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EXPERIMENTAL STUDY ON SWELLING CHARACTERISTICS OF BENTONITE BUFFER MATERIAL CONSIDERING VARIOUS INITIAL CONDITIONS

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ABSTRACT: Swelling pressure tests have been conducted to determine the swelling characteristics of bentonite, which is under consideration for use as a buffer material in the geological disposal of high-level radioactive waste. In a geological disposal environment, the bentonite buffer is subjected to elevated temperatures driven by the waste's decay heat, while its mechanical behavior evolves as groundwater infiltrates and saturation progresses. Therefore, a quantitative understanding of swelling behavior under coupled thermo-hydraulic conditions is essential for designing and optimizing buffer specifications. In this study, swelling pressure tests were conducted on specimens with various initial water contents and temperatures to elucidate the relationship between the time-dependent behavior of swelling pressure and heterogeneous internal state changes. These studies confirmed that there was no clear dependence of time-dependent swelling pressure on initial water content, although transient differences before equilibrium indicated its influence during saturation. Temperature minimally affected overall swelling trends but increased hydraulic conductivity. These findings quantitatively describe the thermo-hydraulic behavior of bentonite buffers and guide the design and optimization of engineered barrier systems in geological disposal.

Keywords: Bentonite, Swelling Pressure, Temperature, Initial Water Content

1. INTRODUCTION

Geological disposal has been considered as a promising disposal method for long-lived radioactive waste, such as the high-level radioactive waste generated from the nuclear fuel cycle in 1950s Japan. To date, scientific and technological studies have investigated the safety and feasibility of geological disposal. The objective of geological disposal is to isolate a geological repository from the human environment physically. This is achieved by constructing a multi-barrier system, in which engineering measures (engineered barriers) are applied to enhance confinement within deep underground rock mass (natural barriers) that exhibit isolation properties [1].

While nuclear power plants are designed to operate for a period of about 40 years, geological disposal is required to ensure long-term safety for tens of thousands of years, considering the half-life of radioactive nuclides.

In geological disposal, bentonite is used as a buffer between the vitrified waste and the bedrock because of its high swelling and low permeability. The buffer material is expected to form self-sealing barriers to water channels by swelling, provide stable support for the overpack, and relieve stress when the rock mass is subjected to creep, thereby ensuring and maintaining performance over the long term. Because of this, attempts have been made to understand the various material properties of bentonite, such as swelling pressure as determined through swelling pressure tests [2-6].

The swelling pressure test measures the pressure generated when water is supplied to a compacted, unsaturated bentonite with a fixed volume of specimen. The swelling pressure measured by the swelling pressure test increases monotonically, or it increases once, then decreases, and then increases again with the passage of time after the start of water application. This indicates that the change in swelling pressure over time differs depending on the initial water content, even under the same dry density conditions [7]. The swelling behavior that occurs during the transition from the unsaturated state to the saturated state is a coupled hydraulic and mechanical behavior that is extremely complex.

In geological disposal, bentonite buffer material is unsaturated when the engineered barrier is installed and is exposed to a high-temperature environment due to decay heat from the waste body. At the same time, the bentonite buffer absorbs water as it is infiltrated by groundwater from the surrounding area, gradually shifting to a saturated state while its mechanical properties change. Thermal, hydraulic, and mechanical factors interact with each other in the bentonite buffer material, forming so-called coupled thermo-hydro-mechanical (THM) processes. To evaluate the long-term mechanical performance of buffer materials, it is essential to understand their mechanical behavior, especially at high temperatures. Therefore, when bentonite is used as an engineered barrier, the effect of heat on the material properties must be considered in the evaluation. On the other hand, few studies have specifically regarded thermal effects, and this has not led to a detailed understanding of swelling characteristics.

In this study, swelling pressure tests were conducted on Na-bentonite and silica sand mixtures compacted to a dry density of 1.6 Mg/m^3 at different initial water contents and temperatures. The purpose of this study was to construct a database based on the test results and to experimentally understand the effects of each initial condition on the change in swelling pressure over time.

