Evaluation of Water Suction for Compacted Bentonite Buffer Under Elevated Temperature Conditions

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A compacted bentonite buffer is a major component of engineered barrier systems, which are designed for the disposal of high-level radioactive waste. In most countries, the target temperature required to maintain safe functioning is below 100°C. If the target temperature of the compacted bentonite buffer can be increased above 100°C, the disposal area can be dramatically reduced. To increase the target temperature of the buffer, it is necessary to investigate its properties at temperatures above 100°C. Although some studies have investigated thermal-hydraulic properties above 100°C, few have evaluated the water suction of compacted bentonite. This study addresses that knowledge gap by evaluating the water suction variation for compacted Korean bentonite in the 25–150°C range, with initial saturations of 0 and 0.22 under constant saturation conditions. We found that water suction decreased by 5–20% for a temperature increase of 100–150°C.

Keywords: Compacted bentonite buffer, Elevated temperature condition, Water suction

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1. Introduction

Korea's high-level radioactive waste management is based on the deep geological disposal concept known as the KBS-3 type, which has been adopted in Sweden and Finland. The Korean reference disposal system plus (KRS+) concept was also proposed based on the KBS-3 type by considering the properties of spent fuel in Korea [1, 2]. However, when the KBS-3 disposal concept was applied to bedrock conditions, the expected area of the Spent Nuclear Fuel (SNF) repository was estimated to be several km². Therefore, it is essential to reduce the space required for the SNF repositories. The EBS concept was developed to dispose of SNFs (Fig. 1) and shows components, such as bentonite buffer material and a disposal canister. Consequently, buffer material is located between rock-mass and a canister that protects a canister and minimizes the release of radionuclide [3].

The design factor with the most significant influence on the site area of the SNF repository is the peak temperature of the bentonite buffer material. Currently, in most countries, the peak temperature of bentonite is limited to 100℃ or less [3-6]. When bentonite buffer material is exposed to high temperatures for a long period, the buffer material may lose its performance (e.g., swelling ability) as an engineered barrier due to mineral transformation.

However, assuming that the temperature criteria of these buffer materials can be raised to 100℃ or higher, the disposal density of high-level radioactive waste can be increased and the disposal area can be dramatically reduced [7-10]. For this purpose, studies on the coupled thermal-hydro-mechanical-chemical (THMC) behavior and mineral transformation of bentonite buffer materials have been conducted [7-11]. The soil-water characteristic curve (SWCC), which shows the water suction according to the water content, represents the saturation and unsaturated behavior among the THMC properties of the bentonite buffer material (Fig. 2) [12]. Some studies have conducted SWCC tests of the bentonite buffer materials using different temperatures, but most were
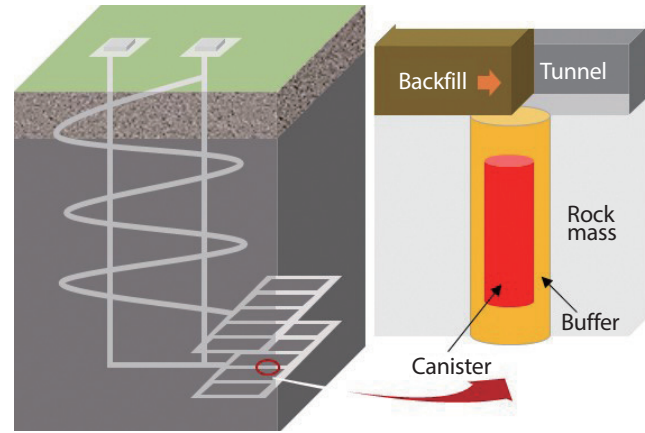

Fig. 1. The concept of EBS.

limited to less than 100℃ [12-14]. Therefore, this study focused on the water suction measurement of bentonite buffer beyond 100℃ under the constant saturation conditions. Yoon et al. (2019) [15] conducted SWCC tests for the Korean bentonites considering the decrease in saturation with increased temperature. The saturation of bentonite buffer will decrease at the initial disposal condition, but after experiencing peak temperature, the saturation will increase or become constant despite the released thermal energy from the canister [16]. Therefore, it is necessary to keep the saturation condition of bentonite buffer constant to investigate the THMC behavior of bentonite buffer.

