Investigation of Soil-Water Characteristic Curves for Compacted Bentonite Considering Dry Density

Seok Yoon[1] Seeun Chang[2] Dowon Park[3,*]

[1] Division of Radioactive Waste Disposal Research, Korea Atomic Energy Research Institute (KAERI), Daejeon, Republic of Korea

[2] Radwaste Disposal Team, Korea Atomic Energy Research Institute (KAERI), Daejeon, Republic of Korea

[3] Department of Civil Engineering, University of Seoul, Seoul, Republic of Korea

[*] Correspondence: dowon@uos.ac.kr

Abstract: Deep geological repositories are often considered for the disposal of high-level radioactive waste (HLW). HLW should be disposed of safely and permanently using two barrier systems: engineered barrier systems (EBS) and natural barrier systems (NBS). An EBS is an artificially suggested concept composed of various components. The buffer is one of the most important components since it plays crucial roles in terms of the safety of the entire disposal system, and compacted bentonite is considered a highly adequate buffer candidate material. As groundwater flows into the rock-mass and buffer, the degree of saturation of compacted bentonite increases. A soil-water characteristic curve (SWCC) represents saturation behavior, and thus it is essential to investigate SWCCs of compacted bentonite. For this reason, this paper conducted SWCC tests of Korean compacted bentonites according to dry density variations, as the dry density of compacted bentonite also varies in actual repository environments. The van-Genuchten fitting parameters (α, n) were obtained; α decreased in exponential form and n increased linearly according to dry density. Furthermore, using the obtained SWCC datasets, a simple empirical model was suggested to estimate the van-Genuchten fitting parameters. The mean relative error between the test results and the empirical model was approximately 8.6%.

Keywords: High-level radioactive waste disposal; Compacted bentonite; Soil-water characteristic curve; van-Genuchten fitting parameters

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Key Points:

- Laboratory SWCC tests were conducted for Korean compacted bentonites according to dry density variations.
- The van-Genuchten fitting parameters (a, n) were obtained considering dry density.
- Simple empirical equations to estimate the values of a and n were suggested according to dry density and were compared with the test results.

1. Introduction

The operation of nuclear power plants inevitably leads to the generation of spent fuel.

Since spent fuel can be classified as high-level radioactive waste (HLW), it must be safely disposed in deep geological repositories based on a multi-barrier system that includes engineered barrier systems (EBS) and natural barrier systems (NBS) (Chen et al., 2017; Zheng et al., 2015). In Korea, as of February 2020, 17,000 tons of HLW has been produced (Yoon et al., 2021). Engineered barriers are features of disposal systems made by humans (IAEA, 2002), and a compacted bentonite buffer is one of the main components of an EBS. Since it is located between the disposal canister and rock-mass, it protects the former from the percolation of groundwater and external physical impact, as shown in Figure 1 (Kim et al., 2019; Sellin et al., 2013). Compacted bentonite buffers are initially in a dry state due to decay heat released from the disposal canister, but the buffer becomes almost fully saturated due to groundwater penetration after several centuries, as shown in Figure 2 (Cho, 2017; Juvankoski, 2009; Yoon et al., 2021). In other words, compacted bentonite buffers experience complex thermal-hydraulic-mechanical-chemical (THMC) variations according to the disposal environment, such as groundwater inflow and the release of decay heat. For this reason, it is essential to investigate the soil-water characteristic curve (SWCC) of compacted bentonites since a SWCC reflects the variation of water content with respect to water potential (Fredlund & Xing, 1994; Nikhil & Lee, 2016; Lee et al. 2014; Park, 2019; van-Genuchten, 1980; Zhai & Rahardjo, 2012). Furthermore, compacted bentonites are subject to volume changes and swelling tendencies in actual repository environments (Villar, 2004). Contraction may take place due to porosity changes and variations in the pore structure (micropores and macropores) of compacted bentonites due to thermal loading (Daniel et al., 2017), or the compacted bentonites may become expansive due to the inflow of groundwater (Cho, 2017). Volume and porosity changes of compacted bentonite may lead to variations in dry density, and thus it is vital to investigate the SWCCs of compacted bentonite buffers according to dry density.

Figure 1. Compacted bentonite buffer in an EBS (Yoon et al. 2021).

Figure 2. Drying and saturation processes of buffer materials.

There have been significant research efforts to evaluate SWCCs for compacted bentonite considering various factors, including temperature variation, dry density, and solution properties (Alberdi et al., 2000; Ballarini et al., 2017; Lee et al., 2014; He et al., 2019; Villar, 2004; Villar et al., 2010). Despite the abundant experimental attempts to derive SWCCs for compacted bentonites according to dry density variations, few studies have attempted to analyze the variations in SWCC fitting parameters based on geotechnical theories. Furthermore, there have been a lack of studies that have suggested SWCC prediction models for compacted bentonite buffers considering dry density. SWCC fitting parameters of compacted bentonite buffers are crucial input parameters for numerical analyses involved in safety evaluations of disposal systems. If SWCCs of compacted bentonite are affected by dry density, a SWCC prediction model with dry density as an independent variable may be necessary to evaluate the safety of disposal systems.

