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(Received December 31, 2024 / Revised January 14, 2025 / Approved January 30, 2025)

The swelling capacity of bentonite buffers is vital in high-level radioactive waste (HLW) repositories, as it minimizes groundwater infiltration, prevents nuclides from reaching the biosphere, and stabilizes the HLW canisters. As swelling capacity is a function of temperature, understanding bentonite's behavior at approximately 100°C (its presumed upper limit) is essential. However, research on this subject has been scarce. Hence, this study explored the effects of thermal treatment of Ca-bentonite at 105°C under injected water pressures. The results suggest a 19% reduction in "swell index" and a 35%–36% decrease in the total pressure in thermally treated bentonite. The heated samples demonstrated higher hydraulic conductivity than the non-heated ones, indicating potential performance deterioration in controlling the fluid movement. Furthermore, the injected water pressure (base pressure) was not fully transmitted to the sample owing to the difference between the base and back pressures, leading to variations in the total pressure despite maintaining a constant differential pressure. Thus, the results demonstrated a degradation in bentonite's swelling capacity and its compromised role in safe HLW disposal, when subjected to treatment at 105°C. The insights from this research can assist in HLW repository design, while highlighting the need for further research into bentonite's performance.

Keywords: Thermal treatment, Bentonite, Swelling, Hydraulic conductivity, Injected water pressure

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1. Introduction

The safe management of high-level radioactive waste (HLW) is a critical challenge in nuclear waste disposal, requiring the design and operation of repositories that can securely isolate radioactive materials over extended periods. These repositories are typically constructed within stable geological formations and comprise both a natural barrier system (NBS) and an engineered barrier system (EBS) [1, 2]. While the NBS leverages the inherent properties of the surrounding rock to contain canisters with HLWs, the EBS enhances containment through engineered components, making its design as pivotal as the selection of an appropriate geological site.

The EBS consists of several key components: the canister, buffer, backfill, and near-field rock (Fig. 1). Among these, the buffer plays a crucial role by minimizing groundwater inflow, preventing the release of radionuclides, providing mechanical support to the canister, shielding it from external forces, and facilitating the dissipation of decay heat into the surrounding environment. The thermal properties of the buffer directly influence the layout of disposal tunnels and deposition holes within the repository. Specifically, the thermal conductivity of the buffer affects the maximum temperature it reaches during HLW disposal, which in turn dictates the spacing of waste packages. Higher thermal conductivity allows for closer spacing, optimizing the repository's footprint, whereas lower thermal conductivity necessitates greater spacing [3-5].

Bentonite clay is widely used as a buffer material due to its exceptional chemical and physical stability over geological timescales. Its swelling capacity is a key property that contributes to canister support and the inhibition of microbially induced corrosion [6, 7]. This swelling capacity is largely determined by the montmorillonite content and the dominant exchangeable cations present in the clay. Bentonite is classified as either sodium (Na)-type or calcium (Ca)-type, depending on whether Na⁺ or Ca²⁺ ions are predominant [8]. Na-type bentonite exhibits a higher swelling capacity compared to Ca-type, making it the preferred choice for buffer materials in HLW repositories [9, 10].

However, the swelling of bentonite buffers does not occur unless sufficient water is supplied, and the buffer may remain unsaturated or even desiccated due to decay heat before groundwater ingress occurs. This raises concerns about the thermal behavior of bentonite under repository conditions. Some studies report an increase in swelling pressure with temperature in saturated bentonite [11]. In contrast, other studies have observed a decrease in swelling pressure as temperature increases [12-14]. For the safety of nuclear waste disposal, it is important to consider the potential decrease in swelling capacity due to thermal effects.

Despite extensive research on the temperature-dependent swelling behavior of bentonite, there is a notable lack of studies focusing on the effects of prolonged thermal treatment at temperatures around 100°C. This temperature is the maximum allowable for buffer materials in HLW repositories [3, 4]. Most previous investigations have examined the effects of short-term exposure to higher temperatures (≥ 150 °C) [15-18]. This leaves a gap in understanding the long-term performance of bentonite under repository-relevant thermal conditions. This knowledge gap is particularly critical for Ca-type bentonite, which has a lower swelling capacity than Na-type bentonite.