Kim et al. (2021) [16] predicted the change in the thermo-hydraulic-mechanical (THM) properties of the buffer material according to the spacing between the deposition hole and disposal tunnel by reflecting the characteristics of KRS+, a domestic disposal system. According to their research results, the peak temperature of the buffer material was calculated to be less than 100℃, which occurred after approximately 14 years, and the degree of saturation was confirmed to be approximately 0.2. Based on these results, the disposal tunnel and deposition hole spacing were estimated to be 40 m and 7.5 m, respectively. If the gap between the disposal tunnel and the deposition hole is narrowed to reduce the area required for the disposal site, the peak
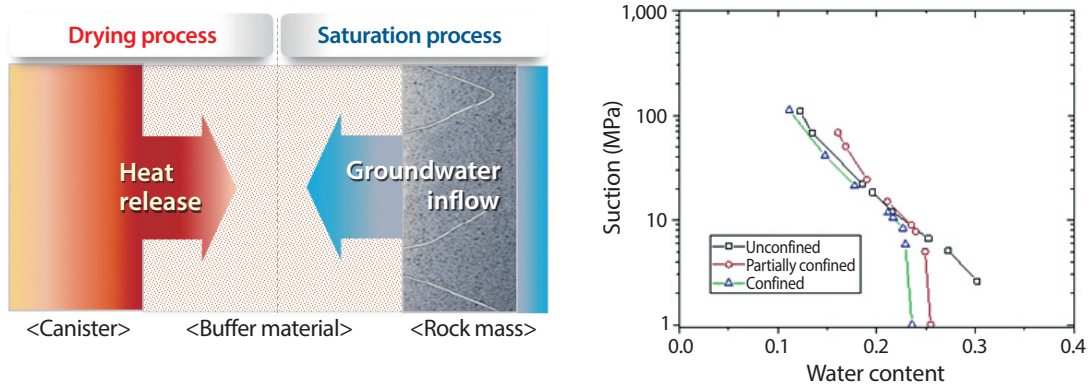
Fig. 2.Water suction variation with respect to saturation process.

temperature of the buffer material will be 100°C or higher, and the degree of saturation at this time will also drop to less than 0.2. Thus, this study measured water suction variation for Korean bentonite between 25°C and 150°C with 0 and 0.22 initial saturations, respectively.

2. Experimental Measurement of Water Suction

2.1 Materials

In this study, the water suction of Gyeongju bentonite, produced by Clariant Korea, was measured. Using the uniform classification method, Gyeongju bentonite powder was classified as highly plastic clay, and the chemical composition was mostly SiO_2 and Al_2O_3. CaO showed a composition ratio approximately 56 times higher than that of NaO [17]. The main minerals in the Gyeongju bentonite include montmorillonite (60.5–63.4%), feldspar (19.4–22.8%), and small amounts of other minerals (15.5–17.2%), including quartz, cristobalite, calcite, and heulandite.

2.2 Water Suction

During the initial stages of disposal, the bentonite buffer exists in an unsaturated state owing to the heat emanat-

ing from the disposal canisters. However, as the disposal canisters cool over time and the groundwater seeps from the surrounding bedrock, the bentonite buffer begins to resaturate (Fig. 2). The process of groundwater penetration into the buffer depends on the water potential (ψ) properties of the buffer. Water potential refers to the energy level of water contained in a medium; energy equilibrium is achieved as moisture moves from a point of high energy to a point of low energy. The overall water potential (ψ_t) of a given system can be calculated using pressure potential (ψ_p), osmotic potential (ψ_o), matric potential (ψ_m), and gravimetric potential (ψ_g) [18].

$$\psi_t = \psi_p + \psi_o + \psi_m + \psi_g \qquad (1)$$

The pressure potential refers to the potential derived from the external pressures acting on the water. Under unconstrained conditions (where no external forces act on the swelling clay), the pressure potential is zero. The osmotic potential refers to the potential of ions or solute molecules in the medium. The gravimetric potential is the energy of water that stems from gravity. The matrix potential is the potential resulting from the energy difference between free water and the water adsorbed on the buffer particles or constrained in the pores of the buffer particles due to capillary forces. In the case of unsaturated compressed clay, the matrix potential value is always negative because the energy state of the

moisture in the clay is lower than that of free water. However, as the buffer became increasingly saturated, the matrix potential value approached zero. In the case of compressed bentonite, the matrix potential is substantially higher than the other potential types; thus, the matrix potential is often used to represent the overall water potential [18]. The water suction (s) was equal to the negative water potential.

$$s = -\psi_t \qquad (2)$$

Water potential is generally measured using thermocouple psychrometry or capacitive thin-film polymer humidity sensors. For bentonite buffers, the latter method is generally preferred because it enables the measurement of water potential without disturbing the system under high-heat and high-pressure conditions [19].