Therefore, this paper conducted SWCC tests for Korean compacted bentonites with various dry densities based on the chilled-mirror dew point temperature condensation (CMDPTC) method. This paper also analyzed how dry density variations affect the SWCC results based on geotechnical theories and principles. Additionally, simple regression models were suggested to estimate SWCC fitting parameters (van-Genuchten, 1980) for compacted bentonite according to dry density.

2. Experimental setup for SWCC tests

2.1 Bentonite

Bentonite is a clay-rich material composed of montmorillonite, quartz, feldspar, cristobalite, and a small amount of organic matter. Montmorillonite, the main component of bentonite, lacks positive charges within its layer structure (JNC, 1999), enabling exchangeable cations such as Na^{+}, Mg^{2+}, and Ca^{2+} to be absorbed between layers to compensate for the lack of positive charges. In South Korea, a Ca-type bentonite referred to as Gyeongju bentonite is produced in the southeastern Gyeongju region and the exchangeable cation involved is Ca^{2+} (Cho, 2017; Yoon et al., 2021). The main minerals in Gyeongju bentonite are montmorillonite (62%), feldspar (21%), and small amounts of other minerals (~17%, including quartz (5%), cristobalite (4%), calcite (5%), and heulandite (3%)), as shown in Table 1. In Korea, physical, hydraulic, thermal, and chemical characterization studies were carried out to investigate the potential of Gyeongju bentonite as a candidate buffer material (Lee et al., 2011). The typical engineering properties of Gyeongju bentonite are presented in Table 2.

Table 1. Quantitative mineral constituents of Gyeongju bentonite (Yoon et al., 2021).

Table 2. Typical properties of Gyeongiu bentonite.

2.2 SWCC test

A change in the water content of a bentonite buffer is directly related to its water potential. The water potential (ψ) of compacted bentonite is usually referred to as its matric potential: the energy of water generated by the adhesion of polarized molecules to the unsaturated medium surface and capillary forces in voids (Lee et al., 2017). SWCC tests were conducted for Korean compacted bentonites mainly using the CMDPTC method. The bentonite powders were tightly compacted in cylindrical steel cells (37 mm in diameter and 4~5 mm in height). A WP4C apparatus (Decagon Devices Inc., USA) was used, which is based on the CMDPTC method (Nguyen-Tuan, 2014; Yoon et al., 2020). Unlike general soil, the water potential of compacted bentonite is usually derived by measuring the relative humidity using Kelvin's law shown in Eq. (1) (Lee et al., 2017; Yoon et al., 2021).

$$\psi = \frac{RT}{M}\ln(\frac{p}{p_o}) \tag{1}$$

where P is the vapor pressure of air (kPa), P_o is the saturation vapor pressure (kPa), R denotes the gas constant (8.31 J/(mol K)), T is the temperature of the compacted bentonite (K), and M is the molecular mass of water. Here, P is measured using the WP4C apparatus and P_o is calculated using the compacted bentonite temperature. The WP4C apparatus, which is composed of a dew point sensor, a fan, and an infrared thermometer inside the sensor block (Figure 3), calculates the water potential by equilibrating the liquid phase water of the specimen with the vapor phase water of the chamber. The dew point temperature of the air is measured, and the temperature of the compacted bentonite is measured by the infrared thermometer (Yoon et al., 2021).

Figure 3. Schematic diagram of the WP4C apparatus (Yoon et al., 2020).

This study also used the VE (vapor equilibrium)-cell/sensor method (Figure 4) to derive the SWCC for comparison with the results derived using the CMDPTC method. The VE-cell/ sensor method also measures the relative humidity of a compacted bentonite sample to obtain water potential, and it is assumed that equilibrium is achieved between the liquid phase water of the sample and the vapor phase water in the headspace. Additionally, water suction can be obtained using Eq. (1). A specimen 50 mm in diameter and 70 mm in height was fabricated for the test. The relative humidity sensor should be inserted into a hole in the specimen. However, it is highly difficult to form a compacted bentonite specimen with a hole, and an extended period of time is required to achieve water content equilibrium using the VE-cell/sensor method (Lee et al., 2017).

Figure 4. Schematic diagram of the VE-cell/sensor method.