2. RESEARCH SIGNIFICANCE

Long-term evaluations of the mechanical performance of bentonite buffer materials are greatly affected by the mechanical state during transient periods of continuous high-temperature environments. Therefore, it is essential that the characteristics of coupled phenomena, including thermal effects, be properly considered in the design of engineered barriers using bentonite.

The results of this research will contribute to the expansion of the basic database for the specification and design of bentonite buffer materials and can also be used as reliable test data to verify the validity of numerical analysis, contributing to the development of rational performance evaluation methods.

3. MATERIAL AND METHODS

3.1 Materials

This study used KuniGel V-1, a sodium-type bentonite from Tsukinuno Mine, Japan. The swelling properties of this bentonite have been widely studied [8]. Table 1 shows the physical properties and chemical composition of KuniGel V-1. It is composed mainly of montmorillonite (about 55%), quartz, feldspar, calcite, and clinoptilolite. The cation exchange capacity is 80.0 (meq/100g), and the major exchange cations are Na$^+$ (53.4 meq/100g), Ca^{2+} (9.3 meq/100g), Mg^{2+} (0.4 meq/100g), and K$^+$ (0.8 meq/100g). Soil particle density was measured by measuring the volume excluded by gas expansion using a Micromeritics helium pycnometer (Accu Pyc) to obtain the true volume, which resulted in a density of 2.605 Mg/m^3, with a plastic limit of 26.4 % and a liquid limit of 436.4 %.

The sample used is bentonite mixed with Tohoku silica sand No. 8, mined from the Ōishida Mine, Japan. Mixing sand with bentonite enhances both thermal conductivity and compaction properties, making this mixed buffer material an important subject of investigation [1]. The soil particle density of the Tohoku silica sand was 2.702 Mg/m³.

Table 1. Basic properties of KuniGel V-1

Properties	KuniGel V-1
Cation type	Na
Main minerals	Smectite, Quartz, Calcite, Zeolite, Plagioclase
Soil particle density (Mg/m^3)	2.605
Liquid limit (%)	436.4
Plastic limit (%)	26.4
Montmorillonite content (%)	55
Cation exchange capacity (meq/100g)	80.0
Extracted cation (meq/100g)	Na$^+$(53.4), Ca^{2+}(9.3), K$^+$(0.4), Mg^{2+}(0.8)

Table 2. Test Conditions

Test case	Dry density (Mg/m^3)	Water content (%)	Temperature (°C)
A-1-1	1.6	7.4	30
A-1-2	1.6	6.9	30
A-2-1	1.6	12.3	30
A-2-2	1.6	12.3	30
A-3-1	1.6	17.8	30
A-3-2	1.6	17.2	30
B-1-1	1.6	7.0	60
B-1-2	1.6	7.1	60
B-1-3	1.6	6.8	60
B-2-1	1.6	12.5	60
B-2-2	1.6	12.5	60
B-3-1	1.6	17.5	60
B-3-2	1.6	18.0	60
C-1-1	1.6	7.0	90
C-1-2	1.6	7.1	90
C-1-3	1.6	6.8	90
C-2-1	1.6	12.6	90
C-2-3	1.6	12.9	90
C-3-1	1.6	17.6	90
C-3-2	1.6	17.6	90

3.2 Sample preparation

All samples used in the swelling pressure tests were bentonite-silica sand mixtures with a bentonite content of 70%. The natural water content of the bentonite mixture was approximately 7%. To

