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Thermal Conductivity Evaluation of Compacted Bentonite Buffers Considering Temperature Variations

압축 벤토나이트 완충재의 온도에 따른 열전도도 평가

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An engineered barrier system (EBS) for the geological disposal of high-level radioactive waste (HLW) consists of a disposal canister packed with spent fuel, buffer material, backfill material, and gap-filling material. The buffer material fills the space between the canister and the near-field rock, thus serving to restrain the release of radionuclides and protect the canister from groundwater penetration. Furthermore, as significant amounts of heat energy are released from the canister to the surrounding rock, the thermal conductivity of the buffer plays an important role in maintaining the safety of the entire disposal system. Therefore, given the high levels of heat released from disposal canisters, this study measured the thermal conductivities of compacted bentonite buffers from Gyeongju under temperature variations ranging 25 to 80~90℃. There was a 5~20% increase in thermal conductivity as the temperature increased, and the temperature effect increased as the degree of saturation increased.

Keywords: Engineered barrier system, Compacted bentonite buffer, Thermal conductivity

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고준위폐기물을 심지층에 처분하기 위한 공학적방벽의 구성 요소로는 처분용기, 완충재, 뒷채움재 등이 있다. 이 중 완충재는 처분용기와 근계암반 사이의 빈 공간에 설치되는 물질로써, 주변 지하수로부터 처분용기를 보호하며 방사성 핵종의 유출을 저지하는 등의 역할을 한다. 또한 처분용기에서 발생하는 고온의 열량은 완충재로 직접 전파되기에 완충재의 열전도도는 처분시스템의 안전성 평가에 있어 매우 중요하다고 할 수 있다. 따라서 본 연구에서는 국내 경주산 압축 벤토나이트 완충재의 열전도도 특성을 규명하였으며 실제 처분용기에서 발생되는 고온의 특성을 반영하여 상온에서 80~90℃까지의 범위에서 압축 벤토나이트의 열전도도를 측정하였다. 온도증가에 따라 압축 벤토나이트의 열전도도는 5~20% 가량 증가하였으며 초기 포화도가 클수록 열전도도 증가는 더 크게 나타났다.

중심단어: 공학적방벽, 압축 벤토나이트 완충재, 열전도도

1. Introduction

Spent fuel from nuclear power plants can be classified as high-level waste with very high radiation levels and must be safely disposed of in multiple barrier (engineered and natural barriers) systems in rock layers 500 to 1,000 m deep [1]. Among such systems, the engineered barrier system (EBS) is an artificially developed concept composed of a canister, backfill, buffer, and gap-filling material. The buffer material is installed in the empty space between the disposal canister containing the spent fuel and the near-field rock. It serves to protect the disposal canister by minimizing physical shocks and the inflow of groundwater from the surrounding rock [2]. Disposal canisters generate high-temperature decay heat that should be rapidly propagated to the surroundings. To secure the integrity of the disposal canister, the buffer material propagates this decay heat and prevents heat from accumulating in the disposal canister [3]. Bentonite clay minerals composed of montmorillonite are known as the most suitable material that satisfies all performance requirements of buffer materials [4-6]. In montmorillonite, one aluminum plate is bonded to two silicon plates, and isomorphous substitution replaces the aluminum and iron in each plate with magnesium [1,7]. Due to the isomorphous substitution, the surface of montmorillonite becomes negatively charged and attracts cations such as Na^+ and Ca^{2+} to form a charge balance; these materials are classified into Na-type bentonite and Ca-type bentonite according to the species of exchangeable cations. In Korea, most research focuses on Ca-type bentonite as it is produced in Gyeongju. As a buffer material, bentonite blocks are used by compressing bentonite powder. Among the various criteria that must be met for a bentonite buffer to achieve proper performance, thermal conductivity is the most important factor in determining the set temperature of the buffer material [4, 8, 9].