Additionally, when measuring the swelling pressure of bentonite, the total recorded pressure reflects contributions from both the swelling of the clay particles and the applied water pressure. The influence of injected water pressure on the total pressure measurement complicates the accurate assessment of the intrinsic swelling behavior of bentonite. There is a need to distinguish between these two components to properly evaluate the swelling capacity of the buffer material.

Thus, this study investigated how the swelling capacity and hydraulic conductivity of Ca-type bentonite buffer materials change when subjected to thermal treatment at a temperature close to the HLW repository design criterion of 100°C, specifically at 105°C. To account for the possibility



Fig. 1. Schematic diagram of the high-level radioactive waste repository.

that the temperature could actually exceed 100°C, a conservative approach was adopted by specifying 105°C. In addition, variations in the total pressure according to the injected water pressure were investigated. By accounting for the effects of the injected water pressure, this study enables the determination of how the swelling pressure caused solely by the bentonite particles should be properly considered under different injected water pressure conditions, as well as the thermal treatment effect on the bentonite buffer in terms of swelling capacity and hydraulic conductivity.

2. Sample and Experimental Procedure

2.1 Sample

KJ-II bentonite, produced in Gyeongju, Korea, was used for the tests to confirm the swelling capacity. The basic properties of the KJ-II bentonite are listed in Table 1 [19], and its element and mineral constituents are shown in Tables 2 and 3, respectively. The main exchangeable cation of KJ-II is Ca²⁺. To create the thermally treated sample, the KJ-II powder was dried in an oven at 105°C for 10 months. After drying, the thermally treated KJ-II powder was mixed with deionized (DI) water to achieve a water content of 12% and allow the comparison of results with those for non-thermally treated KJ-II at the same water content. The powder and water mixture was placed in a mold and compressed by a press to create a block with a diameter of 5 cm and a thickness of 1 cm. The dry density of the samples was 1,580 kg·m⁻³.

2.2 Measurement of Swell Index

The swell index (SI) measures the swelling capacity of a soaked bentonite sample under unconstrained conditions in water. It is determined by measuring how much a 2 g sample swells in volume when placed in a graduated cylinder. The SI was measured following the ASTM D5890-11 [20] standard under unconstrained water conditions.

2.3 Measurement of Swelling Pressure

To measure the swelling pressure, the non-thermally

Table 1. Basic properties of KJ-II bentonite [19]

	Specific gravity	Liquid limit (%)	Plastic limit (%)	Plasticity index	USCS	Specific surface (m ² /g)	D ₅₀	Organic carbon (%)
KJ-II (Non-thermally treated)	2.71	146.7	28.4	118.3	СН	61.5	3.1	0.25

Table 2. Element composition of thermally treated and non-thermally treated KJ-II

	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	K ₂ O	Na ₂ O	TiO ₂	MnO	Ref.
KJ-II (Non-thermally treated)	58.81	15.17	5.28	5.72	2.70	1.27	1.06	0.67	0.13	[19]
KJ-II (Thermally treated)	63.58	12.89	4.73	5.21	2.49	1.18	0.8	0.6	0.11	

Table 3. Mineral composition of thermally treated and non-thermally treated KJ-II

	Montmorillonite	Albite	Quartz	Cristobalite	Calcite	Heulandite	Reference
KJ-II (Non-thermally treated)	61.9	20.9	5.3	4.1	4.8	3	[19]
KJ-II (Thermally treated)	41.4	12.4	22.4	12.1	8	3.8	

treated sample was first placed into an oedometer cell. For the swell test, a force of 0.5 kN was applied vertically to the sample by the ram to ensure full contact with the sample, while all outlets were open to atmospheric pressure to allow air pushed out by the ram to escape (Fig. 2). After ram contact, the base pressure pump with 0.25% volume accuracy of the measured value and 0.15% pressure accuracy of the full range (3 MPa) injected water at 0.2 MPa into the bottom of the sample. The load cell measured the total force exerted by the sample swelling from the injected water; these values were obtained until force convergence. This process was repeated for the thermally treated sample, using an initial base pressure of 0.2 MPa. Especially, for the thermally treated sample, once the measured force stabilized at each pressure, the water pressure was incrementally raised to 0.5, 1.0, and 1.5 MPa to confirm how much water

base pressure is delivered to the load cell depending on the base pressure applied to the sample. The total pressure was calculated using the force measurements from the load cell.

