Hydraulic-Thermal-Mechanical Properties and Radionuclide Release-Retarding Capacity of Kyungju Bentonite

경주 벤토나이트의 수리-열-역학적 특성 및 핵종 유출 저지능

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Abstract

Studies were conducted to select the candidate buffer material for a high-level waste (HLW) repository in Korea. This paper presents the hydraulic properties, the swelling properties, the thermal properties, and the mechanical properties as well as the radionuclide release-retarding capacity of Kyungju bentonite as part of those studies. Experimental results showed that the hydraulic conductivities of the compacted bentonite were very low and less than 10^{-11}m/s. The values decreased with increasing the dry density of the compacted bentonite. The swelling pressures were in the range of 0.66 MPa to 14.4 MPa and they increased with increasing the dry density. The thermal conductivities were in the range of 0.80 kcal/mh°C to 1.52 kcal/mh°C. The unconfined compressive strength, Young's modulus and Poison's ratio showed the range of 0.55 MPa to 8.83 MPa, 59 MPa to 1275 MPa, and 0.05 to 0.20, respectively, when the dry densities of the compacted bentonite were 1.4 Mg/m^3 to 1.8 Mg/m^3. The diffusion coefficients in the compacted bentonite were measured under an oxidizing condition. The values were 1.7×10^{-10} m^2/s to 3.4×10^{-10} m^2/s for electrically neutral tritium (H-3), 8.6×10^{-14} m^2/s to 1.3×10^{-12} m^2/s for cations (Cs, Sr, Ni), 1.2×10^{-11} m^2/s to 9.5×10^{-11} m^2/s for anions (I, Tc), and 3.0×10-14 m^2/s to 1.8×10^{-13} m^2/s for actinides (U, Am), when the dry densities were in the range of 1.2 Mg/m^3 to 1.8 Mg/m^3. The obtained results will be used in assessing the barrier properties of Kyungju bentonite as a buffer material of a repository in Korea.

요약

고준위방사성폐기물 처분장의 완충재 후보물질 선정을 위해 경주 벤토나이트를 대상으로 수리특성, 팽윤특성, 열적특성, 역학특성 및 핵종유출 저지특성을 조사하였다. 실험결과, 압축 벤토나이트의 수리전도도는 10^{11} m/s 이하로 매우 낮았으며 건조밀도가 증가할수록 감소하였다. 팽윤압은 0.66 MPa ~14.4 MPa 사이의 값을 보였으며 건조밀도에 따라 증가하였다. 건조밀도가 1.4 Mg/m³ ~ 1.8 Mg/m³일 때, 열전도도, 일축압축강도 (unconfined compressive strength), 탄성계수 (Young's modulus of elasticity), Poisson 비는 각각 0.80 kcal/mh℃ ~ 1.52 kcal/mh℃, 0.55 MPa ~ 8.83 MPa, 59 MPa ~ 1275 MPa, 0.05~0.20의 값을 나타내었다. 압축벤토나이트 에 대한 핵종 확산계수는 산화환경에서 측정되었으며, 주어진 실험조건에서 삼중수소 (H-3)는 1.7×10^{10} m²/s ~ 3.4×10^{10} m²/s, 양이온 핵종 (Cs, Sr, Ni)은 8.6×10^{14} m²/s ~ 1.3×10^{12} m²/s, 음이온 핵종 (I, Tc)은 1.2×10^{11} m²/s ~ 9.5×10^{11} m²/s, 악티나이드 핵종은 3.0×10^{14} m²/s ~ 1.8×10^{13} m²/s 사이의 값을 나타내었다. 이 때 확산계수는 모든 핵종에 대해 압축벤토나이트의 건조밀도가 증가할수록 감소하는 경향을 보였다.

중심단어 : 방벽특성치, 벤토나이트, 완충재, 고준위폐기물처분장

I. Introduction

A HLW repository in Korea is constructed in a bedrock of several hundred meters in depth [1]. The HLWs are encapsulated in disposal containers, which are deposited into boreholes on the floor of the emplacement rooms. The gap between the container and the wall of a borehole is then filled with a buffer material and the inside space of the emplacement rooms with a backfill material.

In this design concept, the buffer plays the roles of minimizing the water flux into the repository and restricting the release of radionuclides into the host environment. It also dissipates the decay heat from waste into the surrounding rock to avoid the possibility of thermal stress on the container and a high temperature resulting in the loss of the desirable functions of the buffer. Moreover, it supports the container and waste from external mechanical stress.