Although many studies have been conducted on the thermal conductivity of bentonite buffers [8-13], most have been limited to the study of dry density and degree of saturation at room temperature. In disposal environments, the high-temperature heat generated from the disposal canister is propagated to the bentonite buffer, and groundwater penetrating from the near-field rock increases the degree of saturation of the buffer. Thus, the effects of the degree of saturation and the temperature of the buffer must be considered simultaneously. In the initial stages of disposal, the degree of saturation of the bentonite buffer is reduced by the heat generated from the disposal canister [1]. Due to subsequent saturation by groundwater, the degree of saturation will be constant even when high-temperature heat is transferred to the buffer. However, studies on the thermal conductivity of bentonite buffers according to temperature variations have been insufficient, and no further studies have been conducted on the thermal conductivity of

Table 1. Quantitative XRD analysis (wt%) for mineral constituents of KJ-II powders [13-15]

Bentonite Type	KJ-II			
Sample No.	1	2	3	Avg.
Montmorillonite	63.4	61.7	60.5	61.9
Albite (λ=1.96 W/(m·K))	19.4	22.8	20.4	20.9
Quartz (λ=7.69 W/(m·K))	5.8	4.9	5.3	5.3
Cristobalite (λ= 6.15 W/(m·K))	4.0	4.5	3.7	4.1
Calcite (λ=3.59 W/(m·K))	4.3	3.3	6.8	4.8
Heulandite (λ=1.09 W/(m·K))	3.0	2.7	3.3	3.0

Fig. 1. Steel mold and bentonite specimen.

Ca-type bentonite produced in Korea.

Therefore, this study measured the thermal conductivity of compacted bentonite produced in Korea for temperatures ranging from room temperature to 80~90℃, with consideration of the degree of saturation and dry density.

2. Thermal conductivity test

2.1 Materials and test equipment

In this study, the thermal conductivity of Gyeongju bentonite produced by Clariant Korea was measured. Prior to 2015, the Korea Atomic Energy Research Institute (KAERI) conducted research on Gyeongju bentonite samples designated as KJ-I; since 2015, research has been conducted on KJ-II [16]. As such, KJ-II was used in this study. Using the uniform classification method, KJ-II bentonite powder was classified as highly plastic clay [7], and the chemical composition was mostly SiO_2 and Al_2O_3; CaO exhibited a composition ratio approximately 5 ~ 6 times higher than that of NaO [16]. Table 1 shows the quantitative XRD analysis results for mineral constituents of KJ-II powders [13]. The KJ-II bentonite powder was placed in a steel mold, and compacted bentonite was produced with a certain density using a hydraulic press, as shown in Fig. 1.

Using the transient hot wire method, the thermal conductivity of the compacted bentonite was measured using a QTM-500 (Kyoto Electronics) instrument. The transient hot wire method is a principle for thermal conductivity measurement according to Equation (1): it involves the heating time and temperature rise relationship between heating wires when a constant amount of heat is applied to a heating wire in a medium [17].

$$\lambda = K \cdot R \cdot I^2 \cdot \ln(t_2/t_1)/(T_2 - T_1) - H \tag{1}$$

where λ is the thermal conductivity (W/(m·K)) of the sample, and K and H represent the probe constants, which are obtained by measuring reference specimens with known thermal conductivity values. The reference materials used in this experiment were Quartz (1.42 W/(m·K)), silicon rubber (0.24 W/(m·K)), and Styrofoam (0.036 W/(m·K)) [17]. In addition, I is the heating current (A), t_1 and t_2 represent measurement times (s), and T_1 and T_2 represent the temperature (K) at times t_1 and t_2. The precision and reproducibility of this method for measuring thermal conductivity is ±5% and ±3%, respectively.

2.2 Test procedure

In this study, the thermal conductivity values of the compacted bentonite samples were measured with increasing temperature. The compacted bentonite samples and

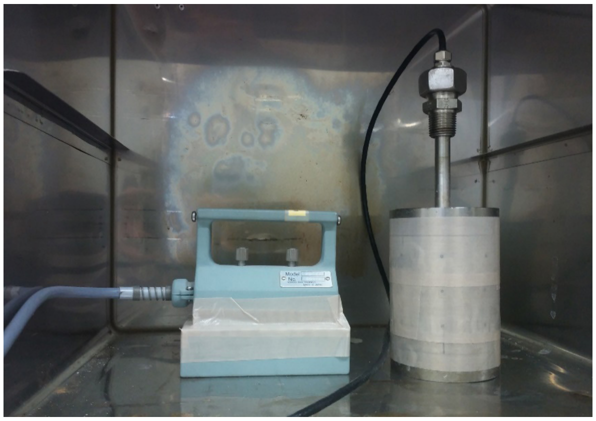

Fig. 2. Temperature measurements of the compacted bentonite.