3. van-Genuchten SWCC theory

In unsaturated soils, the volumetric water content is controlled by matric suction. Whereas laboratory measurements of these parameters are discrete points, engineering applications require the results presented as continuous functions. The mathematical relationship between water content and soil suction is known as the soil-water characteristic curve (SWCC), typically plotted on a semi-log scale. Many mathematical expressions have been proposed in the literature, among which the van Genuchten equation (1980) has become the preferred equation for unsaturated soil mechanics. The advantage of this equation above others is its flexibility: it covers a wide range of suction and captures smooth transitions. Detailed reviews regarding SWCC models have been summarized by Leong and Rahardjo (1997) and Sillers et al. (2001).

The fundamental expression of the van Genuchten equation (1980) as functions of soil volumetric water content θ and matric suction ψ reads as follows:

$$\Theta = \frac{\theta - \theta_r}{\theta_s - \theta_r} = \left[\frac{1}{1 + (\alpha\psi)^n} \right]^m \qquad (2)$$

where Θ is the normalized volumetric water content (dimensionless) that is equal to the degree of saturation, θ_s is the saturated water content ($cm^3 \cdot cm^{-3}$), θ_r is the residual water content ($cm^3 \cdot cm^{-3}$), α is a parameter corresponding approximately to the inverse of the air-entry value, and m and n are empirical shape-defining parameters (with m often being $1 - 1/n$). The constants n and m are related to the pore size distribution of the soil and the asymmetry of the model, respectively.

Figure 5 presents the influence of the van Genuchten parameters (Eq. (2)) on the shape of SWCCs. The van Genuchten equation uses three curve fitting parameters obtained generally by linear regression: α, n, and m. Among these parameters, one among α and n is varied whereas the other is kept constant, as shown in Figure 5(a). Constant m is assumed to be $1 - 1/n$. Parameter α induces a shift of the curve, but the shape of the curve is not affected. Curves with a smaller α move toward the higher matric suction range in the plot, as α is inversely related to the air-entry value. When it comes to varying n with α as a constant ($\alpha = 0.1$, $m = 1 - 1/n$), as shown in Figure 5(b), the larger the n, the steeper the curve in the capillary regime. The slope of the curve in the capillary regime is primarily dependent on the pore size distribution of the soil: soils with a more uniform pore size distribution exhibit a steep soil-water characteristic curve as most pores are drained in the narrow range of matric suction, which is related to larger values of n (Hillel 1998). Note that the air-entry value is constant owing to the fixed value of α.

Figure 5. Effects of parameters on the shape of van Genuchten soil-water characteristic curves: (a) varying α with $n = 1.5$, $m = 1 - 1/n$, and (b) varying n with $\alpha = 0.1$, $m = 1 - 1/n$.

4. Test results and discussion

4.1. Comparison between CMDPTC and VE methods

As this paper mainly used the CMDPTC method to derive SWCCs for compacted bentonites, it is important to verify the results of the CMDPTC method through comparisons with results obtained using the classical VE-cell/sensor method. The VE-cell/sensor method has been widely adapted to measure SWCCs for compacted bentonites (Blatz et al., 2008; Jacinto et al., 2009; Lee et al., 2017). Figure 6 shows the SWCC results of the two aforementioned methods for Korean compacted bentonites with a dry density of 1.6 g/cm^3 (±0.5%) under the confined condition. This figure shows highly similar curves between the two methods in medium suction ranges. However, it can be said that there are some difficulties in forming homogenous samples under high water content conditions, especially for the VE-cell/sensor method which requires significantly higher quantities to form compacted samples.

Figure 6. SWCC results between the two methods.

4.2. SWCC results considering dry density

Figure 7 presents the SWCCs of soils with different dry unit weights ranging from 1.40 g/cm^3 to 1.73 g/cm^3. The saturated volumetric water contents (θ_s) of the tested samples are plotted as volumetric water contents at ψ = 1 MPa. Overall, volumetric water content decreases with increasing matric suction. However, the volumetric water contents appear to be independent to dry unit weight when the samples are unsaturated. Romero et al. (1999) and Dieudonne et al. (2017) reported similar results where the influence of dry unit weight on SWCCs appeared to be negligible at low water content levels. For each bentonite sample, the best-fitted van Genuchten parameters, α and n ($m = 1 - 1/n$), were computed as shown in Table 3.

Figure 7. Soil-water characteristic behaviors of specimens with different soil dry unit weights.