2.4 Measurement of Hydraulic Conductivity

After measuring the swelling pressure, the hydraulic conductivity for the thermally treated sample was determined using the same setup by applying back pressure using the back pressure pump with the same specifications as the base pressure pump. After the force measured by the load cell was converged when the base pressure was 1.5 MPa, the outlet valve connected to the atmosphere was closed, and 0.5 MPa was applied on top of the sample by the back-pressure pump. When the load cell forces under each back-pressure condition stabilized, the back pressure



Fig. 2. Schematic of the experimental setup for measuring swelling pressure and hydraulic conductivity.

was incrementally increased to 0.7 and 1 MPa. The hydraulic conductivity was calculated using Darcy's law, based on the volume change of the back-pressure pump and the differential pressure between the base and back pressure.

3. Results

3.1 Swell Index

The SI of the thermally treated sample decreased by 19% from 6.8 to 5.5 mL/2 g (Fig. 3), indicating that the swelling capacity of the sample was reduced by the thermal treatment.



Fig. 3. Comparison of swell index between (a) non-thermally treated and (b) thermally treated samples.

3.2 Swelling Pressure

The total pressure of the non-thermally treated sample with a dry density of 1,580 kg \cdot m⁻³ was approximately 2.25

MPa when the base pressure of 0.2 MPa was applied (Fig. 4). However, under the same injected water pressure, the total pressure for the thermally treated sample was lower at 1.43 MPa, a decrease of 36% compared with that of the



Fig. 4. Results of measuring the total pressure of the sample.

non-thermally treated sample. Compared to the total pressure result of approximately 2.85 MPa reported by Kim et al. (2021b) [21] for the non-thermally treated sample with a dry density of 1,600 kg·m⁻³ at a base pressure of 1 MPa, the total pressure for the thermally treated sample decreased by 35% to 1.85 MPa (Fig. 4). The effect of thermal treatment was confirmed by the discrepancy in swelling pressures. Even if swelling of the sample does not occur, the swell index does not become 0 due to the volume of the sample itself, so the rate of decrease in swelling pressure is greater than the rate of decrease in swell index.

For each base pressure of 0.2, 0.5, 1, and 1.5 MPa, the total pressures for the thermally treated sample were 1.43, 1.61, 1.85, and 2.09 MPa, respectively (Fig. 5). Assuming that the swelling pressure of the thermally treated sample is 1.23 MPa (excluding the base pressure) at a base pressure of 0.2 MPa, the actual applied base pressures at 0.5, 1, and 1.5 MPa were 0.37, 0.62, and 0.85 MPa, respectively (Fig. 6). In other words, only 74, 62, and 57% of the 0.5, 1, and 1.5 MPa base pressures, respectively, were effectively applied.

When the increase in the injected base pressure was 0.3 MPa, the ratio of the actual pressure increase was higher than when the increase in the injected base pressure was 0.5 MPa (Fig. 7). Actually, applied base pressure to the



Fig. 5. Results of measuring the pressure of the thermally treated sample with the base pressure: (a) total pressure, and (b) normalized total pressure.



Fig. 6. Actual base pressure to the thermally treated sample measured by the load cell with base pressure.

total pressure was 0.17 MPa which is 57% of the increase amount of base pressure of 0.3 MPa. When the base pressure increased from 0.5 to 1 MPa and from 1 to 1.5 MPa, the applied base pressures to the total pressure were 0.246, and 0.233 MPa, respectively, which are approximately 49% and 47% of the increased amount of base pressure of 0.5 MPa. Even though the increased amount of base pressure was the same as 0.5 MPa, the actual base pressure was different. The higher the base pressure was, the more the



Fig. 7. Increased pressure amount of the thermally treated sample with increase in amount of base pressure.