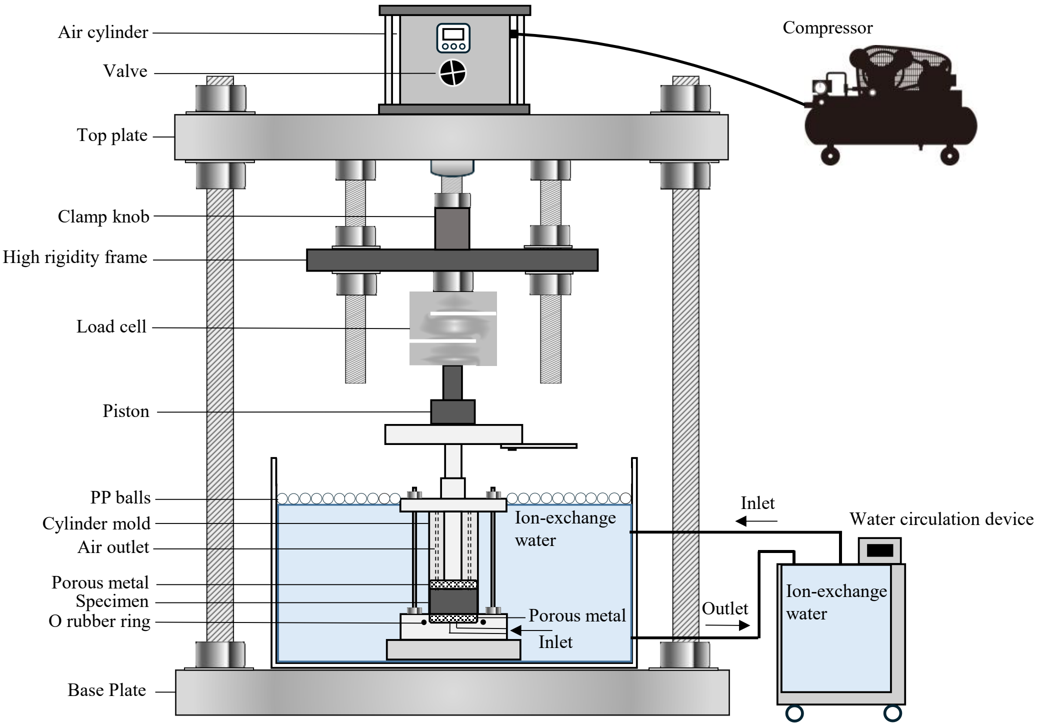

Fig. 1 Schematic diagram of the swelling pressure test apparatus

compare the change over time in swelling pressure at different initial water contents, this test was conducted twice at higher initial water contents of 12% and 17%. In adjusting the initial water content, water was added by misting ion-exchanged water. To prevent water droplets from accumulating, the water content of the sample was gradually increased while alternating between kneading and water addition.

Specimen details for the swelling pressure tests are presented in Table 2. The water-adjusted cylindrical specimens used in the swelling pressure tests were prepared by static compaction with a dry density of 1.6 Mg/m^3, a diameter of 28 mm, and a height of 10 mm. To prepare a specimen, the sample was first placed in a metal mold with an inner diameter of 28 mm, and a stick was inserted into the mold. A piston was then lowered into the top of the rod, and the specimen was compacted with a hydraulic jack so that its height was 10 mm. The compaction pressure was approximately 6 to 8 MPa, depending on the initial water content, and the pressurization holding time was 10 minutes. After unloading, the compaction rod was removed, and the specimens were placed in the mold for testing.

3.3 Testing methodology

The main components of the swelling pressure test apparatus are the reaction frame, load cell, compressor, air cylinder, water tank, temperature control device, and thermocouple. Figure 1 shows a schematic diagram of the swelling pressure test apparatus. The load cell is connected to a data logger and PC, and data can be collected every second. The specimen was transferred to the pedestal of the testing machine one mold at a time, and the upper and lower pedestals were fixed with screws. Membrane filters and porous metal were inserted between the specimen and the pedestal as filter materials. To prevent swelling and deformation of the specimen during the test, the piston was lowered onto the top plate of the testing machine, and vertical displacement was fixed with a clamp knob. The vertical load at this time was taken as the initial vertical load, which was approximately 0.08 MPa. The swelling pressure was then measured by supplying ion-exchanged water to the tank. Water was supplied only from the underside of the specimen to expel air from inside the specimen. Ion-exchanged water was used as the test water, and the test temperature was set at 30°C,60°C, and 90°C.

