was weighed with an electronic balance.

3.2 Temperature Measurement Test Inside the Specimen

In order to determine the time needed to apply the temperature to the test piece, the internal temperature was measured while applying the temperature. A sample without temperature history was compacted to make a specimen, and paulownia was used to make a hole. After that, a thermocouple was inserted inside the specimen, and the rate of temperature increase was measured while the temperature was applied.

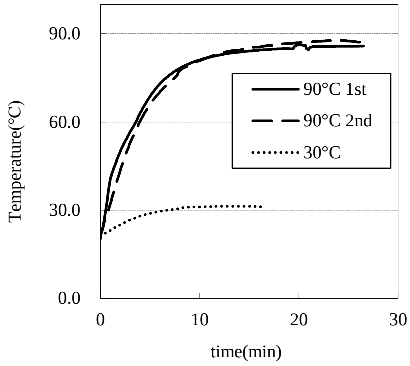

Fig. 1 Relationship between temperature inside the specimen and time

Figure 1 shows the relationship between internal temperature and time when the specimen was placed in a water tank maintained at various temperature. When the specimen was placed in a water tank maintained at 30 °C, it can be seen that 10 minutes after the temperature was applied, the internal temperature reached 30°C and achieved an equilibrium state. When the specimen was placed in a water tank maintained at 90 °C, it can be seen that the internal temperature reached about 90°C 20 minutes after the temperature was applied and achieved an equilibrium state. Therefore, temperature is considered to be adequately applied inside the specimen by these set heating times. Based on these results, the heating time was set to 15 minutes at 30°C, 20 minutes at 60°C, and 25 minutes at 90°C.

3.3 Uniaxial Compression Test

Figure 2 shows a schematic diagram of the Uniaxial compression tester. Table 1 shows the test condition, and table 2 shows the sample condition.

Fig. 2 Uniaxial compression tester

Table 1 Test condition

silica sand No.	No.8
silica sand ratios (%)	30
saturation (%)	30
dry density (Mg/m^3)	1.6

Table 2 Sample condition

Silica sand No. 8 particle density (Mg/m^3)	2.702
Kunigel V1 particle density (Mg/m^3)	2.606
Silica sand No. 8 particle size (mm)	0.08～0.2
Kunigel V1 particle size (mm)	～0.05

The experiment was conducted with reference to JIS A 1216 [5]. The procedure is shown below.

(1) A compression auxiliary device and a rubber sleeve were attached to the specimen. Heat resistant rubber was attached to the top and bottom to prevent water from entering. The specimen was placed in a water tank and heated with warm water.

(2) The heating time was set to 30°C for 15 minutes, 60°C for 20 minutes, and 90°C for 25 minutes.

(3) After applying heat, the uniaxial compression test was performed with the specimen kept in the water tank. At this time, the loading speed was set to 0.4 mm/min. The reason for this is that the bentonite-silica sand mixed specimen used in this test exhibits a very brittle fracture morphology, and this speed allowed for minute deformation up to the fracture to be observed in detail.

(4) The water content of the destroyed specimen was measured.

(5) The maximum compressive stress, the strain at the maximum compressive stress, and the elastic modulus were obtained based on the results.

3.4 Methylene Blue Adsorption Test

In Japan, there are JIS Z 2451 [6], Horiuchi and Takagi [7], Watanabe and Yokoyama [8], and Japan Bentonite Industry Association [9] methods for the methylene blue adsorption amount test. In this test, reference [7] was adopted. The procedure is shown below.

(1) 1.000 g of methylene blue powder was weighed in a crucible and dried in a drying oven for 10 to 26 hours.

(2) The material was removed from the drying oven, let stand to cool in a desiccator for about 1 minute, and then weighed with an electronic balance.

(3) The amount of methylene blue needed was calculated according to the following formula, and the original reagent bottle was used to weigh the medicine packing paper. The dried methylene blue was discarded.

Methylene blue amount (g)

$$=\left(\frac{\mathbf{0.855}}{\textbf{weight after drying(g)}}\right)\times 3.74\times\frac{1}{2} \qquad (1)$$

(4) Methylene blue was transferred to a beaker, ion-exchange water was added, and the mixture was stirred with a glass rod to dissolve methylene blue.

(5) The solution was transferred to a 500 ml brown volumetric flask, and ion-exchanged water was added to a volume of 500 ml.

(6) The solution was allowed to stand overnight with occasional shaking to ensure complete dissolution.

(7) 17 g of sodium diphosphate + hydrate was

weighted and dissolved it in 500 ml of ion-exchanged water.

(8) 50 ml of 2% sodium pyrophosphate solution was placed in a 200 ml conical beaker, and the sample (bentonite) was placed in the solution.

(9) The beaker was covered, stirred gently with a magnetic stirrer, and then boiled gently with the rotor in place.

(10) After allowing to cool, the solution was stirred and gently boiled again.