2.3 Experimental Apparatus

The general method of measuring water suction in soils is the axis translation technique (where gradually increasing air pressure is applied to a sample up to 1.5 MPa). However, the axis translation technique is not suitable for measurements involving compressed bentonite samples with high water-suction properties, as in the case of bentonite buffers in high-level waste disposal repositories. Instead, the vapor equilibrium (VE) method is generally used [18-20]. Therefore, we also employed the VE method to measure the water suction of bentonite according to its volumetric water content. The VE method assumes that the water potentials of the pore water and water vapor in the pores of a sample are in equilibrium. The Kelvin equation (3) was used to calculate the water suction (s) from the measured relative humidity (RH) values of the compressed bentonite.

$$s = -\frac{RT}{v_{\omega 0}\omega_v} \ln\left(\frac{RH}{100}\right) \qquad (3)$$

where R is the gas constant (8.3143 J·(mol^{-1}·K^{-1})), T is the absolute temperature (K), $v_{\omega 0}$ is the specific volume
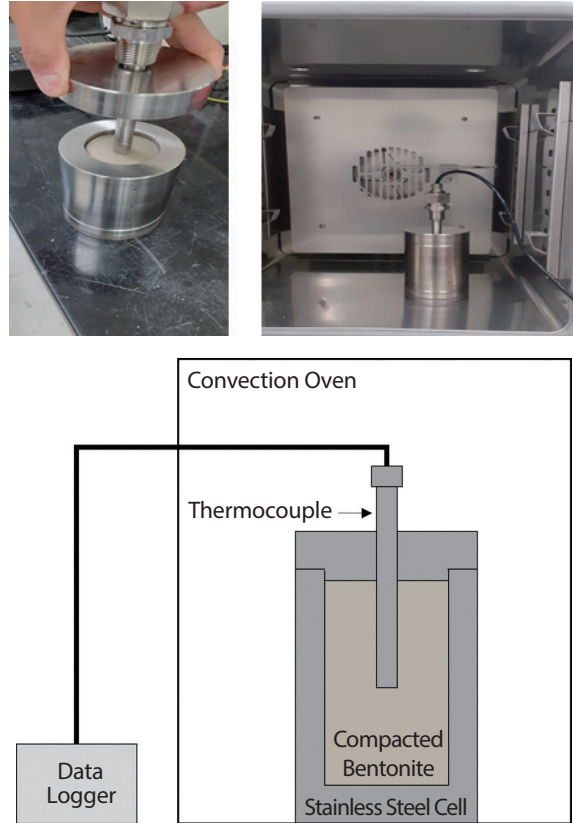

Fig. 3. Experimental apparatus for the measurement of water suction.

of water (0.001 m³·kg^{-1}), and ω_v is the molar mass of water (18.016 kg·kmol^{-1}) (Nguyen-Tuan, 2014). A relative humidity sensor was installed at the center of the compressed bentonite sample via carving (Fig. 3). The compressed bentonite sample was molded into a cylindrical shape with a diameter of 50 mm and a height of 70 mm, and the relative humidity sensor was 12 mm in diameter and 30 mm in height. A convection oven was used to maintain the temperature at high-temperature levels of 150°C and higher. Additionally, a stainless-steel cell was used to ensure airtightness.

