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《Technical Note》

Basic Physicochemical and Mechanical Properties of Domestic Bentonite for Use as a Buffer Material in a High-level Radioactive Waste Repository

W.J. Cho, J.O. Lee, and K.S. Chun
Korea Atomic Energy Research Institute
150 Dukjin-dong, Yusong-gu, Taejon 305-353, Korea
wjcho@nanum.kaeri.re.kr

D.S. Hahn
Korea Institute of Geology, Mining & Materials
P.O. Box 111 Yusong, Taejon 305-350, Korea

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Abstract

The physicochemical, mineralogical, hydraulic, swelling and mechanical properties of a domestic bentonite for use as the buffer material in a high-level waste repository have been measured. The bentonite is identified to be a Ca-bentonite, and the hydraulic conductivity of the compacted bentonite with the dry density higher than 1.4 Mg/m^3 is lower than 10^{-11} m/s. When the dry densities are 1.4 to 1.8 Mg/m^3, the swelling pressures are in the range of 6.6 to 143.5 kg/cm^2. The unconfined compressive strength is about 94 kg/cm^2, and the coefficient of volume change and the coefficient of consolidation are in the range of 0.00249 to 0.02142 m^2/MN and 0.018 to 0.115 $m^2/year$, respectively.

Key Words : buffer material, high-level waste repository, physicochemical, mineralogical, hydraulic, swelling, mechanical properties, bentonite

1. Introduction

Clays are being used increasingly as sealing materials in the design of disposal facilities for hazardous wastes. Placed as engineered barriers, the materials are required to inhibit the migration of contaminants from the waste to the surrounding environment. A repository for high-level radioactive wastes would be constructed in the bedrock deep below ground surface, and the disposal safety can be assured through the combinational function of multi-barriers, namely, the engineered barriers such as buffer and backfill, and the natural barrier such as geosphere. The present design concepts [1,2] of an underground repository in granite formation for the disposal of high-level wastes include the use of compacted clay-based materials as both buffer surrounding waste containers and separating them from the host rock, and backfill for tunnels and access shafts.

In the repository, the buffer material should have a low hydraulic conductivity and high sorption capacity in order to minimize the penetration of groundwater and to restrict the release of radionuclides into the host environment when the waste container would be breached. The buffer material should also be required to have good mechanical properties to support the waste container without significant deformation resulting in the formation of void either under the weight of the container and other imposed loads. The cracks in the bentonite due to shrinkage should also not occur when the temperature is increased because of decay heat.

Bentonite has been considered as a candidate buffer material in the underground repository for the disposal of high-level radioactive waste because of its low permeability, high sorption capacity, self sealing characteristics, and durability in nature [3]. Bentonites are classified into two groups, that is, Na-bentonite and Ca-bentonite depending on the type of exchangeable cations existing in the interlayers of bentonite particle. Na-bentonite is generally considered to be preferable for the buffer material because of high swelling potential and high sorption capacity. However most countries have considered the use of domestic bentonite without distinction of types of bentonite for buffer material in a high-level waste repository because the required quantities of bentonite would be large. In Korea, bentonite is mainly produced from tertiary sediments in eastern Kyungsangbuk-do, and several studies have been carried out to investigate its possible use in a radioactive waste repository. Choi et al. [4,5] investigated the basic properties such as surface area, free swell rate and radionuclide distribution coefficient of heat-treated and untreated domestic bentonites. Cho et al. [6] summarized the hydraulic and diffusive properties and the mechanical properties of the low-density bentonite and the bentonite-crushed rock mixture for a backfill in the low- and intermediate-level waste repository. For the buffer material in a high-level waste repository, the bentonite should be compacted to a high density, and the sealing and mechanical properties of highly compacted bentonite are different considerably from those of low-density bentonite and bentonite-crushed rock mixture. These properties of domestic highly compacted bentonite have however not been investigated and are needed to be characterized.

This study intended to present the physical, chemical and mineralogical properties of the domestic bentonite. The hydraulic, swelling and mechanical properties of highly compacted bentonite were also analyzed.

2. Experimental

2.1. Bentonite

The bentonite used in this study was a product mined by Taekwang Chemical Co., Kyungju, Kyungsangbuk-do, and will be refered hereafter as 'Kyungju bentonite' . The bentonite is similar to those used in previous studies [4-6] although the mining companies are different, and is untreated except for drying and powdering. In the experiment, the portion of bentonite being passed through a 200 mesh of ASTM standard sieve were used. This bentonite is a potential buffer material for a high-level waste repository.

2.2. Physical Analyses

The physical analyses of the Kyungju bentonite including the determination of density, specific surface area, Atterberg limits and linear shrinkage were performed. The test procedures are as follows:

Density : The density of bentonite was measured by using the test procedure of ASTM D 854-83 (KS F 2308) [7].