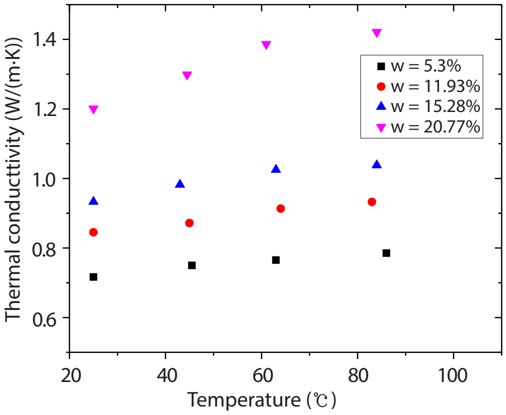

Fig. 3. Thermal conductivity variation considering various water content.

thermal conductivity measurement probe were sealed with silicon and heat-resistant tape to prevent water from escaping. The temperature was adjusted using a convection oven. A temperature sensor was inserted into a separate bentonite sample that was not used in the thermal conductivity test to measure the temperature of the bentonite, as shown in Fig. 2. The accuracy of the temperature sensor (HMT334, VAISALA) is ±0.2℃. The thermal conductivity was measured when the temperature of the bentonite became constant.

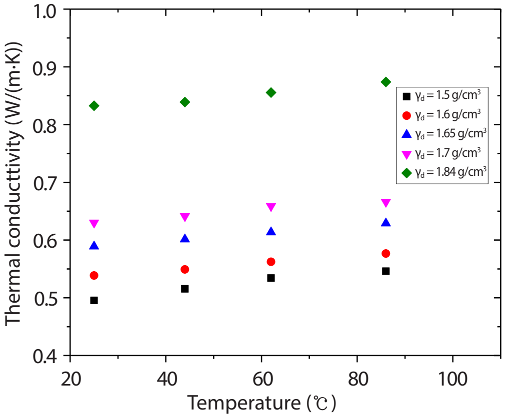

Fig. 4. Thermal conductivity variations (w=0%).

3. Test results

As dry density greater than 1.6 g/cm^3 is required for the buffer to achieve satisfactory performance [18], thermal conductivity was measured for compacted bentonite with a dry density of 1.61 g/cm^3 (±1%) according to temperature variations and water content. Fig. 3 shows the change in thermal conductivity from room temperature to 80~90℃ for various initial water contents. As the temperature increased, the thermal conductivity of the compacted bentonite increased by approximately 10~20% compared to that at room temperature. The higher the water content, the greater the thermal conductivity. Bentonite is a three-phase system consisting of soil particles, water, and air, and the thermal conductivity of soil is known to increase with temperature [9,19]. This is caused to the formation of water-islands [19] as the temperature increases by the movement of water vapor arising from the meniscus of water in the bentonite. Thermal conductivity increases with heat transfer by latent heat. For this reason, it is thought that higher initial water content results in greater increases in thermal conductivity [9,20].

In addition, thermal conductivity was measured according to temperature increases under dry conditions, in which the water in the compacted bentonite was removed. Fig. 4 shows the thermal conductivity of bentonite under dry

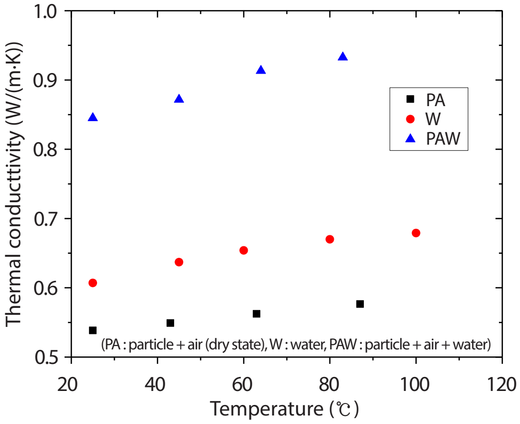

Fig. 5. Summary of thermal conductivity.

