THE SWELLING BEHAVIOUR OF BENTONITE BUFFER MATERIAL CONSIDERING THE EFFECT OF TEMPERATURE

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ABSTRACT: In the nuclear fuel cycle of nuclear power generation, reprocessing generates high-level radioactive waste. Geological disposal is a widely accepted disposal method for high-level radioactive waste. This method can isolate waste from the human environment for a very long time. Geological disposal utilizes bentonite as a buffer material in order to suppress the transfer of radioactive materials to groundwater. In practice, waste may be exposed to a high-temperature environment due to decay heat or geothermal heat, or heated groundwater may permeate the water. Nonetheless, few studies consider the effect of heat on bentonite, and it is difficult to understand the long-term swelling performance of bentonite material under these conditions. Therefore, it is necessary to investigate the effect of temperature on bentonite. In this study, swelling properties of bentonite were investigated by performing a swelling pressure test considering temperature history and temperature change. Results demonstrated that swelling pressure tended to increase with increasing temperature. Furthermore, a threshold value for the equilibrium swelling pressure may exist.

Keywords: bentonite, swelling pressure, temperature, geological disposal, High-level radioactive waste

1. INTRODUCTION

In the nuclear fuel cycle, which reuses nuclear fuel, radioactive effluent is generated when uranium and plutonium are extracted. The mixture of radioactive effluent and glass into canisters is called high-level radioactive waste. This method can isolate waste from the human environment for a very long time. In Japan, geological disposal for the safe disposal of high-level radioactive waste. It is believed that this method can safely dispose of high-level radioactive waste via the multiple barrier system, which combines an engineered barrier system and a natural barrier system. In Japan, bentonite is used as a buffer material [1]. Since bentonite has swelling properties, low permeability, and cation exchange properties, it is expected to fill gaps, control transfer of radioactive material, and control groundwater invasion. In the actual disposal environment, the buffer material is exposed to high temperatures. Because of this, it is necessary to examine the effects high temperature on the buffer. High temperature environments arise from the decay heat of the vitrified body, the geothermal energy of the rock mass, and permeation of heated groundwater. However, there are few studies considering the effect of heat on bentonite, which poses a problem for understanding the material swelling performance of the long-term. Therefore, it is necessary to investigate the effect of temperature on bentonite. In this study, the swelling characteristics of bentonite were investigated using a swelling pressure test and considering the temperature history of the buffer material exposed to high temperatures and the infiltration of heated groundwater.

2. MATERIALS AND METHODS

2.1 Materials

In this study, a 70% bentonite and 30% silica sand mixture was used because of its excellent workability and economy. The mixture comprised Kunigel V1 for bentonite and silica sand with a particle size diameter of 212 μm or less. Table 1 shows the physical properties of Kunigel V1 in [2]. The samples were placed in stainless steel vats and heated in a drying oven at 30 °C, 60 °C and 100 °C for 14 or 90 days, respectively, to obtain the temperature history. Since the water content of the sample after drying was as low as 1 ~ 2%, and the preparation of the sample was difficult, the water content of the sample was adjusted using ion exchange water. The sample was then placed in a desiccator until the reaching nearly uniform water content. The temperature history and test conditions for each heating period are summarized in Tables 2, 3 and 4. The montmorillonite content shown in Tables 2, 3 and 4 is the value of a mixed sample of bentonite and silica sand. The montmorillonite content was determined by the boiling method of methylene blue adsorption determination in [3].

Table 1 The physical properties of Kunigel V1

Properties	Kunigel V1
soil particle density (Mg/m^3)	2.61
montmorillonite content(%)	51.0

Table 2 Test conditions (No history)

Properties	No history
water content (%)	6.95~9.20
void ratio	0.647
degree of saturation (%)	28.3~37.5
montmorillonite content(%)	35.7

Table 3 Test conditions (Heating period of 14 days)

Properties	30°C	60°C	100°C
water content (%)	3.00	1.72~4.00	1.62~6.95
void ratio	0.647	0.647	0.647
degree of saturation (%)	12.2	13.2~16.3	16.7~28.3
montmorillonite content(%)	35.7	34.3	34.3

Table 4 Test conditions (Heating period of 14 days)

Properties	30°C	60°C	100°C
water content (%)	4.00	2.54	4.40
void ratio	0.647	0.647	0.647
degree of saturation (%)	16.4	10.5	10.2
montmorillonite content(%)	32.1	32.9	32.1