Bentonite has been favored as a candidate buffer material in many countries because it has low hydraulic conductivity, high sorption capacity, self-sealing characteristics, and durability in nature [2]. The design concept of a Korean repository as well is considering the use of domestic bentonite as the buffer material [1].

The present study, in this connection, investigates the hydraulic-thermal-mechanical properties and radionuclide release-retarding capacity of Kyungju bentonite to see how well the bentonite meets the functional criteria of a buffer for a constituent barrier of repository.

II. Kyungju Bentonite

In Korea, the bentonite is produced mainly from tertiary sediments in the southeastern area of the peninsula. The Kyungju bentonite used in this study is a product by the Taekwang Chemical Co., which was taken from Jinmyeong mine located in Kyungju, Kyungsangbuk-do. It was dried below 110°C, pulverized, and passed through No. 200 of ASTM(American Society for Testing and

Table 1. Chemical composition of the bentonite.

Chemical Constituent	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	K_2O	Na_2O	FeO	SO_3	TiO_2
Percentage(%)	56.8	20.0	6.00	2.6	0.8	0.9	1.3	0.2	1.3	0.8
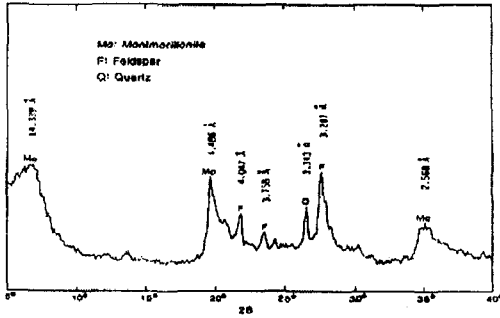
Figure 1.X-ray diffraction pattern of the bentonite.
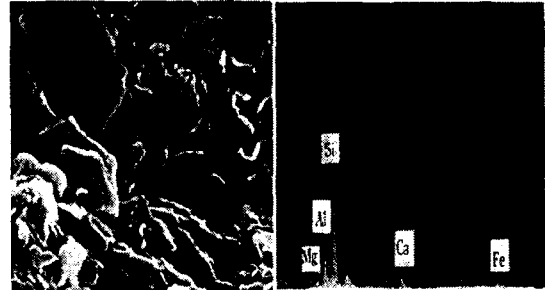
(a)SEM micrograph   (b) EDX pattern

Figure 2. Results of SEM and EDX analysis for the bentonit

Materials)standard sieves. It is similar to those used in the previous studies [3] although the mining companies are different. The X-ray diffraction pattern is shown in Fig. 1 and the results of SEM(Scanning Electron Microscope) and EDX(Energy Dispersive X-ray) analysis in Fig. 2 [13]. The bentonite contains montmorillonite (70%), feldspar (29%), and small amounts of quartz (~ 1%), and its chemical composition is listed in Table 1. It has a cation-exchange capacity of 57.6 meq/100g

III. Hydraulic Properties

The hydraulic conductivities in water-saturated bentonites with the dry densities of 1.4 Mg/m^3 to 1.8 Mg/m^3 were measured. They were very low and less than 10^{-11} m/s [4]. It isassumed that the high swelling potential of the bentonite contributes significantly to the development of low resultant hydraulic conductivities. The hydraulic conductivities decreased with an increasing dry density of the bentonite. The logarithm of the hydraulic conductivity at 20°C versus the dry density of the

bentonite has been plotted in Figure 3. The relation between the logarithm of the hydraulic conductivity and the dry density of the bentonite can be fitted to a straight line expressed as follows:

$$\log K = -4.07 \text{ rho } d - 6.13 \text{ (at 20°C)} \cdots\cdots\cdots (1)$$