The water temperature was controlled to be ±1°C above the target temperature, and the temperature was measured using thermocouples. The water level was maintained at approximately 100 mm from the top of the specimen. The test period was about one week, when the swelling pressure of the bentonite was expected to reach equilibrium.

4. EXPERIMENTAL RESULTS

Figures 2 to 10 show the change over time of swelling pressure for each initial water content and temperature. In this study, swelling pressure tests were conducted multiple times under each condition, which were categorized based on the initial water content as follows: 7% was defined as small, 12% as medium, and 17% as large.

4.1 Effect of initial water content on swelling pressure change over time

Figures 2 to 4 show the change over time of swelling pressure for each initial water content at 30°C. In the initial water content is relatively small, swelling pressure increased, then temporarily decreased, and then increased again. On the other hand, as the initial water content increases, the swelling pressure did not decrease under the condition of low initial water content, and it increased relatively slowly. Furthermore, where the initial water content was large, the swelling pressure tended to increase monotonically, and the change over time in the swelling pressure differed depending on the initial water content. The final equilibrium swelling pressure was about 0.4 MPa to 0.6 MPa. The trend of monotonically increasing swelling pressure with increasing initial water content and the equilibrium swelling pressure were similar to the results of previous studies conducted on mixed KuniGel V-1 and silica sand samples and under the same dry density conditions [7].

The differences in the time-dependent behavior of swelling pressure with varying initial water contents are caused by the continuous occurrence of water absorption and compressive behavior inside the specimen, and the presence or absence of wetting-induced collapse in the unsaturated region and its magnitude have a significant impact. Since the swelling pressure test is conducted under a constant-volume condition, both swelling and compression are expected to occur continuously within the specimen during water uptake [9]. Takayama investigated the temporal variations in swelling pressure and wet density distribution using X-ray CT observations, and reported that, under low initial water content conditions, a temporary decrease in swelling pressure was accompanied by a transient reduction in wet density near the infiltration surface [10]. This reduction in wet density indicates a decrease in dry density, suggesting local expansion near the infiltration surface and compressive deformation in the opposite region. These results imply that the rapid reduction in suction, which had acted as a negative confining stress, may have induced localized structural collapse and subsequent compression. Also, Zeng analyzed the microstructure of compacted granular bentonite specimens with different initial water contents using X-ray micro-computed tomography (micro-CT) and mercury intrusion porosimetry (MIP) [11]. They reported that specimens with lower initial water contents exhibited a higher proportion of macropores and were therefore more susceptible to wetting-induced collapse during subsequent hydration. Although involving a different type of bentonite, it suggests that suction acting as a negative confining stress decreased rapidly upon wetting, leading to localized structural collapse and the associated compression.

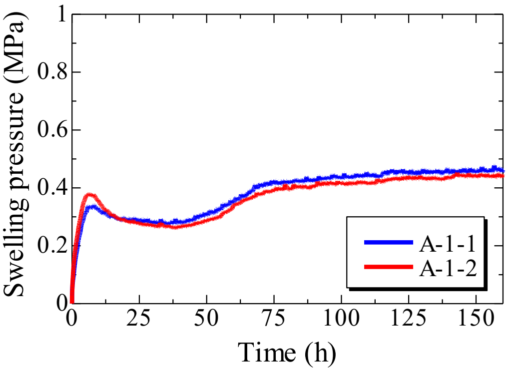

Fig. 2 Time history of swelling pressure (Initial water content: small, Temperature: 30°C)

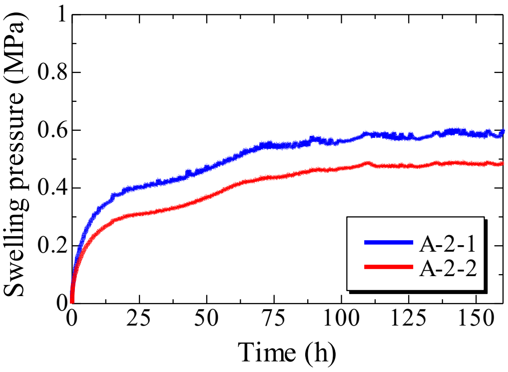

Fig. 3 Time history of swelling pressure (Initial water content: middle, Temperature: 30°C)