Table 3. Saturated water contents and estimated van Genuchten parameters of Gyeongju compacted bentonites

Figure 8 shows the best fitting curves with samples of different dry unit weights. Using the curve fitting toolbox in the MATLAB environment, the van Genuchten parameters α and n can be presented as functions of the dry unit weight of soil:

$$\alpha = 179.9e^{-4.882\gamma_d}$$
$$n = 0.3787\gamma_d + 0.8196 \quad (3)$$

Parameter α and soil dry unit weight γ_d exhibit a strong relationship where α exponentially decreases as γ_d increases, as shown in Figure 8(a). This trend is not surprising, considering that the air-entry value increases linearly with the dry unit weight of the soil (Vanapalli et al., 1999). Soil samples with smaller dry densities have larger pores, indicating that they desaturate at lower matric suctions (air-entry value) according to the Young-Laplace equation (Benson et al., 2014). Thereby, soils with larger pores have larger α values (a smaller air-entry value corresponds to a larger α value, as illustrated in Figure 5(a)). Despite the significant variation in the experimental measurements in Figure 8(b), the linear increase in n with respect to dry unit weight may also be explained by pore size. Romero et al. (1999) presented that the dry density of soil affects macro-pores: as the dry density increases (denser), the pore size distribution becomes narrower, with small-to-medium-sized pores becoming dominant at the expense of larger pores. From Figure 5(b), this results in larger values of n in specimens with greater dry unit weight. This increased descent gradient of soils with low porosity was also reported by Romero et al. (1999) and Dieudonne et al. (2017).

Figure 8. Influence of the dry unit weight of soils on the van Genuchten curve fitting parameters: (a) parameter α, and (b) parameter n.

In Figure 9, curve fits for the van Genuchten equations using the proposed function (Eq. (3)) are compared with the experimental measurements results from the wetting test. The fitted curves were not drawn according to the best estimate of α and n for each sample (scatters in Figure 8) but were obtained from the suggested relationship in Eq. (3) or the corresponding values on solid lines in Figure 8 at given dry unit weights. For five different dry unit weights, Figures 9(a) and (b) were plotted with water content (θ) and normalized volumetric water content (Θ), respectively. The results displayed on the $\theta - \psi$ plot (Figure 9(a)) show that samples with large porosity held greater amounts of water at low matric suctions. When plotted in terms of normalized volumetric water content (Figure 9(b)), the influence of the dry unit weight on the soil-water characteristic behaviors is clear, as water content is normalized by different pore volumes at different dry unit weights. The predicted SWCCs follow the experimental measurements with reasonable accuracy, considering that the fitting parameters were estimated using only the dry unit weight of soil. The numerical results are summarized in Table 4. The average difference between the SWCC test results and those obtained by the regression analysis is 8.6%.

Figure 9. Comparison of experimental measurements and soil-water characteristic curve fits in terms of: (a) water content θ, and (b) normalized volumetric water content Θ.

Table 4. Comparisons of the van Genuchten parameters

The water retention behaviors according to different dry unit weights of bentonite samples in Figure 9(b) are consistent with the findings in Figures 5 and 8. It can be expected that soils with lower dry unit weights have larger pores (larger void ratio). Furthermore, the soil-water characteristic behaviors of such soils exhibit smaller air-entry values (larger α), as air is able to easily enter the largest pores in the soil at low suction ranges. In addition, the slopes of the soil-water characteristic curves in the capillary regime increase as dry unit weight increases. When soil is heavily compacted, the total volume of void decreases; large pores (macropores) are especially reduced in size without difficulty. From a phenomenological point of view, a decrease in the population of macropores leads to narrow pore size distributions comprising mostly of micro-to-medium-sized pores. Thereby, most of the saturation takes place across a narrow range of matric suction. This may be the reason for the steeper slopes and larger values of n in the case of low-porosity samples. However, further investigation is required to determine the correlated connection by taking into account the complex hydro-mechanical processes that affect soil-water characteristic curves.

5. Conclusion

As a component of engineered barrier systems in deep geological repositories, a buffer protects the disposal canister from environmental impacts. A typical buffer material involves compacted bentonite with a low flow rate, preventing the leakage of high-level radioactive waste and groundwater inflow. Although numerous studies have extensively investigated SWCCs of compacted bentonite for varying dry unit weights with regard to its performance, the underlying mechanisms of the dry-unit weight dependency of SWCCs are not fully understood.

In this paper, SWCC tests were carried out for compacted bentonites with different dry unit weights to determine the influence of the dry unit weight on the SWCC, particularly the van Genuchten (1980) model parameters. Two experimental techniques, CMDPTC and VE, were utilized to characterize the soil-water characteristic behaviors of compacted bentonite samples; the former appeared to be the more effective method with regard to sample preparation and the testing procedure, whereas the latter proved ineffective. As both techniques presented similar outcomes, the CMDPTC method was adopted as the primary experimental tool in this study.

The SWCC results showed that the van Genuchten parameters depend on the dry unit weight of compacted bentonites: an increase in dry unit weight results in an exponential decrease in α and a linear increase in n. Such dependencies are interpreted to be related to the differences in pore size distributions according to the dry densities of the bentonite. The van Genuchten parameters for a given dry unit weight of bentonite were successfully reproduced using the proposed empirical estimation. Given that the dry unit weight of a bentonite buffer continuously varies due to groundwater inflow and the release of decay heat, the proposed empirical estimations of the van Genuchten parameters derived from dry unit weight could be used as a quick reference to assess the safety of a disposal system without the need for advanced experiments.