2.2 Method for Producing Test Material

Specimens with a dry density of 1.6 Mg/m3, a diameter of 28 mm, and a height of 10 mm were prepared by static consolidation using the mixtures prepared by the above method. Each specimen was treated with a given temperature history described above. The drying density was determined by referring to the design and manufacturing of the buffer material shown in the second report 1. The mass assigned to the specimen was calculated for each test condition such that the dry density was 1.6 Mg/m3 when compacted to the target specimen height. Test equipment consisted of a hydraulic jack, a reaction frame, and a piston, which can load a maximum pressure of 100 MPa. First, the sample was put into a metal mold with an inner diameter of 28 mm. A compaction rod was put into the mold. Then, the piston was lowered to the upper part of the compaction rod. The specimen was then compressed and molded using a hydraulic jack so that the sample height was 10 mm. The compaction pressure was about 8MPa, and the pressure retention time was 10 minutes. After unloading, the compaction rod was removed, and testing was performed with the specimen in the mold.

2.3 Test Method

The main components of the swelling pressure testing equipment are a reaction frame, load cell, compressor, air cylinder, displacement gauge, water tank, temperature control tool, and thermocouple, as shown in the schematic diagram, Fig.1. The piston of the testing machine can load at maximum of 1 tf using compressed air. The load cell is connected to a data logger and a PC, and data can be collected every second. The specimen was moved to the pedestal of the tester for each mold, and the upper and lower pedestals were fixed with screws. A membrane filter and a porous metal were inserted as filter materials between the specimen and the pedestal. The piston was lowered to the top plate of the testing machine, and the vertical displacement was fixed by a clamp knob. The initial vertical pressure was defined as the vertical pressure at this time. The initial vertical pressure was about 50 N. After this, ion exchange water warmed to the target test temperature was supplied to a water tank, and the specimen wetted to start the swelling pressure measurement. The water supply was applied only from the underside of the specimen, as groundwater only permeates from one side of the buffer material in the actual disposal environment. The test water temperature was controlled by a hot water circulator (or a heater and a thermocouple) so that the water temperature was ± 3 °C of the target temperature. Temperature was measured by the thermocouple. The reason why the temperature control was carried out by either a hot water circulation equipment or a heater and a thermocouple is that when the temperature of the test water was high it dropped to less than -3 °C of the target temperature during its transfer through a hose. Therefore, hot water circulation equipment was only used for tests using 30 °C water. At 60 °C and 100 °C, a heater and thermocouple was used. To suppress evaporation from the water surface during testing, an evaporation inhibitor (pp sphere) was set up, and ion exchange water, heated to a target temperature, was injected as needed so that the water level could be maintained at about 50 mm from the upper part of the specimen. In addition, to confirm that the specimen position was maintained during the test, a displacement meter was installed on the upper part of the specimen, and the displacement amount

during the test was measured. A displacement gauge capable of measuring up to a minimum of 0.001 mm was used. The test period was about 1 to 2 weeks, during which time the swelling pressure of bentonite was considered to reach equilibrium. After the completion of the test, each specimen was immediately sliced into 2 mm sections, and the water content distribution at each height was measured. The test results showed that the maximum swelling pressure and the equilibrium swelling pressure correlated with temperature.
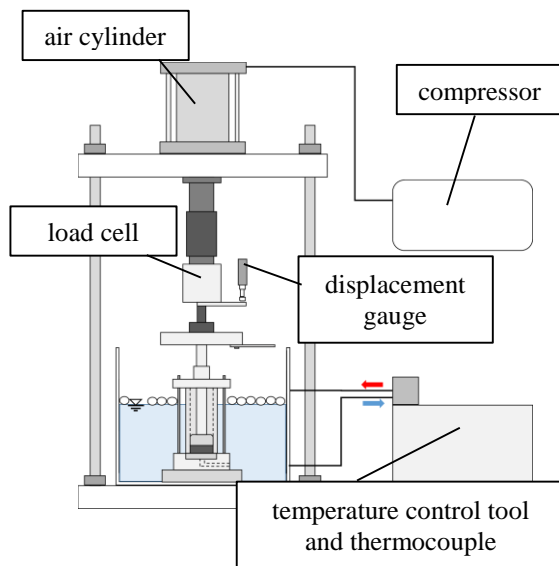
Fig.1 Swelling pressure tester

3. RESULTS AND DISCUSSION

3.1 Changes in Swelling Pressure with Time

Figs.2 to 8 show changes in swelling pressure over time.