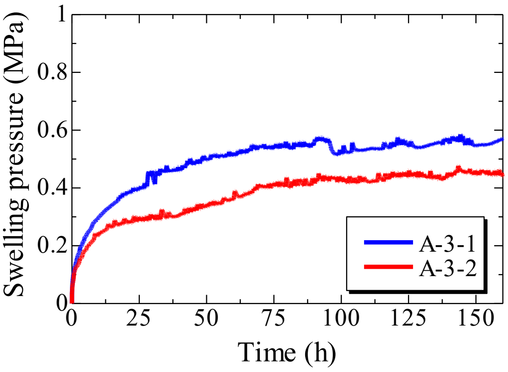

Fig. 4 Time history of swelling pressure (Initial water content: large, Temperature: 30°C)

On the other hand, under higher initial water content conditions, the amount of water absorption is relatively small, resulting in a weaker influence of collapse. Consequently, the swelling pressure tends to

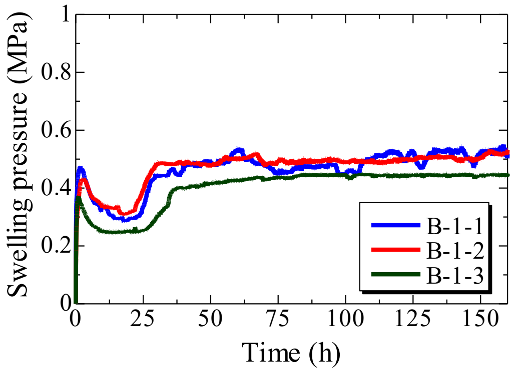

Fig. 5 Time history of swelling pressure (Initial water content: small, Temperature: 60°C)

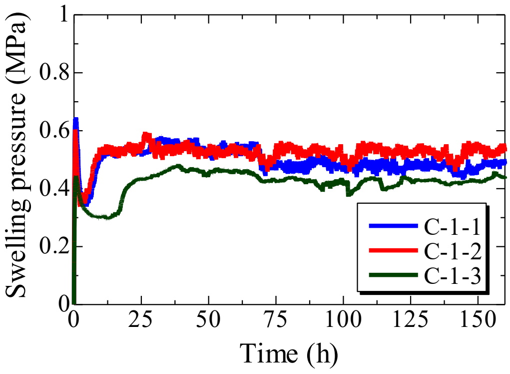

Fig. 8 Time history of swelling pressure (Initial water content: small, Temperature: 90°C)

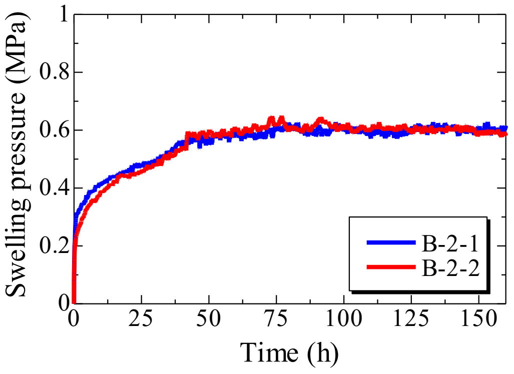

Fig. 6 Time history of swelling pressure (Initial water content: middle, Temperature: 60°C)

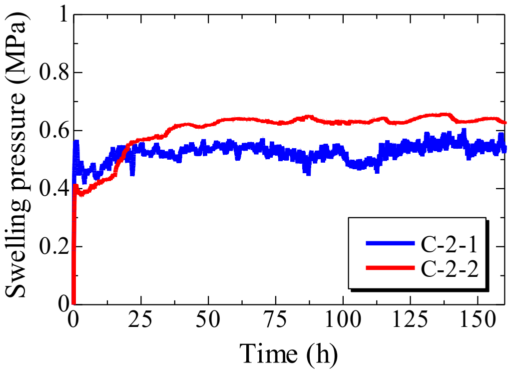

Fig. 9 Time history of swelling pressure (Initial water content: middle, Temperature: 90°C)