(11) After allowing the solution to cool to room temperature, 60% of the saturated adsorption amount of methylene blue solution was added and stirred for 30 seconds.

(12) The solution was sampled with continuous stirring and by dropping it onto the filter paper.

(13) Titration was performed by adding 1 ml of methylene blue solution.

(14) Steps (12) and (13) were repeated until a pale blue halo was confirmed.

(15) Titration was performed, and when a pale blue halo was confirmed, the mixture was stirred for 2 minutes and then dropped onto the filter paper. Negative ion-bearing montmorillonite adsorbs positive ion-bearing methylene blue. The halo appears as a blue smear around spots of unabsorbed methylene blue.

(16) Step (15) was repeated until the halo width became 1.5 to 2.0 mm. The halo width was measured three times with a caliper and the average value was used.

(17) The montmorillonite content (methylene blue adsorption amount) was calculated from the amount of methylene blue added when the halo width was about 1.5 to 2.0 mm. The calculation formula is shown below.

Adsorption amount of methylene blue (mmol / 100g) = Methylene blue solution consumption (ml)×2 (2)

(18) The amount of methylene blue adsorbed on 100% montmorillonite Kunigel V1 is 140 mmol / 100g. The montmorillonite content was determined by this ratio. After this, the bentonite-silica sand mixed sample was converted to the value of 100% bentonite. The calculation formula is shown below.

Adsorption amount of methylene blue

$$= \frac{0.50 \times (montmorillonite\ content)}{0.35} \quad (3)$$

0.50 : Mass of sample used for test (g)

0.35 : Mass of bentonite during the test (g)

4. TEST RESULTS AND DISCUSSION

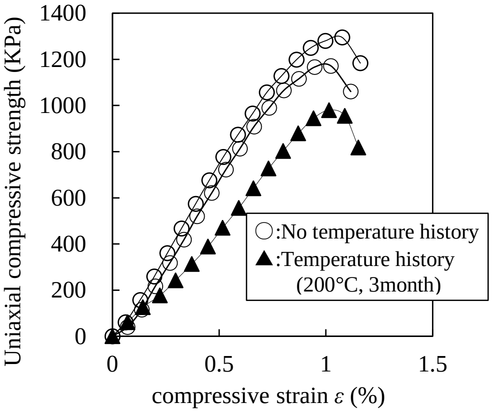

Fig. 3 Maximum compressive stress-compressive strain result

Table 3 compressive strain and elastic modulus

compressive strain (temperature history) ε_f (%)	1.05
Elastic coefficient (temperature history) E_{50} (MN/m^2)	149.3
compressive strain (temperature history) ε_f (%)	0.97
Elastic coefficient (temperature history) E_{50} (MN/m^2)	138.9
compressive strain (no temperature history) ε_f (%)	1.02
Elastic coefficient (no temperature history) E_{50} (MN/m^2)	94.9

Figure 3 shows the changes in the stress-strain curve of the specimen with and without temperature history. The maximum compressive stresses of the samples without temperature history were 1180kN/m^2 and 1300kN/m^2. For the samples with temperature history maximum compressive stresses were 980kN/m^2. It can be seen that the maximum compressive strength of the samples with temperature history is reduced by 200-320kN/m^2 compared to the samples without temperature history. Table 4-7 shows that the montmorillonite content of the sample given a temperature of 200°C decreased by 6.1 to 9.2%. In general, it is said that the swelling property decreases as the montmorillonite content decreases. Takegahara and Masuda [10] stated that the higher the content of montmorillonite, the higher the swelling property, so the energy required to fabricate specimens with the same density increases and the strength increases. On the other hand, in this study, as the specimens were manufactured so that the dry density was constant, it is considered that the compaction energy at the time of preparation decreased and the strength decreased due to the decrease in the montmorillonite content.