The hydraulic conductivities of bentonite with dry densities of 1.4 Mg/m^3, 1.6 Mg/m^3 and 1.8 Mg/m^3 as a function of temperature are presented in Figure 4 to Figure 6. As shown in these figures, the hydraulic conductivities increase with an increasing temperature. The hydraulic conductivities of the bentonite with dry densities of 1.4 Mg/m^3 to 1.8 Mg/m^3 at a temperature of 150°C are up to about one order of magnitude higher than the hydraulic conductivities at 20°C. The change of hydraulic conductivities in the bentonite samples at the elevated temperatures is attributable to the changes in the viscosity factor [5]. Although the hydraulic conductivity increases with temperature, the experimental results also indicate that the hydraulic conductivities of the compacted bentonite with dry densities of 1.6 and 1.8 Mg/m^3 are still low and are
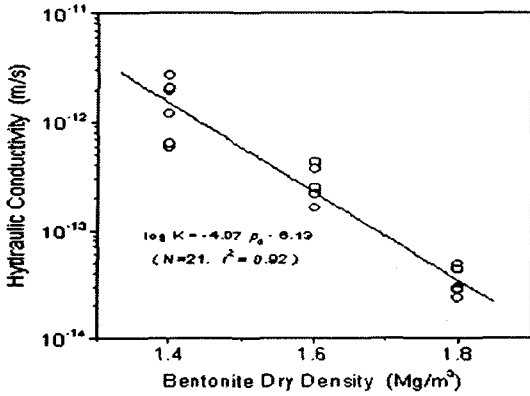
Figure. 3. Hydraulic conductivity versus the dry density of compacted benonite at 20°C

Figure 4. Hydraulic conductivity of compacted bentonite with a dry density of 1.4 Mg/m³ at different temperatures (symbols: measured over different samples)

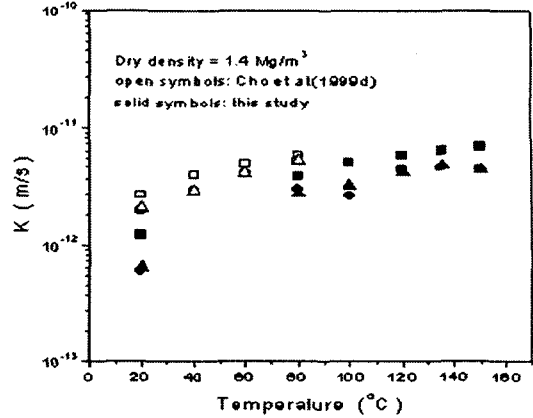**Figure 5. Hydraulic conductivity of compacted bentonite with a dry density of 1.6 Mg/m³ at different temperatures (symbols: measured over different samples)**
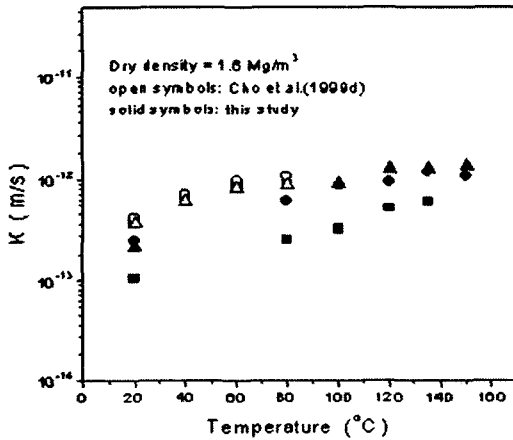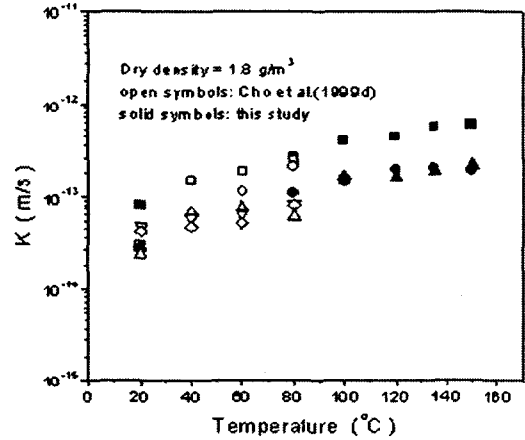

Figure 6. Hydraulic conductivity of compacted bentonite with a dry density of 1.8 Mg/m³ at different temperatures (symbols: measured over different samples)

in the order of 10^{-12} m/s even at a temperature of 150 ℃.