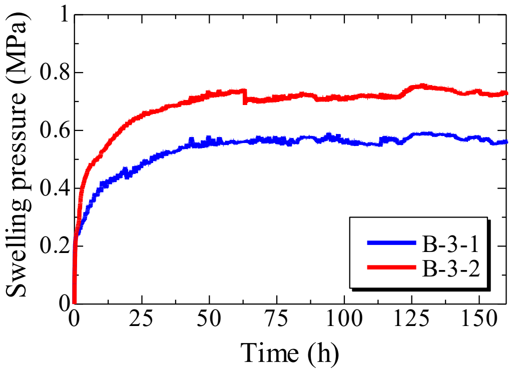

Fig. 7 Time history of swelling pressure (Initial water content: large, Temperature: 60°C)

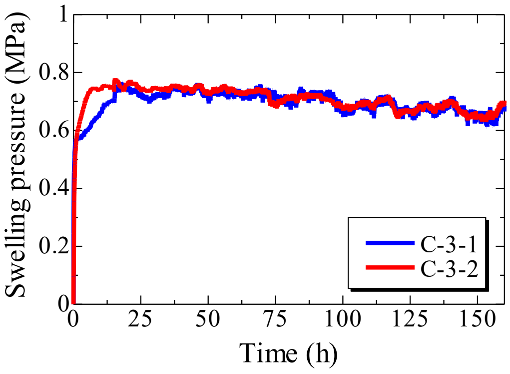

Fig. 10 Time history of swelling pressure (Initial water content: large, Temperature: 90°C)

increase monotonically. This indicates that, above a certain threshold of initial water content, the effect of collapse becomes negligible, and the deformation mechanism transitions to a continuous process of swelling accompanied by compression. Since no clear dependence of the final equilibrium swelling pressure on the initial water content was observed, it is suggested that variations in the initial water content significantly affect the swelling behavior throughout the transition from the unsaturated to the saturated state.

4.2 Effect of temperature on swelling pressure change over time

Figures 5 to 10 show the change over time of swelling pressure for each initial water content at temperatures of 60°C and 90°C. In the initial water content is relatively small, the swelling pressure increased after the start of water supply, reached a peak, temporarily decreased, and then increased again to reach equilibrium for all temperature conditions. In particular, the initial swelling pressure peaked after the start of the water supply and tended to increase as the temperature increased. In addition, the time required for the swelling pressure to reach the equilibrium state tended to be shortened at higher temperatures. When the test temperature was 30°C, it took about 60 hours for the swelling pressure to increase again after the initial peak, while at 60°C it took about 24 hours, and at 90°C it took about 10 hours. These trends were also observed at high initial water contents.

The increase in the initial swelling pressure peak is caused by the increase in osmotic pressure. Kanazawa reported that the increase in maximum swelling pressure in a swelling pressure test that considered temperature history and temperature variation was due to the increase in osmotic pressure and the expansion of the diffusion electric double layer [12]. In the swelling pressure test, ion-exchanged water was used, and it is considered that the concentration difference between the montmorillonite interlayer and the external solution caused water to move into the interlayer, and the increase in temperature accelerated this movement, resulting in the increase in the initial peak. In addition, Towhata investigated the effects of elevated temperature on the volume-change behavior of clays [13]. They reported that, in swelling clays with low permeability, thermal expansion of pore water results in an instantaneous increase in sample volume. In this study, specimens with high initial water content exhibited pore spaces saturated with considerable amounts of pore water. As a result, the thermal expansion of pore water with increasing temperature may have contributed to the increase in initial swelling pressure.