In all test results, the swelling pressure rapidly increased after receiving water, decreased after reaching the peak swelling pressure, and then increased again to reach the equilibrium state. Previous studies have confirmed a similar trend [4-6]. Under some test conditions, the swelling pressure at equilibrium was larger than at the initial peak. Pressure peaked about 7 hours after the start of the test at 30 °C and about 0.3 hours after the start of the tests at 60 °C and 90 °C. The slope of the initial swelling pressure graph, which increase with increases temperature, indicates that with higher temperature, the time until the swelling pressure reaches the equilibrium state decreases. In addition, for tests at 90 °C, the swelling pressure after the peak repeatedly increased and decreased by about 100kPa.

That the time until the swelling pressure reaches the equilibrium state becomes shorter as the temperature rises is thought to be facilitated by a faster hydration reaction with rising temperature. Bentonite swells when water molecules enter between the layers of montmorillonite crystals due to the hydration reaction between interlayer cations and solution water. In addition, the hydration reaction is accelerated at higher temperatures. Therefore, the hydration reaction likely advanced more rapidly as the temperature rose. The reaction ended when the hydration number reached the upper limit, and the swelling pressure reached equilibrium state.

The reason that the swelling pressure after the peak repeatedly increased and decreased by about 100kPa at the test temperature of 90 °C is likely caused by the use of a heater and a thermocouple for temperature control of the test water. The heater cools after reaching the target temperature, and when the temperature decreases by about 3 °C from the target temperature, the heater repeatedly activates to control the temperature. Since the time course of the swelling pressure after the peak agrees with the heating and cooling cycles of the heater, swelling pressure likely changes due to the change in the temperature.

3.2 Maximum Swelling Pressure

Maximum swelling pressure increased with increasing temperature under all test conditions. In addition, maximum swelling pressure of samples with temperature history decreased slightly at a test temperature of 30 °C, and increased by about 100 ~ 200kPa at test temperatures of 90 °C and 60 °C. This suggests that the higher the test temperature, the greater the effect on the maximum swelling pressure of samples with temperature history, regardless of the

The increase in maximum swelling pressure is caused by the expansion of the diffusion electric double layer and the increase in osmotic pressure. First, we will discuss the expansion of the diffusive electric double layer. Since the surface of clay minerals is charged, an electric double layer is formed. Debye distance ($1/\kappa$) is defined as a parameter representing the thickness of the electric double layer. Eq. (1) is Debye distance magnitude.

$$\frac{1}{\kappa} = \sqrt{\frac{\varepsilon_r \varepsilon_0 kT}{2e^2 n_0 z^2}} \tag{1}$$

Where e: elementary charge, n_0: ion molar concentration, z: valence of ion, ε_r: dielectric constant, ε_0: dielectric constant of vacuum, k: Boltzmann constant, and T: absolute temperature. Debye distance increases with increasing absolute temperature. Overlapping electric double layers
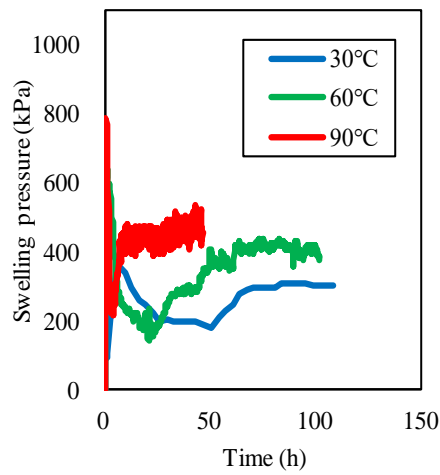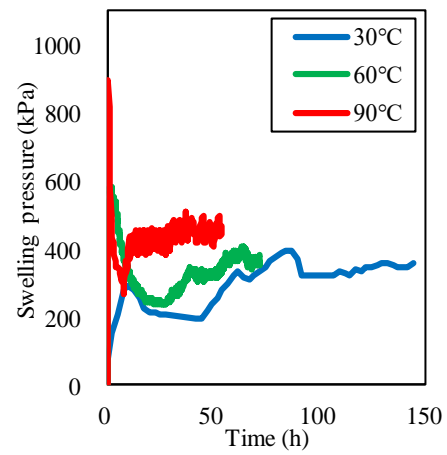
Fig.2   Samples with no history