conditions for various dry densities; the rate of increase from room temperature to 80~90℃ was approximately 4~10%. This is due to the lack of water in the dry state, which means water-islands are not formed by steam movement. As such, the increase in thermal conductivity by air and soil particles is smaller than when water is present. In particular, as the dry density increases, heat transfer due to air movement is small, so the increase in thermal conductivity according to temperature increases is insignificant. In Spain, FEBEX bentonite also exhibited a linear increase in thermal conductivity of approximately 3~4% from room temperature to 80~90℃ [21, 22]. Fig. 5 shows the results of the thermal conductivity test and the thermal conductivity change of water when the dry density of the compressed bentonite was 1.6 g/cm^3 for initial water contents of 0 and 11.93% [23]. The results show that the thermal conductivity of the bentonite particles, water, and air increases with increasing temperature. However, it is known that the thermal conductivity of water increases at a slowing rate with increasing temperature, with thermal conductivity decreasing beyond 120℃ [23]. The thermal conductivity of the compacted bentonite under dry conditions, in which only bentonite and air are present, increased linearly in the temperature range from room temperature to 80~90℃. However, the increase rate of thermal conductivity with the three-phase condition slightly decreased in the same temperature range.

4. Conclusion

In disposal environments, bentonite buffers inevitably undergo temperature increases due to the high-temperature decay heat from the disposal canister in addition to groundwater inflow from the surrounding rock. With the thermal conductivity of compacted bentonite buffers being one of the most important parameters in the safety assessment of EBS, this study evaluated thermal conductivity variations in compacted Gyeongju bentonite by considering temperature increases and various initial water contents.

First, a compacted bentonite sample with a dry density of 1.6 g/cm^3 (±1%) was prepared by compressing Gyeongju bentonite powder in a press. Thermal conductivity was measured for temperatures in the range of 25℃ to 80~90℃ according to various initial water contents. The thermal conductivity of the compacted bentonite was measured using a QTM-500 instrument based on the principle of the transient hot wire method. The compacted bentonite samples and probe were completely sealed with heat-resistant tape and placed in a convection oven to measure thermal conductivity according to temperature variations.

Compared to the measured value at 25℃, the thermal conductivity of the compacted bentonite increased by approximately 10~20% at the 80~90℃ range. As the temperature increases, water-islands are formed by the movement of water vapor due to the meniscus structure of water. The thermal conductivity of bentonite, which forms a three-phase system, increases due to heat transfer by latent heat. The results showed that the higher the initial water content, the greater the thermal conductivity. In addition, the thermal conductivity of the compacted bentonite under dry conditions increased by approximately 4~10% at 80~90℃ compared to the measured values at 25℃, which was dependent on the initial dry density. As heat transfer by water does not occur under dry conditions, thermal conductivity did not exhibit significant increases. The results showed that the higher the dry density of the compacted bentonite, the smaller the increase in thermal conductivity. The thermal conductivity of the compacted bentonite in the three-phase system, in which soil particles, water, and air are all present, tended to increase as the temperature increased.

Through this study, it was found that the thermal conductivity of compacted bentonite increases by 5~20% as temperature increases, depending on the water content. Considering the mid to long-term conditions of actual disposal environments, even though heat is generated from disposal canisters, groundwater penetration form the near-field rock continuously increases the degree of saturation of the buffer, which naturally leads to an increase in the thermal conductivity of the bentonite buffer. In addition, the thermal conductivity of bentonite buffers was determined to increase to a greater degree in the temperature range of 80~90℃. Thus, the test results from this study can be effectively used to evaluate the soundness of bentonite buffers: the results are expected to be used as important data for repository design and engineering barrier performance evaluations for the goal of raising the maximum buffer temperature or reducing the overall disposal area.

Additionally, the thermal-hydro-mechanical properties and the thermal conductivity of bentonite buffers at the set temperature of 100℃ or higher should be investigated with consideration of actual complex repository conditions.

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