IV. Swelling Properties

The free swell index of the Kyungju bentonite measured by the Forster's method was 89.1 ± 2.8 ml/10g of dry bentonite. The typical swelling pressure characteristics of the compacted bentonite at 20°C are presented in Figure 7 [6]. The swelling pressure developed rapidly over the early period, and then reached a nearly constant value after 15

days. The transient behavior of the swelling pressure can be explained by the changes in the fabric structure of the bentonite and moisture redistribution [7]. The swelling pressures were measured after 30 days assuming a steady state, and the vertical swelling pressures were found to be higher than the horizontal ones suggesting an anisotropic structure of the compacted bentonite. When the dry densities were 1.4 to 1.8 Mg/m³, the swelling pressures were in the range of 6.6 kg/cm² to 143.5 kg/cm² and increased with an increasing dry density (Figure 8). The dependency of the swelling pressures on the dry
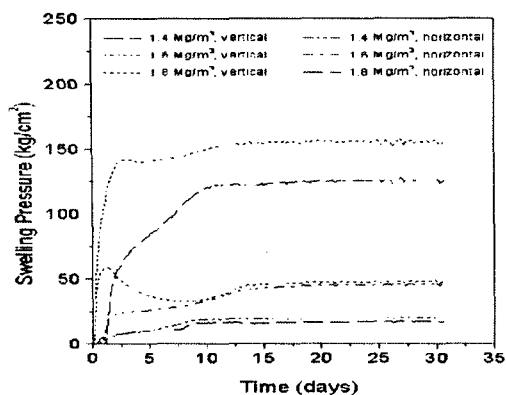
Figure 7. **Typical swelling pressure development pattern for the compacted bentonite at 20°C.**
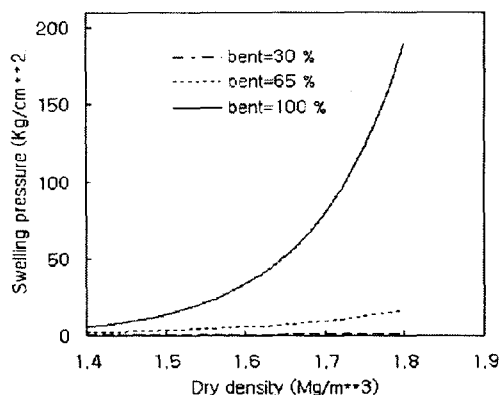

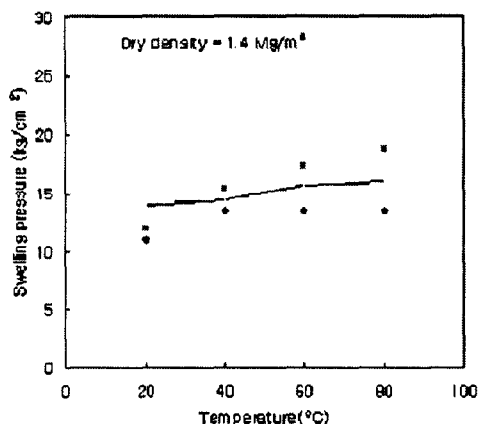Figure 8. **Effect of dry density on the swelling pressure.**

Figure 9. **Swelling pressure of compacted bentonite with a dry density of 1.4 Mg/m³ at different temperatures.**
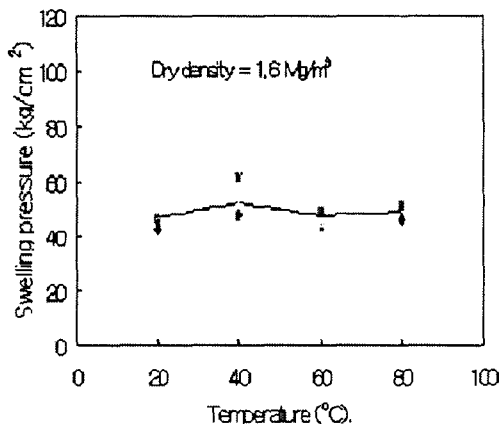

Figure 10. **Swelling pressure of compacted bentonite with a dry density of 1.6 Mg/m³ at different temperatures.**

density was more sensitive in a higher dry density. The swelling pressures of the bentonite for dry densities of 1.4 Mg/m³ and 1.6 Mg/m³ in the range of temperatures from 20°C to 80°C are 12 kg/cm² to 18 7 kg/cm² and 46.2 kg/cm² to 51.2 kg/cm², respectively (Figure 9 and 10), although there is an outlier of more than 60 kg/cm² for a dry density of 1.6 Mg/m³ at 40°C which is a possible experimental deviation [8]. As shown in the figures, the swelling pressures of the compacted bentonite increased with increasing temperature, but the change was not large. This phenomenon can be explained by the

changes of the hydration pressure, osmotic pressure and pore water pressure with increasing temperature. The hydration pressure decreases rapidly by increasing the temperature because of the reduction of the water molecular layer on the surface of the bentonite [9]. On the other hand, the osmotic pressure increases due to the reduction of the thickness of the electric double layer [9, 10] and the pore water pressure is also increased due to the differential expansion of the pore water and the skeleton [11, 12]. The latter two effects almost exactly counterbalance the former. The