Data Availability Statement

All datasets supporting this work are publicly available online (https://doi.org/10.7910/DVN/4K544E).

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Figure Captions

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Figure 2. Drying and saturation process for the buffer materials.

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Figure 4. Schematic diagram of the VE-cell/sensor method.

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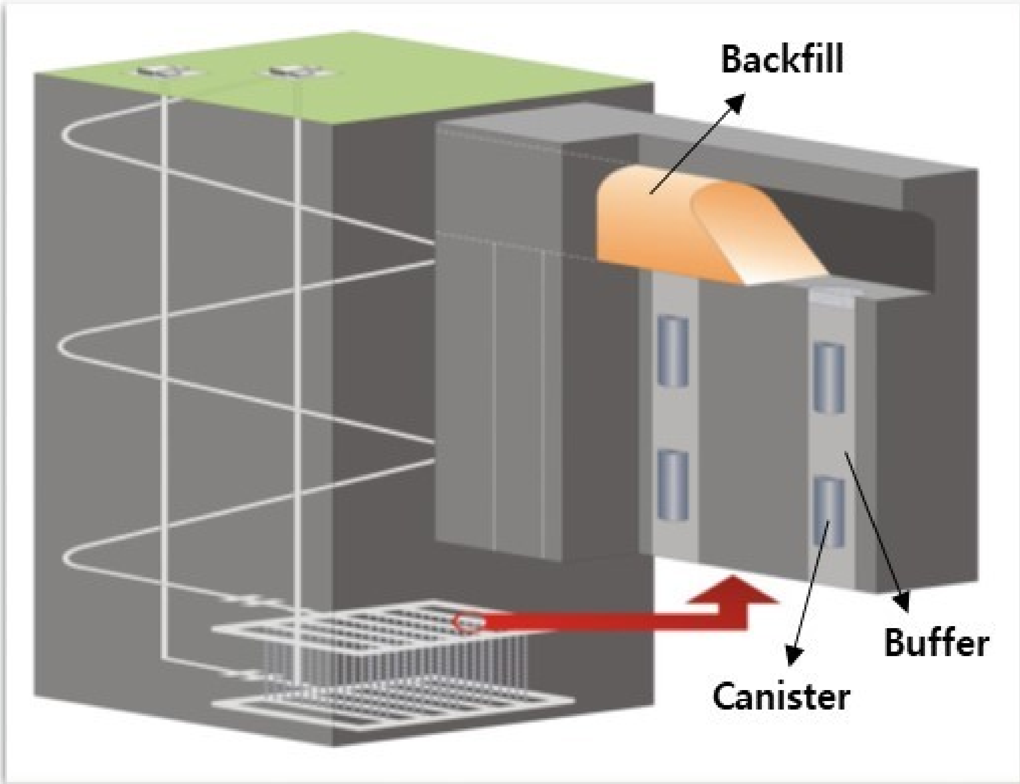

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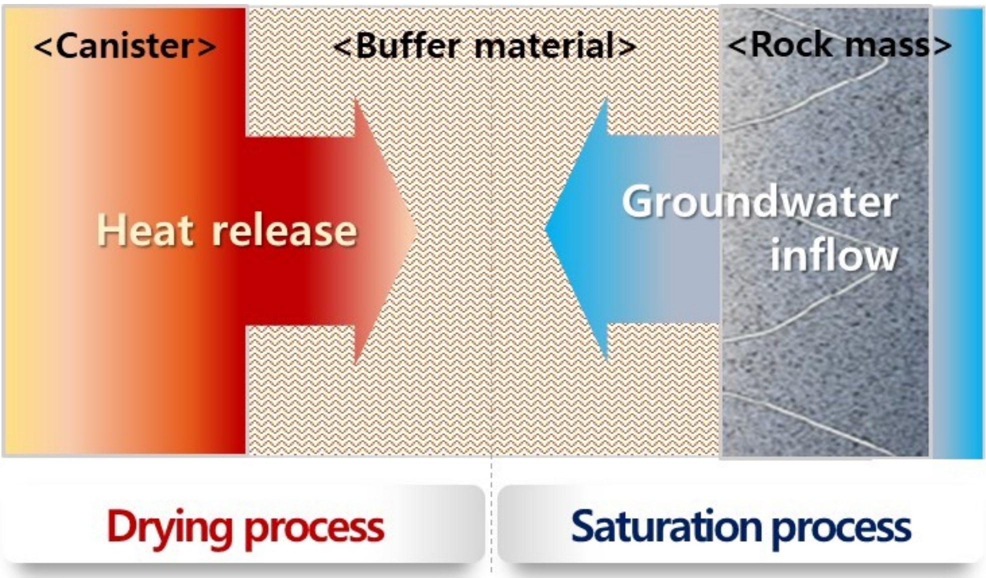