Fig. 8. Total pressure of the thermally treated sample measured by the load cell at the same difference between base and back pressures. (Legend of Base means when the back pressure is 0).

actual base pressure applied became less than the amount of increased pressure. When the difference between base pressure and back pressure is 0.5 MPa and 1 MPa, respectively, the pressure measured by the load cell was higher when back pressure was applied to the sample than when the back pressure was atmospheric (Fig. 8).

3.3 Hydraulic Conductivity

Regardless of the hydraulic head difference when the



Fig. 9. Measurement of flow volume for thermally treated sample with base and back pressures.

base pressure was constant at 1.5 MPa, the hydraulic conductivity of the thermally treated sample remained relatively stable, measuring 6.21×10^{-13} , 6.42×10^{-13} , and 6.11×10^{-13} m·s⁻¹ when the back pressure was 0.5, 0.7, and 1 MPa, respectively (Fig. 9). The hydraulic conductivity of the thermally treated sample was higher than that of the non-thermally treated sample, with a dry density of approximately 1,600 kg·m⁻³, and 3.5×10^{-13} m·s⁻¹ [19], which indicates that the swelling capacity of the thermally treated sample was reduced.

4. Discussion

4.1 Reduction of Swelling by Thermal Treatment

The swelling mechanisms are classified into two mechanisms. One is crystalline swelling, and the other is swelling by hydration in the diffusion double layer (DDL). Since the swelling in the DDL is affected by the spacing of aggregates, the swelling pressure of the bentonite depends on the crystalline swelling in the particles for block-type bentonite with a high dry density such as a buffer that has low spacing of aggregates [13, 22].



Fig. 10. Schematic of the structure of montmorillonite composed of several silicates.

The crystalline swelling occurs when water is adsorbed to the surface of the basal crystal [23], and it is regulated by maintaining equilibrium between potent electrostatic attraction and repulsive hydration forces [24-29]. However, the repulsive hydration force is reduced by heat treatment. Furthermore, when bentonite undergoes prolonged hightemperature thermal treatment in a dry state, the subsequent reaction with water results in a decrease in the repulsive hydration force compared to that before undergoing hightemperature thermal treatment.

The crystalline swelling capacity can be predicted roughly by the variation of the basal spacing of the montmorillonite (Fig. 10). For the thermally treated sample at 105°C for 10 months, the basal spacing was decreased from 1.5 to 1.43 nm (Fig. 11). Also, the basal spacing was decreased from 1.45 to 0.98 nm by thermal treatment at 200°C for 27 months [18], and similarly, thermally treated bentonite up to a temperature of 650°C also exhibits a decrease in basal spacing from 1.5 to 0.98 nm [30]. Decreased basal spacing means decreased interlayer induced by equivalent force changes between silicate layers. According to the results of the basal spacing changes depending on the thermal treatment temperature, it can be observed that changes in basal spacing occur up to 200°C (Fig. 11). Additionally, it



Fig. 11. Basal spacing variation with temperature of thermal treatment.

is noted that an increase in temperature at 105°C leads to a further decrease in basal spacing. Furthermore, it can be inferred that the hydration capacity is structurally reduced in the interlayers of the aggregates completely by the thermal treatment at 200°C. The SI of the bentonite also decreased by 24% after thermal treatment at 200°C for more than 15 months [18], which indicates that the temperature of thermal treatment may affect the reduction rate of swelling capacity at least up to 200°C to compare the decrease ratio of the SI, 19% when the temperature of thermal treatment is 105°C, and the duration is 10 months.

In addition, the interlayer can be collapsed by thermal treatment, reducing the hydration capacity of cations in the DDL [18]. Through XRF analysis, it was confirmed the exchangeable cations in the thermally treated sample were reduced compared to those in the non-thermally treated sample (Table 2). This collapsed interlayer structure limits swelling compared to untreated bentonite. The prolonged thermal treatment is presumed to prevent full rehydration and the restoration of hydrated cations in the DDL that enable swelling. The interlayer collapse and decreased hydration sites for cations lead to lower swelling.