On the other hand, the shortening of the time required to reach equilibrium is caused by the increase in hydraulic conductivity due to the rise in temperature. Matsumoto conducted hydraulic permeability tests on bentonite with temperature as a parameter and reported that at 60°C and 90°C, the hydraulic conductivity increased approximately 2 to 3 times compared to that at room temperature, which they attributed to temperature changes in the density and viscosity of water [14]. In this test, the viscosity of the water decreased as the temperature increased, and the permeability of the bentonite increased, which is thought to have shortened the time required to reach equilibrium. In addition, the time required to reach equilibrium was significantly shorter at 90 °C, suggesting that, in addition to the improvement in permeability, the effect of osmotic pressure may have facilitated water movement between the layers.

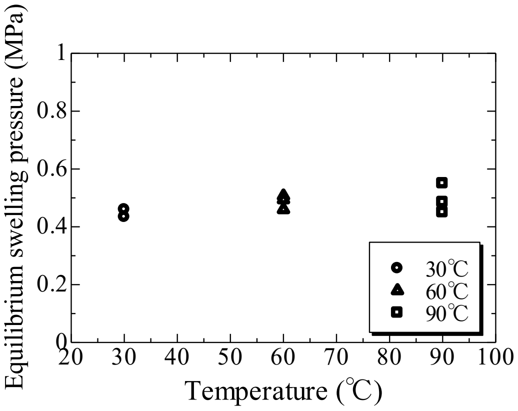

Fig. 11 Equilibrium swelling pressure at each temperature (Initial water content: small)

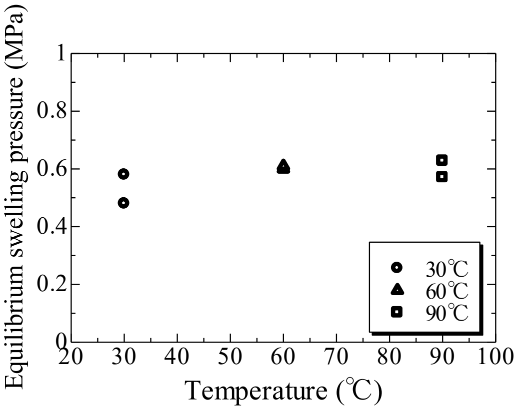

Fig. 12 Equilibrium swelling pressure at each temperature (Initial water content: middle)

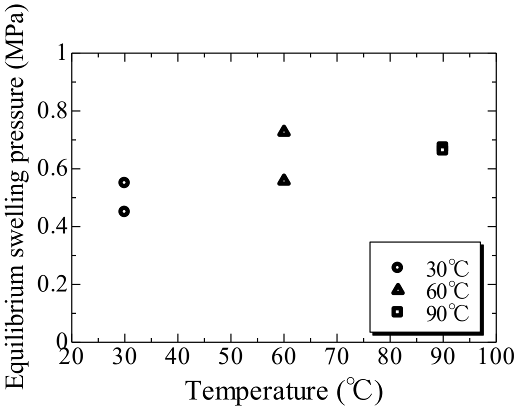

Fig. 13 Equilibrium swelling pressure at each temperature (Initial water content: large)

4.3 Effect of initial water content on swelling pressure change over time

Figures 11 to 13 show the equilibrium swelling pressure at each temperature for different initial water contents. Under all test conditions, the equilibrium swelling pressure tended to increase with increasing temperature. Specifically, it increased by approximately 0.05 to 0.1 MPa as the temperature rose. On the other hand, the equilibrium swelling pressures at 60°C and 90°C were nearly the same, suggesting that the effect of temperature on equilibrium swelling pressure is relatively small when the effective clay density is approximately 1.36 Mg/m³.

Previous studies have pointed out the following four main factors in the effect of temperature on equilibrium swelling pressure [15]. 1) change in the state of adsorbed water in the interlayer, 2) expansion of the diffused electric double layer, 3) volume expansion of free water, and 4) thermal expansion of mineral particles.