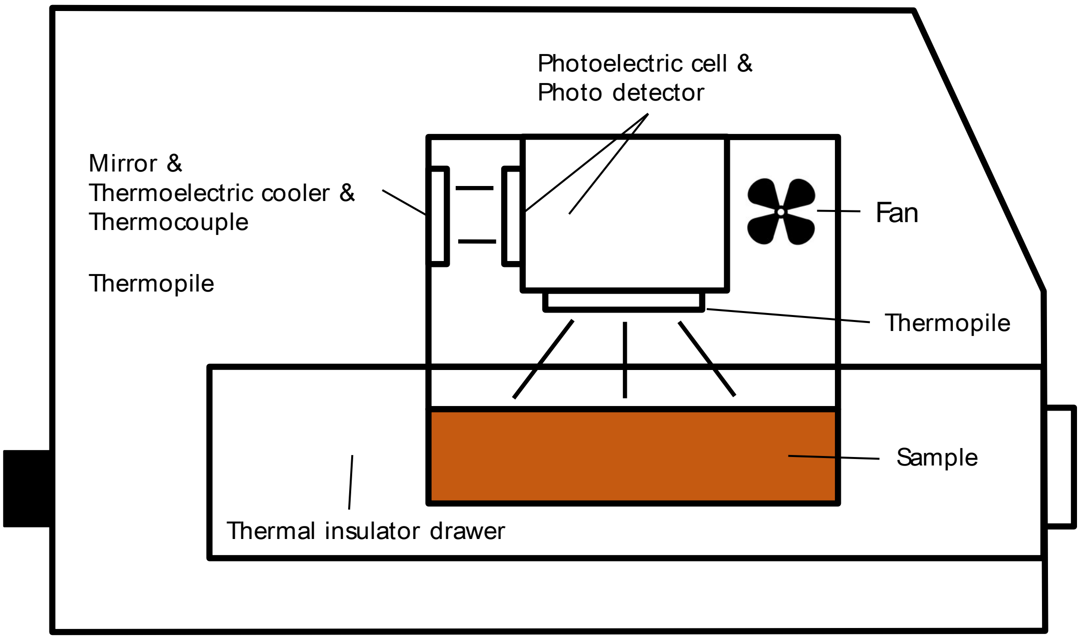

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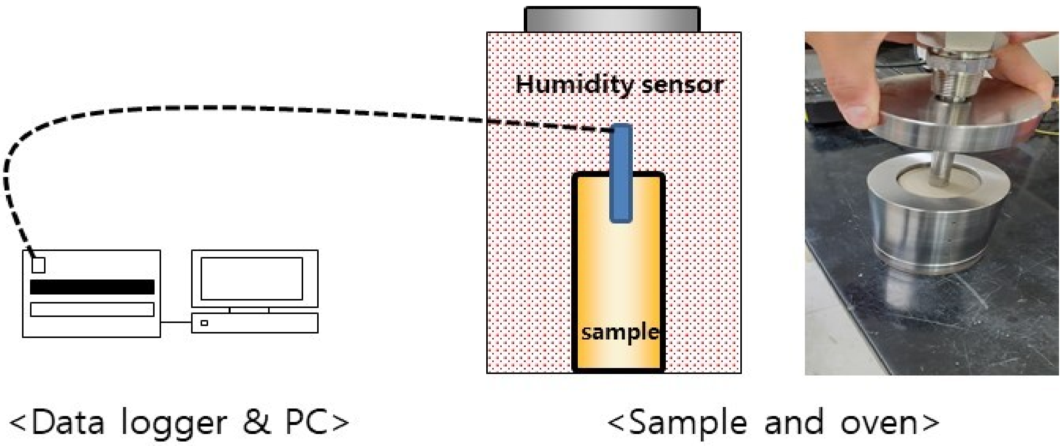