3. Results and Discussion

According to a study by Kim et al. (2021) [16], the degree of saturation of a buffer is approximately 0.2 when
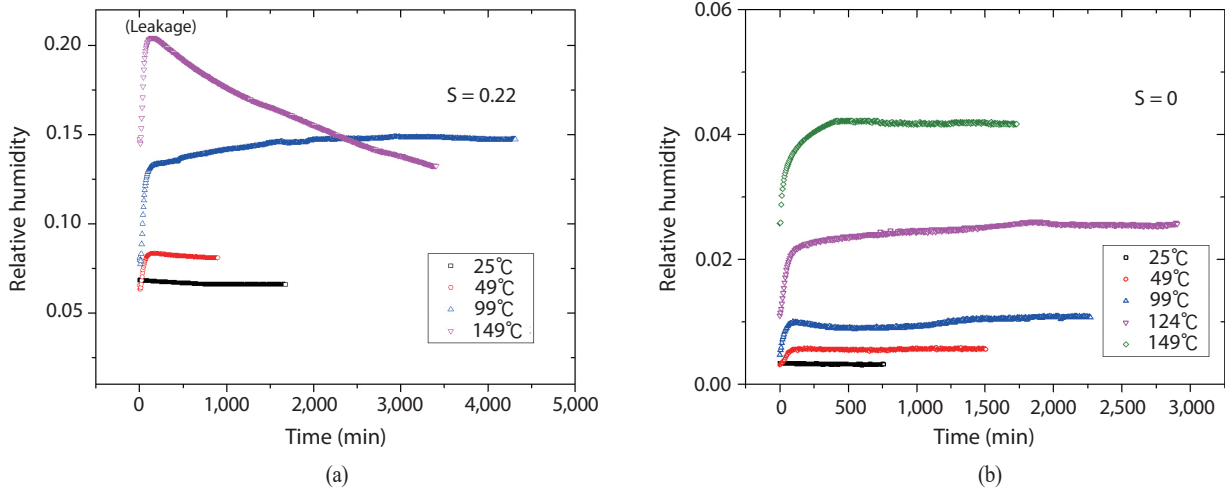
Fig. 4. Relative humidity (RH) variation.

approaching the maximum temperature near 100°C. In our experiments, we recreated conditions similar to the assumed conditions during the initial 10–20 years of an operation with initial saturation (S) values of 0 and 0.22 and an initial dry density of approximately 1.61 (±1%) g·cm⁻³. Under these conditions, we measured the water suction of the compressed bentonite sample over a temperature range of room temperature to 150°C. Fig. 4 shows the change in relative humidity over time for bentonite buffer samples with initial saturation values of 0 and 0.22. The experiment was conducted until a specific temperature was reached at which the relative humidity value plateaued to an almost constant value. The relative humidity value was entered into Equation (3) to derive the water suction of the buffer. According to the results, the relative humidity value of the buffer increased as the temperature increased. This is due to an increase in water vapor as the moisture in bentonite vaporizes into water vapor. Additionally, this phenomenon is a result of water flowing from the micropores between elementary layers between the bentonite aggregates as the temperature increases, thus leading to an increase in the overall saturation [18, 20]. In contrast, Yoon et al. (2019) [15] showed a relative decrease in humidity with respect to temperature because it was conducted considering decreased saturation of bentonite buffer. The buffer sample

with an initial saturation of 0.22, the relative humidity increased until the sample reached a temperature of 149°C, then abruptly declined. This phenomenon was caused by a leak in the cell containing the buffer, resulting in the leakage of water vapor. At a temperature of 149°C (far higher than the boiling point of water), the water in the pores of the bentonite sample all rapidly vaporized, thus increasing the water vapor pressure in the air. Moreover, the air itself expands owing to the high temperature, which increases the air pressure [21]. As a result, the significant increase in the gas (air + water vapor) pressure renders it difficult to seal the sample completely. In the case of the completely dried sample with an initial saturation value of 0, the absence of moisture results in little water vapor being produced, even at 149°C. Therefore, the overall gas (air + water vapor) pressure did not substantially increase, and we believe this is why the dried sample did not result in any leakage.

Fig. 5 shows the change in water suction (obtained by converting the relative humidity values from Fig. 4) according to the temperature of the bentonite buffer. For the sample with an initial saturation of 0.22, the water suction of the buffer exhibited a tendency to gradually decrease as the temperature increased. The water suction value at 149°C corresponds to the point at which the maximum relative humidity value is achieved. Due to increasing
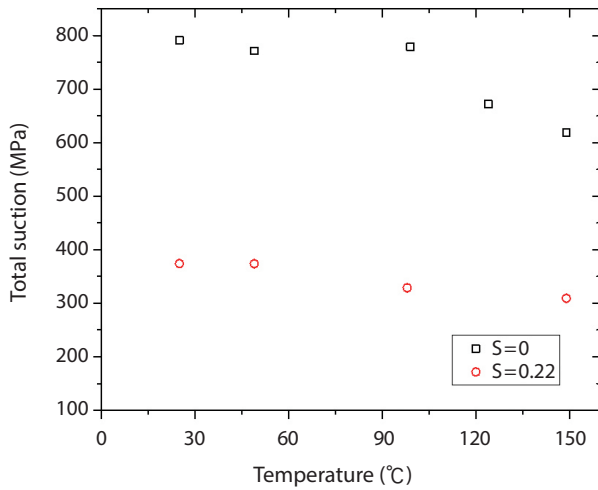
Fig. 5. Water suction variation with respect to temperature.