Fig.5 Heating period of 14 days and temperature history of 100 °C
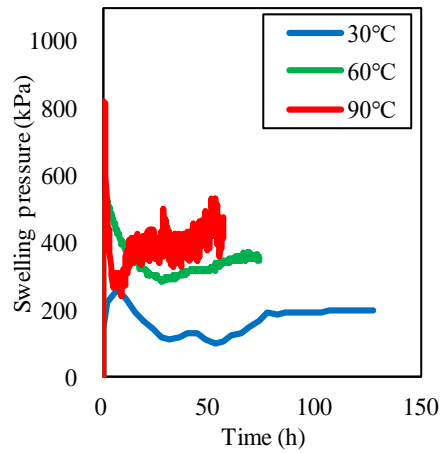

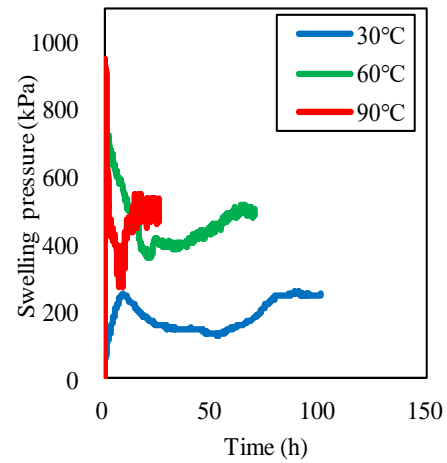Fig.3   Heating period of 14 days and temperature history of 30 °C

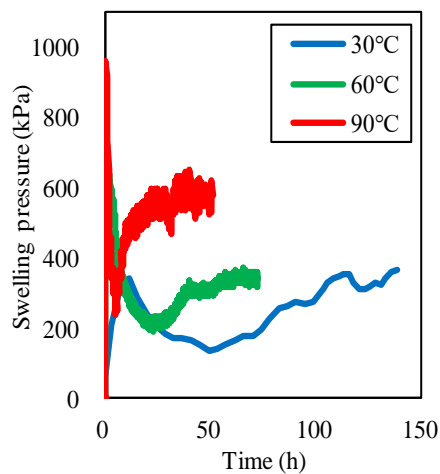Fig.6 Heating period of 90 days and temperature history of 30 °C

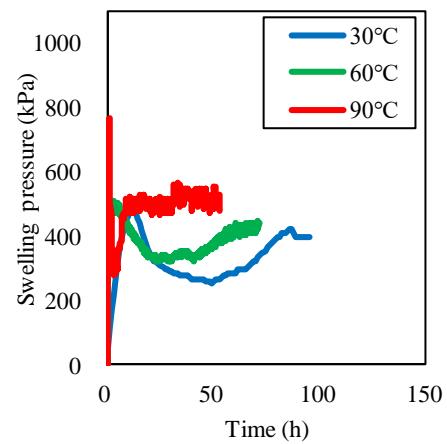Fig.4   Heating period of 14 days and temperature history of 60 °C

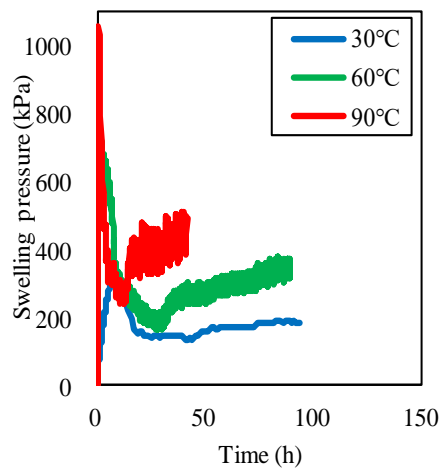Fig.7 Heating period of 90 days and temperature history of 60 °C

Fig.8   Heating period of 14 days and

generate repulsive forces, and as the Debye distance increases, the area where the electric double layers overlap grows larger. Therefore, it is thought that the repulsive force and the swelling pressure both increased.