Table 2. Thermal conductivities of the compacted bentonite

Specimen, No.	Dry density, g/cm^3	Water content, %	Degree of saturation, %	Thermal conductivity, Kcal/mh°C	Specimen temperature
1	1.8	15.0	77.8	1.312	
2	1.8	17.2	89.1	1.313	23°C
3	1.8	19.0	98.9	1.520	
4	1.6	17.2	65.5	1.016	"
5	1.6	26.0	99.5	1.273	
6	1.4	17.2	48.9	0.801	"
7	1.4	34.6	98.8	1.045	

Table 3. Unconfined compressive strength, Young's modulus and Poison's ratio for the compacted bentonite

Dry density (g/cm^3)	Water Content (%)	Cohesion (kPa)	Internal friction angle(degree)
1.8	17	1100	50
1.6	17	1000	37
1.4	17	500	27

Table 4. shear properties of the compacted bentonite

Dry density (g/cm^3)	Water Content (%)	Unconfined compressive Strength(MPa)	Young's modulus ($\times 10^2$MPa)	Poisson's ratio
1.4	17	0.55	0.59	0.05
1.6	17	4.90	7.84	0.20
	17	4.90	5.88	0.18
1.8	17	7.84	10.79	0.21

experimental results showed that the influence of the temperature on the swelling pressure is not important if the maximum temperature is maintained below 100°C

V. Thermal Properties

The thermal conductivities of the Kyungju bentonite were measured using a Quick Thermal Conductivity Meter [13], and the results are shown in Table 2. The thermal conductivities are in the range of 0.80 to 1.52 kcal/mh°C in the case of the dry density of 1.4 to 1.8 Mg/m^3. The thermal conductivity increases by increasing the dry density and the water content of the bentonite.

VI. Mechanical Properties

The unconfined compressive strength, Young's modulus, Poison's ratio, and shear properties of the compacted Kyungju bentonite were measured [14]. The compaction test was performed using the ASTM D 1557-78 Method A [15]. The dry density-water content relationships are presented in Figure 11. This compaction curve shows that, as the molding water content is increased, the dry density increases to a maximum value and then decreases. The addition of water increases the amount of free water in the voids. This decreases the intergranular, or interped, shearing resistance and aid compaction. The stress, the axial strain and the lateral strain for the compacted bentonite were measured using the unconfined compression tester (Material Test System 815, USA). The stress-strairelationship for the bentonite specimen is shown in Figure 12. Young's module is the tangent modulus corresponding to 40 % of the curve (vertical stress), and the Poisson's ratio is a ratio between the axial strain and the lateral strain. The results are listed in Table 3. The shear strength of the compacted bentonite was
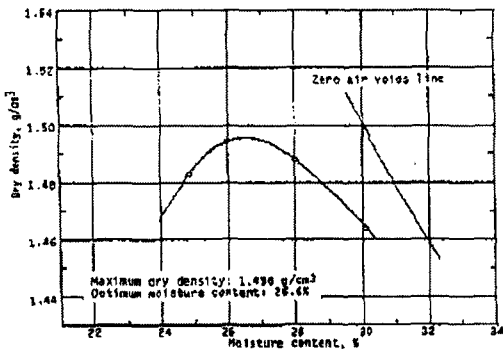
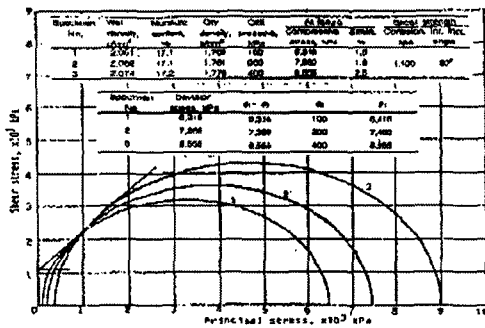**Figure 11. Moisture content-dry density relation-ship of the bentonite**

Figure 12. Stress-strain relationship of the bentonite

Figure 13. More's circle diagram with the results of the triaxial compression test.