The mineral composition changes caused by thermal treatment probably contributed to the reduced swelling capacity. Some studies suggest that when montmorillonite undergoes heat treatment, it transforms into a more stable silicate mineral, resulting in a decrease in the montmorillonite content [31, 32]. Likewise, in this study, the substantial montmorillonite content of the sample decreased by 33% from 61.9 to 41.4% after 105°C treatment, leading to an increase in the proportion of other minerals without new phase formation (Tables 2 and 3). Similar variations occurred with 200°C treatment for 27 months [18]. Additionally, 650°C treatment for 4 hours increased silica and alumina contents [17]. This indicates that both prolonged thermal treatment (even at relatively low temperatures above 100°C) and brief exposures to high temperatures can induce structural changes in bentonite, reducing montmorillonite content. The decreased proportion of swelling montmorillonite minerals plausibly contributed to the lower SI and swelling pressure after 105°C thermal treatment.

Above all, the decrease of montmorillonite in the thermally treated sample was clearly confirmed through the XRD intensity results (Fig. 12). Indeed, after prolonged thermal treatment of bentonite, a comparison of its cation exchange capacity (CEC) before and after thermal treatment revealed a decrease in CEC [18]. Since the CEC which is related to hydration affects the swelling, the reduced CEC means the reduced swelling.



Fig. 12. XRD analysis results for the non-thermally and thermally treated samples.

4.2 Swelling Pressure With Injected Water Pressure Conditions

The bentonite swells in the direction in which layers are piled up. Thus, when the bentonite is compressed by a press, the particles are likely to be mainly aligned in the direction of compression, so the swelling pressure in the radial and vertical directions of the buffer material can be measured differently [33] (Fig. 13). In addition, when bentonite particles swell, forces act up and down, and the forces acting up and down between layers cancel each other out in constraint conditions [34]. Therefore, when a water pressure of 1 MPa is applied to the bottom of the sample, all of the water pressure of 1 MPa is not transferred to the load cell. Therefore, it should not be judged that the total pressure minus the water pressure is the swelling pressure of the sample itself [35]. In addition, after saturation, all the interlayers in the sample are filled with water. At that time, if the base pressure increases, rearrangement of aggregation, and squeeze phenomenon occur, so that the water molecules are exchanged with injected water molecules and the water comes out through outlet at the top of the sample by squeezing effect, which draws damping of pressure [36].



Fig. 13. Schematic of particle swelling in the sample.

Thus, as base pressure increases in an already saturated state, the degree of influence of base pressure on total pressure decreases [37].

5. Conclusions

This study investigated the swelling behavior of thermally treated bentonite and determined how applied hydraulic pressure affects the swelling pressure. The following conclusions can be drawn:

■ The swelling capacity of bentonite decreased, and the hydraulic conductivity increased by thermal treatment at 105°C for 10 months. The increase in hydraulic conductivity indicates a reduction in pore space, thereby confirming a decrease in swelling capacity. Also, in the case of bentonite thermally treated at a temperature between 105 and 200°C, the reduction rate of the swelling capacity may increase with increasing thermal treatment temperature.

- At temperatures above 100°C, structural changes in the bentonite can occur if thermal treatment is prolonged. Conversely, high temperatures might also lead to structural changes in bentonite, even if only briefly applied.
- The hydraulic conductivity remains almost constant regardless of the injected water pressure, but the swelling pressure of bentonite has to be considered according to the injected water pressure. By the swelling mechanism, the swelling pressure among each layer in bentonite is cancelled out in constraint conditions, so the injected water is not fully taken into account in the pressure of the buffer in constraint conditions.

It is thought that this study can contribute to the assessment of potential performance degradation of bentonite buffer in the HLW repository by demonstrating the anticipated decrease in swelling capacity at 105°C.

Conflict of Interest

No potential conflict of interest relevant to this article was reported.

Acknowledgements

This research was supported by the National Research Foundation of Korea (NRF) grant funded by the Korea government (Ministry of Science and ICT) (No. RS-2021-NR056237), and carried out under the KICT Research Program (project no. 20250260-001, Evaluation and Analysis of Radionuclide Release from Support Materials for a High-Level Radioactive Waste Disposal Tunnel) funded by the Ministry of Science and ICT.

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