Bentonite consists of montmorillonite as its main component, together with other minerals. In the dry state, montmorillonite exists as secondary particles formed by the stacking of multiple primary particles, and these secondary particles, together with the other minerals, form the skeletal structure. When water infiltrates, water is taken into the interlayer spaces, causing swelling, and the layered structure gradually collapses, resulting in changes to the skeletal structure. As water uptake progresses, the swollen montmorillonite begins to fill the pore spaces within the material, and under saturated conditions, these pores are almost completely occupied. Thus, as saturation is approached, the influence of temperature changes on the volumetric behavior of pore water and the adsorption behavior of interlayer water becomes relatively small compared with the unsaturated state, where free water is dominant. The observed increase in equilibrium swelling pressure with rising temperature can be attributed to the expansion of the diffuse electric double layer and the thermal expansion of the mineral particles. The surface of clay minerals carries either a positive or negative charge, attracting oppositely charged ions and forming an electric double layer around the particles. The Debye distance ($1/\kappa$) is defined as a parameter representing the thickness of the electric double layer and is expressed by equation (1).

$$\frac{1}{\boldsymbol{\kappa}} = \sqrt{\frac{\varepsilon_r \varepsilon_0 \boldsymbol{k} \boldsymbol{T}}{2\boldsymbol{e}^2 \boldsymbol{n}_0 z^2}} \qquad (1)$$

Here $\boldsymbol{e}$: elementary charge, $\boldsymbol{n}_0$: ion molar concentration, z : valence of ion, ε_r : dielectric constant, ε_0 : dielectric constant of vacuum, $\boldsymbol{k}$: Boltzmann constant, and $\boldsymbol{T}$: absolute temperature. Equation (1) shows that the Debye distance increases as the absolute temperature rises and the molar concentration of ions decreases. The thickness of this electric double layer increases with rising absolute temperature, and when the double layers of adjacent particles overlap, repulsive forces are generated. In the present study, the electric double layer was observed to expand with increasing temperature, leading to an increase in repulsive forces and, consequently, an increase in the equilibrium swelling pressure. However, the increase in the thickness of the electric double layer at 90°C was only about 1.1 times greater than that at 30°C. Therefore, although the effect of enhanced repulsion was evident, it is presumed to have been limited.

In addition, Shimizu conducted consolidation tests on both swollen and unswollen clays under varying temperature conditions and determined the thermal expansion coefficient of each sample based on the relationship between temperature and soil particle density [16]. The results indicated that the thermal expansion coefficient of montmorillonite, a major component of bentonite, is higher than that of other minerals due to the weak bonding between its crystal particles. During the temporal evolution of swelling pressure, swelling and compression induced by water absorption occur continuously within the specimen. In this process, the thermal expansion of mineral particles caused by the rise in temperature is believed to contribute to compression, thereby resulting in an increase in swelling pressure.

5. CONCLUSION

In this study, swelling pressure tests were conducted, considering initial water contents and temperature, to experimentally understand the effect of each initial condition on the change in swelling pressure over time. The following is a summary of the findings.

(1) No clear dependence of the time-dependent evolution of swelling pressure on the initial water content was identified. However, differences in swelling behavior were observed before reaching equilibrium, indicating that the initial water content influences the swelling process during the transition from the unsaturated to the saturated state.

(2) The overall trend of swelling behavior showed little variation with increasing temperature. Nevertheless, the temperature rise enhanced the hydraulic conductivity, thereby shortening the time required to reach the equilibrium swelling pressure. In addition, an increase in osmotic pressure tended to raise the initial peak of the swelling pressure.

(3) Under conditions of low effective clay density, the influence of temperature on the equilibrium swelling pressure increased with rising temperature; however, this effect appeared to be limited. This behavior is likely attributed to the combined effects of the expansion of the diffuse double layer and the thermal expansion of clay

minerals, both of which contribute to variations in equilibrium swelling pressure.

These findings are expected to provide a fundamental database for the development of a coupled THM analysis code aimed at evaluating the long-term performance of bentonite buffer materials in geological disposal systems. Moving forward, additional specimens and testing conditions will be incorporated to further investigate the detailed swelling characteristics.

6. ACKNOWLEDGMENTS

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