Secondly, we consider osmotic pressure. Bentonite swells by the hydration reaction of water molecules with interlayer cations. Large osmotic pressure is considered to be generated by the ion concentration difference between the ion exchange water and montmorillonite layers. Since the osmotic pressure increases with increasing temperature, it is likely that the swelling pressure increases because water molecules entry between the layers becomes easier.

The decrease in the maximum swelling pressure of the sample with a temperature history of 30 ℃ is thought to be due to the decrease in the montmorillonite content of the sample with increasing temperature history. As the montmorillonite content decreased by about 3% in the 14- day history period and by about 2% in the 90- day history period, the maximum swelling pressure of the sample during the 90- day history period is thought to be lower than that of the sample without a temperature history. However, it has been reported that clay minerals may be altered by drying if the water content falls below 7% [7-9]. Since the water content of the sample taken out of the drying furnace after the temperature history was as low as 1 ~ 2%, the decrease of montmorillonite content is considered to be due to the alteration of bentonite by drying. Necessary future analysis should include examination of temperature history application to prevent sample drying and clarify the causal of montmorillonite content reduction. Since the maximum swelling pressure increases at the test

temperatures of 60 ℃ and 90 ℃, further collection of data is necessary.

3.3 Equilibrium Swelling Pressure

The equilibrium swelling pressure increased with increasing temperature, and the swelling pressure tended to increase more slowly than the maximum swelling pressure under all test conditions. A sample with a history temperature of 60 ℃ shows a larger equilibrium swelling pressure than a sample without a temperature history. In addition, it was confirmed that the equilibrium swelling pressure of the sample with a temperature history decreased by about 100kPa at the test temperature of 30 ℃.

First, the expansion of the diffusion electric double layer is considered to cause an increase in the equilibrium swelling pressure with the rise in temperature. The previous section explained that the diffusion electric double layer expands with rising temperature, a trend observed even to equilibrium swelling pressure. The repulsive forces are considered to increases due to the expansion of the electric double layer and the equilibrium swelling pressure increases due to the repulsive force of the particles.

The gradual increase in swelling pressure compared with the increase in maximum swelling pressure is thought to be due to the decrease in osmotic pressure and dehydration of interlayer water. Although the osmotic pressure increased with increasing temperature, the osmotic pressure likely decreased because water entered the interlayer and the ion concentration difference between the peripheral solution and the interlayer decreased. As for the dehydration of the interlayer water, the increase in the swelling pressure likely slowed, due to the separation of the interlayer cations and hydrated water molecules with increasing temperature. A TG (thermogravimetric decrease) index exists, which illustrates the relation between temperature and mass. In this, TG is defined as a method to measure the mass of the sample as a function of temperature, while the temperature of the sample is changed according to a fixed program. By this method, weight reduction was confirmed even at temperatures below 100 ° C. in [10]. The higher the test temperature, the more the water molecules in the montmorillonite layers become dehydrated, because higher temperatures enhance dehydration. Therefore, the tendency of the equilibrium swelling pressure to increase became less pronounced the distance between layers decreased. However, since the rate of the hydration reaction and the time until specimen saturated are unknown, it is necessary to estimate from the water content distribution of the specimen in the initial stage of swelling by changing the duration of the

swelling pressure test.

The decrease in equilibrium swelling pressure at a test temperature of 30 °C for samples with a temperature history regardless of the specific temperature history or heating period) is likely caused by the decrease in montmorillonite content due to the temperature history. However, since the swelling pressure increased at a test temperature of 90 °C and a hysteresis temperature of 60 °C, it is necessary to continue testing and data collection.

4. CONCLUSIONS

In this study, the swelling pressure test was used to understand the swelling characteristics of bentonite buffer material, considering temperature history and temperature change.

First, it was confirmed that the swelling pressure rapidly increased at the start of water supply, decreased after reaching the peak swelling pressure, and then increased again to reach the equilibrium state. This tendency was confirmed in all test conditions. It was also confirmed that the slope of the graph in the initial stage of swelling increased and that the time to reach the equilibrium state was shorter with increasing test temperature.

Second, the maximum swelling pressure tended to increase as the temperature of the test water increased. Expansion of the diffusion electric double layer and an increase in the osmotic pressure are considered as important factors for this.

Thirdly, the equilibrium swelling pressure tended to increase with increasing temperature of the test water. This is likely due to the expansion of the diffusion electric double layer.

Finally, various condition tests will be conducted in the future to understand the swelling characteristics of bentonite.

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