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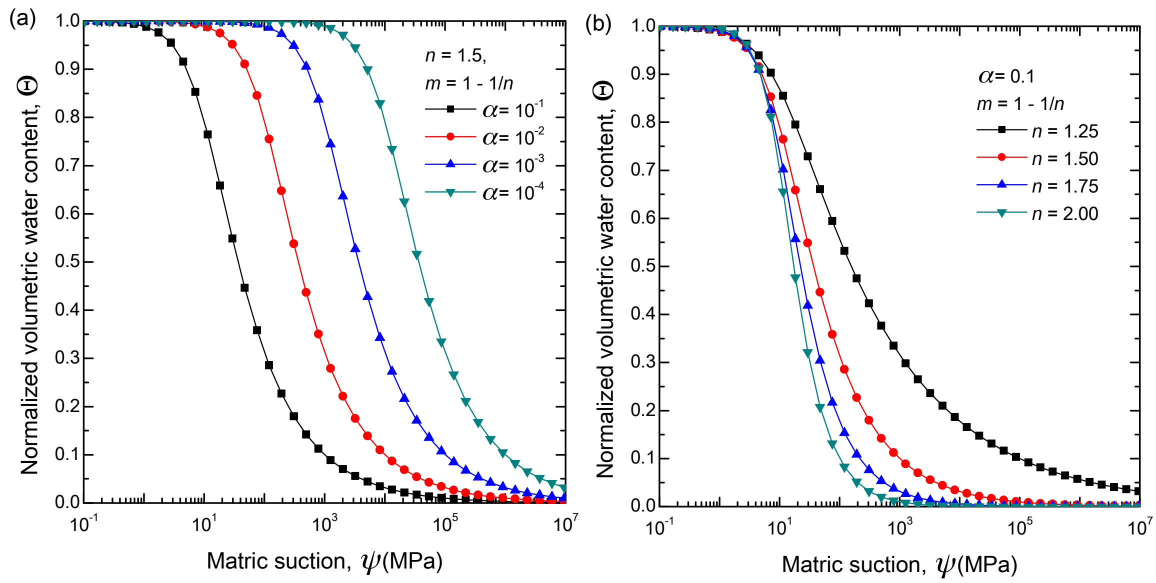

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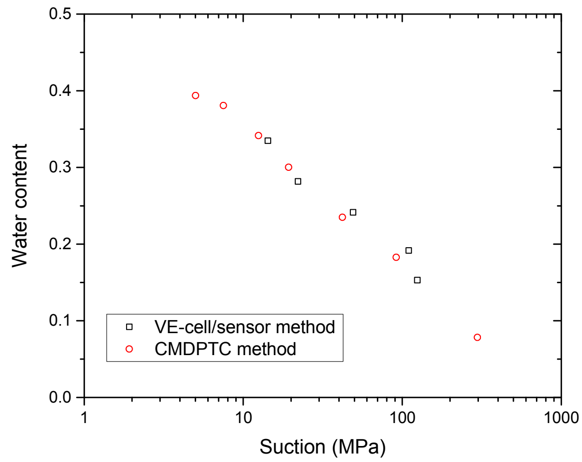

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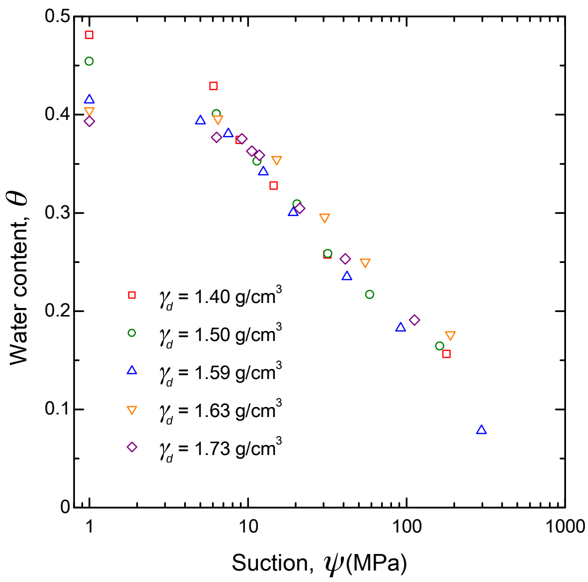

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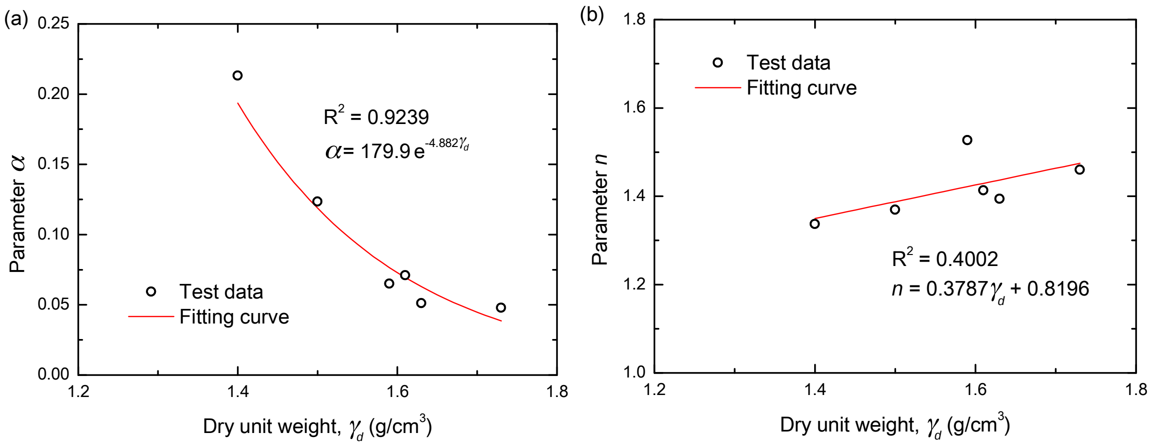