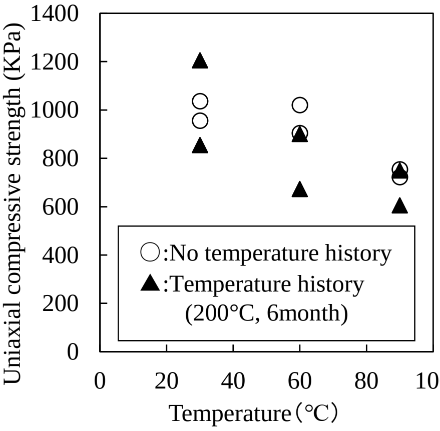

Fig. 4 Relationship between maximum compressive strength and temperature

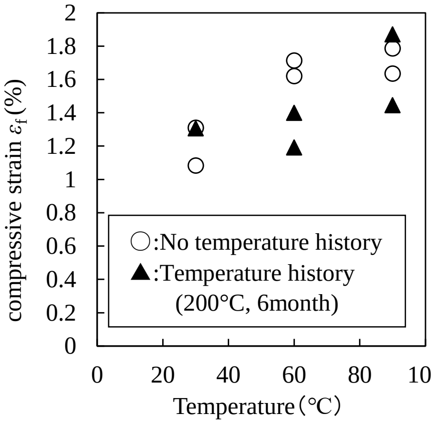

Fig. 5 Relationship between compressive strain and temperature

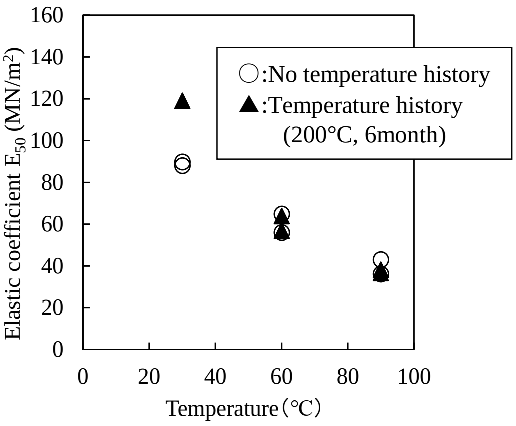

Fig. 6 Relationship between Elastic coefficient and temperature

Table 4 Results of methylene blue adsorption test (No temperature history, no temperature change)

MB solution consumption (ml)	28	29.5
MB adsorption amount (mmol/100g)	56	59
Montmorillonite content (%)	57.1	60.2

Table 5 Results of methylene blue adsorption test (Temperature history 3 months, No temperature change)

MB solution consumption (ml)	25
MB adsorption amount (mmol/100g)	50
Montmorillonite content (%)	51.0

Table 6 Results of methylene blue adsorption test (Temperature history 6 months, Temperature changes 30 degrees)

MB solution consumption (ml)	25
MB adsorption amount (mmol/100g)	50
Montmorillonite content (%)	51.0

Table 7 Results of methylene blue adsorption test (Temperature history 6 months, Temperature changes 90 degrees)

MB solution consumption (ml)	25
MB adsorption amount (mmol/100g)	50
Montmorillonite content (%)	51.0

Figure 4 shows the results of a uniaxial compression test with varying temperatures. It can be seen that the maximum compressive stress decreases with increasing temperature regardless of the presence or absence of temperature history. A possible cause of the decrease in compressive strength due to temperature rise is the effect of cracks caused by thermal expansion of the specimen. Bentonite is generally considered to have very low air permeability. Microcracks may be generated on the surface or inside the material due to the thermal expansion of internal interstitial air and the evaporation of interstitial water caused by the influence of heat. It is considered that the compressive strength decreased due to the decrease in rigidity caused by the inclusion of microcracks. As this effect increases with rising temperature, compressive strength is considered to decrease with increasing temperature. Figure 5 shows the relationship between compressive strain and temperature.

It can be seen that the compressive strain increases as the temperature rises, regardless of whether or not there is a temperature history. Figure 6 shows the Relationship between Elastic coefficient and temperature. It can be seen that the Elastic coefficient decreases as the temperature rises, regardless of whether or not there is a temperature history.

5. CONCLUSION

In this paper, the mechanical behavior of bentonite cushioning material was investigated in consideration of the existence of temperature history and the method of applying temperature in the geological disposal facility. From this, the following conclusions were obtained.

(1) When a temperature history of 200°C for 6 months was provided, the maximum compressive stress and elastic modulus of the specimen decreased, and the montmorillonite content also decreased. It is considered that the decrease in montmorillonite content decreased the compaction energy specimen production. With this, the strength also decreased.

(2) The maximum compressive stress decrease as the temperature of the specimen rises, regardless of the presence or absence of temperature history. The reason for this is considered to be the decrease in rigidity and the decrease in compressive strength due to the effect of cracks caused by thermal expansion of the specimen. As this effect increases with rising temperature, the compressive strength is considered to decrease with increasing temperature.

In actual disposal facilities, the use of bentonite compacted at high pressures is considered from the viewpoint of improving the performance and workability of the cushioning material. The compacted bentonite has a high dry density and is considered to be affected by the decay heat of high-level radioactive waste. Furthermore, it is said that the temperature inside the cushioning material is in the range of 65 to 165°C due to the decay heat. However, in this test, using powdered bentonite compacted after given a temperature history of 200°C, the characteristics of the cushioning material in the actual disposal facility were not reproduced. Therefore, in order to reproduce the cushioning material in an actual disposal facility, it is necessary to apply heat at high temperature and perform a uniaxial compression test after manufacturing the specimen. This time, the temperatures applied after the specimens were set to 30℃, 60℃, and 90℃. In the future, we would like to improve the water tank heater and perform a uniaxial compression tests at higher temperatures.

A swelling test is also planned in parallel with the uniaxial compression test. Future work would investigate the swelling characteristics of bentonite cushioning materials under various conditions, with and without temperature history, and clarify the relationship between strength characteristics and swelling characteristics. We would also like to investigate the mechanical properties of bentonite. Furthermore, we would like to build a database using a methylene blue adsorption amount measurement test results and improve the accuracy of the data.

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