(a)In-diffusion test method   (b) through-diffusion test method

Figure 14. Experimental apparatus used in measurement of diffusion coefficients

measured using the triaxial compression test under an unconsolidated-undrained condition(Figure 13). The shear properties of tcompacted bentonites are summarized in Table 4. The consolidation tests were performed by the oedometer (ELX, UK) using the applied load of 200 kN/m², 400 kN/m², 800 kN/m² and 1,600 kN/m², respectively. The coefficient of the volume change and the coefficient of the consolidation are in the range of 0.00249 to 0.02142 m²/MN and 0.018 to 0.115 m²/year, respectively.

VII. Radionuclide Release-Retarding Capacity

One of the major functions of a buffer from the chemical point of view is to retard the release of radionuclides from the wastes to the surrounding environment. Previous hydraulic tests [4] with Kyungju bentonite revealed that the hydraulic
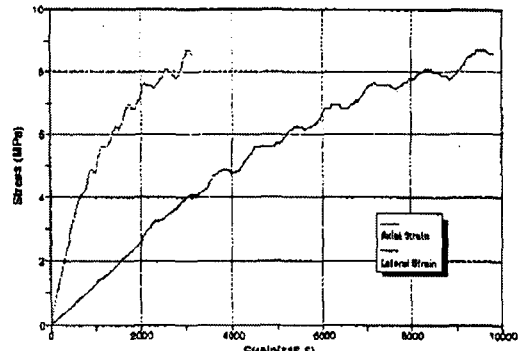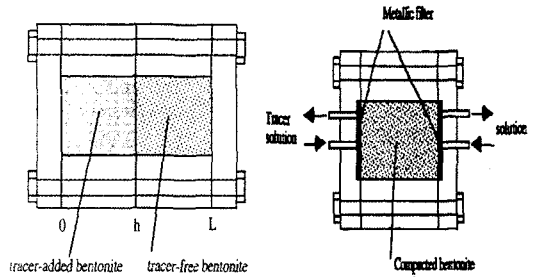
conductivities were below 10^{-11}m/s for the compacted bentonite with a dry density of more than 1.6 Mg/m³. If such a highly compacted bentonite is employed for the buffer of a repository, the advection and dispersion can virtually be ignored and thus the transport mechanism of the radionuclide will be controlled by diffusion, as reported in other papers [16, 17]. Determining the diffusion coefficient and understanding the diffusion behavior of the radionuclide in compacted bentonite, therefore, is required for the assessment of the radionuclide release through the buffer of the repository.

Measurements of the diffusion coefficients were conducted by either an in-diffusion test or a through-diffusion test. Literature [18] reported that these two diffusion tests gave negligible differences in the values of the diffusion coefficient. In the in-diffusion test, two sections of bentonite plug are saturated with solutions and a tracer solution is

added to one of the bentonite sections. The sections are compacted to the same density and brought into contact with each other in the diffusion cell, as shown in Figure 14(a). After a predetermined diffusion period, the bentonite specimens are removed from the diffusion cell and sectioned into slices. The concentration profile in the bentonite plug and subsequently the diffusion coefficients are determined [19]. In the through-diffusion test, a compacted bentonite plug is sandwiched between two reservoirs, as shown in figure 14(b). Initially, one of the reservoirs contains a tracer, whereas the other one is free of a tracer. The tracer penetrates from the high concentration reservoir through the bentonite specimen into the other reservoir. The tracer concentration is monitored in an initially trace-free solution. This provides a cumulative amount of tracer diffused through the bentonite plug. When the amount of radionuclide transported through the plug is at the stage of a steady state, the apparent diffusion coefficients are determined by comparing the theoretical values and experimental breakthrough curve at the steady state of the diffusion [20].

The experimental measurement of the diffusion coefficients have been conducted for 8 major radionuclides(i.e., neutral H^3, cations Cs, Sr, Ni; anions I, Tc; actinides U, Am). The measurements were conducted under an oxidizing condition. The dry densities of the compacted bentonite were given in the range of 1.2 Mg/m^3 to 1.8 Mg/m^3, and the temperature was maintained at 20°C. Table 5 lists the measured diffusion coefficients. As shown in the table, under oxidizing conditions, the diffusion coefficients were 1.7×10^{-10} m^2/s to 3.4×10^{-10} m^2/s for electrically neutral tritium, 8.6×10^{-14} m^2/s to 1.3×10^{-12} m^2/s for cations, 1.2×10^{-11} m^2/s to 9.5×10^{-11} m^2/s for anions, and 3.1×10^{-15} m^2/s to 1.8×10^{-13} m^2/s for actinides, when the dry densities were in the

Table 5. Apparent diffusion coefficients measured from diffusion tests.