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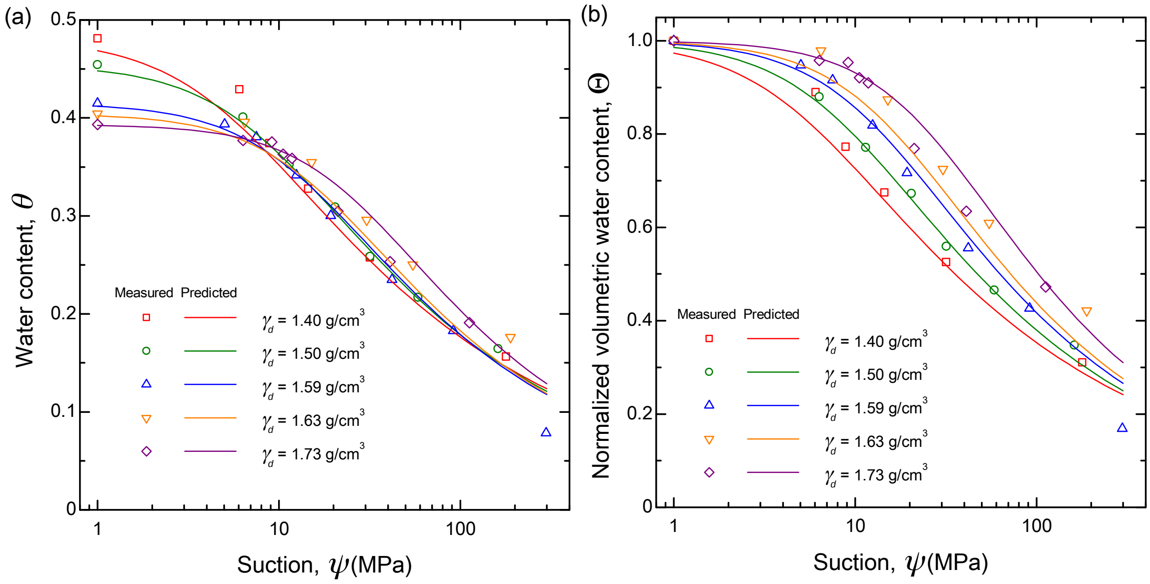

Figure 9. Comparison of experimental measurements and soil-water characteristic curve fits in terms of: (a) water content θ, and (b) normalized volumetric water content Θ.

Table Captions

Table 1. Quantitative mineral constituents of Gyeongju bentonite (Yoon et al., 2021).

Table 2. Typical properties of Gyeongju bentonite

Table 3. Saturated water contents and estimated van Genuchten parameters of Gyeongju compacted bentonites

Table 4. Comparisons of the van Genuchten parameters

Table 1. Quantitative mineral constituents of Gyeongju bentonite (Yoon et al., 2021).

Bentonite Type	KJ-II			
Sample No.	1	2	3	Avg.
Montmorillonite	63.4	61.7	60.5	61.9
Albite	19.4	22.8	20.4	20.9
Quartz	5.8	4.9	5.3	5.3
Cristobalite	4.0	4.5	3.7	4.1
Calcite	4.3	3.3	6.8	4.8
Heulandite	3.0	2.7	3.3	3.0

Table 2. Typical properties of Gyeongju bentonite

Property	Gyeongju bentonite
Water content (%)	11~12%
Swelling Index (mL/2g)	8
Specific gravity	2.71
Specific surface area (m^2/g)	61.5
Cation Exchange Capacity (meq/100g)	64.7
Plastic Index (%)	118.3
Liquid limit (%)	146.7

Table 3. Saturated water contents and estimated van Genuchten parameters of Gyeongju compacted bentonites

Dry unit weights γ_d (g/cm^3)	θ_s	α	n
1.40	0.481	0.2131	1.337
1.50	0.454	0.1235	1.369
1.59	0.415	0.0650	1.527
1.61	0.406	0.0710	1.413
1.63	0.404	0.0511	1.394
1.73	0.393	0.0478	1.460

Table 4. Comparisons of the van Genuchten parameters

Van Genuchten parameters	Dry unit weights (g/cm^3)				
	1.40	1.50	1.59	1.63	1.73

α	Measured	0.2131	0.1235	0.0650	0.0511	0.0478
	Predicted	0.1935	0.1188	0.0765	0.0630	0.0386
n	Measured	1.337	1.369	1.527	1.394	1.460
	Predicted	1.350	1.388	1.422	1.437	1.475