Fig. 6. Pore waters in a porous material [24].

temperature, water from the intra-aggregate pore flows into the inter-aggregate pore. Thus, the density of intra-aggregate pore water is higher than 1 g·cm⁻³ [22], and the degree of saturation increases with increasing temperature resulting in the decrease of the water suction.

In the case of the sample with zero initial saturation, the suction value did not change up to 100℃, but at 100–150℃, the suction sharply decreased with increasing temperature. Bentonite consists of a 2:1 layered structure, where a layer of gibbsite is sandwiched between two silica sheets [23], and water in the bentonite particles exists in three forms: the bound water adsorbed on the surface of basic layers, the trapped water in the micro dead-end pore, and the capillary pore water (Fig. 6). The capillary pore water can be drained and regarded as free water. The bound water and the trapped water can be regarded as the residual water [24]. The general process of drying soil samples at 110℃ prior to water content measurements is a process that removes capillary pore water, not residual water [23]. In other words, capillary pore water was removed from soil samples dried at 110–150℃, and thus the water suction decreased as the residual water detached and moved to the capillary pores. Conversely, samples dried at temperatures below 100℃ do not exhibit the movement of adsorbed water, resulting in
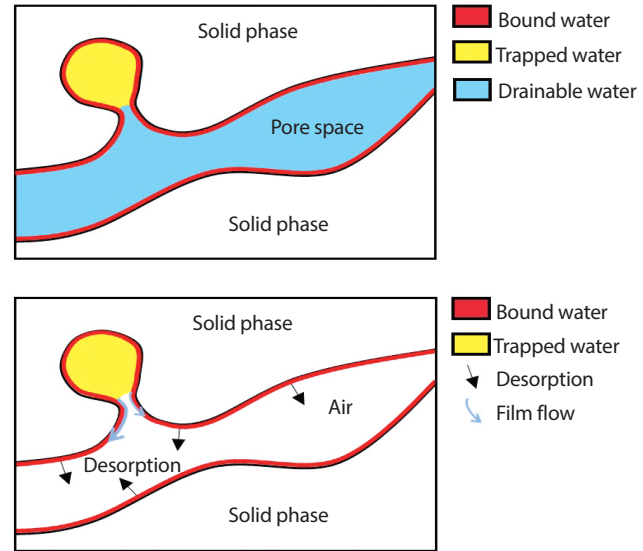
temperature increases having little effect on suction. However, as the temperature increased to 125℃ or 150℃, the remaining moisture began to move to the capillary pores; thus, the suction began to decrease.

4. Conclusion

It is necessary to study the complex behavior of bentonite buffer above 100℃ to estimate temperature change for reducing disposal area. Therefore, this study investigated the change in matric suction of bentonite buffer material under high-temperature conditions up to 150℃. A system to measure the matric suction of the bentonite buffer between 25℃ and 150℃ under the constant saturation condition was constructed. Compacted bentonite blocks were prepared with an initial saturation degree of 0.22 and a dry density of 1.61 (±1%) g·cm⁻³ based on the current literature, in which the degree of saturation was 0 to 0.22 at less than 100℃. The matric suction at constant temperature was derived using the VE method.

As a result, when the temperature increased, the relative humidity of the bentonite buffer material increased, and the

matric suction tended to decrease. This is because the moisture in the bentonite block evaporates and the amount of water vapor in the air increases as the temperature increases. In addition, the total degree of saturation increased as the water between bentonite layers moved to the macropores. In the case of dry bentonite block, there was no change in matric suction from room temperature to 100℃, but the matric suction decreased by approximately 20% between 100 to 150℃. This is because the adsorbed water of bentonite moves to the capillary pores under high-temperatures above 100℃. Moreover, in the case of the bentonite block with an initial saturation degree of 0.22, the matric suction between 100 to 150℃ decreased by 5 to 6%.

Overall, when the initial degree of saturation of the bentonite buffer material was 0 to 0.22, the matric suction from 100 to 150℃ tends to decrease by approximately 5 to 20%. Therefore, it is necessary to clearly identify the SWCC of bentonite buffer materials under high-temperature conditions and apply it to the safety evaluation of repositories.

Acknowledgements

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