Element		Dry density (Mg/m^3)	D_a ($\times 10^{-12}$ m^2/s)	Test method[2]
Neutral	Tritium	1.4	343	ID
		1.6	222	"
		1.8	173	"
Cation	Cs	1.4	0.39	ID
	Sr	1.2	1.32	TD
		1.4	1.28	"
		1.7	1.20	"
	Ni	1.4	0.20	ID
		1.6	0.14	"
		1.8	0.086	"
Anion	I	1.2	71.2	TD
		1.4	58.0	"
		1.7	12.5	"
	Tc	1.4	94.6	ID
		1.6	45.3	"
		1.8	34.6	"
Actinides	U	1.4	0.177	ID
		1.6	0.057	"
		1.8	0.030	"
	Am	1.4	0.0073	ID
		1.6	0.0058	"
		1.8	0.0031	"

(1) Test conditions: domestic bentonite, distilled water, oxidizing condition
(2) ID: in-diffusion test method, TD: through-diffusion test method

range of 1.2 Mg/m^3 to 1.8 Mg/m^3. The diffusion coefficient of tritium was similar in the order of magnitude to the water diffusion coefficient. Cations had the following order for the value of diffusion coefficient: Sr > Cs > Ni. The low diffusion coefficients of nickel could be explained by its precipitation at the high pH of the solution. Iodine and technetium as anions had the diffusion coefficients one order of magnitude less than tritium, which was probably attributed to the anion exclusion between the anions and bentonite particles with a negative surface charge [21]. Uranium and americium of the actinide series were found to have a wide range of diffusion coefficients. This may be

attributed mainly to their complex chemistry and speciation in the water-bentonite system. The uranium existed largely as an anionic carbonate complex in the pore solution of the bentonite under the oxidizing conditions. Its diffusion is expected to be dependent on the anion-exclusion and the pore structure of the compacted bentonite, but it was negligibly affected by the sorption of uranium onto the bentonite particles [22]. The americium revealed that the precipitation of the hydrous and carbonate americium complexes existed at the high pH region of the solution. It caused much lower diffusion coefficients than those of the other radionuclides. For the americium, the diffusion seems to be dependent on the precipitation, the sorption of americium onto the bentonite particles as well as the pore structure of the compacted bentonite. A combination of these affecting factors could probably explain the changes in the diffusion coefficients of the uranium and americium in the compacted bentonites.

VIII. Conclusions

The studies on Kyungju bentonite gave experimental data and information which is useful in assessing its feasibility and suitability as a candidate buffer material of a Korean repository. The hydraulic conductivities of the compacted bentonite were very low and less than 10^{-11}m/s above the dry density of 1.6 Mg/m^{3}and they decreased by increasing the dry density of the compacted bentonite. The swelling pressures were in the range of 0.66 MPa to 14.4 Mpa under the given experimental conditions. They were sensitive to the dry density, but the temperature change between 20°C and 80°C did not have a large effect on the swelling pressures.The thermal conductivities were in the range of 0.80 kcal/mh°C to 1.52 kcal/mh°C. The unconfined compressive

strength, Young's modulus and Poison's ratio showed the range of 0.55 MPa to 8.83 MPa, 59 MPa to 1275 MPa, and 0.05 to 0.20, respectively, when the dry densities of the compacted bentonite were 1.4 Mg/m^3 to 1.8 Mg/m^3. The diffusion coefficients of the major radionuclides in the compacted bentonite were 1.7×10^{-10} m^2/s to 3.4×10^{-10} m^2/s for electrically neutral tritium, 8.6×10^{-14} m^2/s to 1.3×10^{-12} m^2/s for cations, 1.2×10^{-11} m^2/s to 9.5×10^{-11}m^2/s for anions, and 3.0×10^{-14} m^2/s to 1.8×10^{-13} m^2/s for actinides, when the dry densities were in the range of 1.2 Mg/m^3 to 1.8 Mg/m^3.

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