Total surface area : The total surface areas of bentonite and montmorillonite were measured by using EGME (Ethylene Glycol Monoethyle Ether) technique suggested by D. L. Carter [8]

Atterberg' s limit : The liquid limit and the plastic limit were measured by using the procedures of ASTM D 4318-84 (KS F 2303 and KS F 2304) [7].

Linear shrinkage : The measurements were carried out by BS 1377: 1975, Test 5 [9]. About 150 g of dry bentonite was mixed with distilled water to meet the liquid limit condition, and then was filled into the cylindrical mold (50 mm(D) × 160 mm(L)). The inner surface of the mold was coated with the thin film of silicon grease. The bentonite in the mold was dried in air at first and then in the convective oven at 60℃. After shrinkage, the temperature was increased to 110 ℃ for additional drying.

2.3. Chemical Analyses

The cation exchange capacity, pH and the chemical composition of the bentonite were measured. For measurement of pH, the Electrometric Method (BS 1377: 1975, Test 11(A)) was used [9].

2.4. Mineralogical Analyses

For the characterization of mineralogical properties of the bentonite, X-ray diffraction (XRD) analysis, Scanning Electron Microscope (SEM) analysis, and Energy Dispersive X-ray (EDX) analysis were performed.

XRD : To determine the mineralogical composition of bentonite, a Philips PW 1730 diffractometer and X' Pert-MSD were used.

SEM and EDX : SEM(Scanning Electron Microscope) and EDX(Energy Dispersive X-ray) analyses were performed to investigate qualitatively the texture and chemical elements of montmorillonite in the bentonite, using the JSM 6400 (JEOL, Japan) and the SERIES II (NORAN, USA), respectively.

2.5. Hydraulic Analyses

To measure the hydraulic conductivity of compacted bentonite, the modified constant head test procedure using constant helium gas pressure was applied [10,11]. The apparatus used to determine the hydraulic conductivity is designed to supply water to the sample at 9 to 20 kg/cm^2 depending on the dry density of bentonite. The stainless steel cylindrical chamber has an inside diameter of 50 mm. The bentonite samples are rigidly confined using a restraining ram. The compacted samples with pre-determined dry density were placed in the sample chamber, and the water flows from the bottom to the top of the sample chamber. The hydraulic conductivities of the samples were then determined at room temperature (20℃) using deionized water. The penetrated water volumes were measured by weighing. The hydraulic conductivity at a given experimental condition was determined when equilibrium was reached.

2.6. Swelling Analyses

Free swell rate : The free swell rate of bentonite was measured by using the technique suggested by Gibbs and Holtz [12]. The pre-determined weight (10g) of air-dried bentonite powder was freely dropped slowly into the distilled water (50 ml) in a 50 ml cylinder in order to avoid the possibility of agglomeration of clay particles. After a sufficient time, the free swell rate was determined from the swelled volume in the distilled water.

Swelling pressure : The apparatus used to measure the swelling pressure of compacted bentonite includes four sections: main test section, pressurized water-feeding section, vacuuming section, and signal and data processing section. The main test section consists of a stainless steel cylindrical cell of which the dimension is 30 mm in height and 50 mm in diameter, porous metal filters, and swelling pressure sensors mounted in both vertical and horizontal directions. In the pressurized water-feeding section, the water in a stainless steel reservoir is pressurized by helium gas and fed to the main test section. In the vacuuming section, a vacuum pump is connected to the outlet of the main test section to pull out the air entrapped in the compacted sample. The signal and data processing section is designed to acquire swelling pressures from pressure sensor and then to display their values on digital indicator.

2.7. Mechanical Analyses

The mechanical analyses of bentonite include compaction test, unconfined compression test, and consolidation test.

Compaction test : The test was performed using ASTM D 1557-78 Method A [7]. The clay samples were compacted by drop of a rammer with the weight of 4.5 kg from the height of 457 mm.

Unconfined compression test : The bentonite of 180.4 g was divided into three equal parts. Each part was put into the mold, and was compacted using temper and compacter to the dry density of 1.73 Mg/m^3. The test specimens have a diameter of 38 mm and a height of 76 mm. The stress, the axial strain and the lateral strain were measured by using the unconfined compression tester (Material Test System 815, USA).

Consolidation test : The bentonite with water content of 20.3 wt% was compacted to the dry density of 1.75 Mg/m^3 (wet density of 2.10 Mg/m^3) to prepare the specimen for the consolidation test. The diameter and height of test specimen are 50 mm and 20 mm, respectively, and the initial void ratio and the initial saturation were 0.567 and 98 %, respectively. The consolidation tests were performed by using the oedometer (ELX, UK) for the applied load of 200 kN/m^2, 400 kN/m^2, 800 kN/m^2 and 1,600 kN/m^2, respectively.

3. Results and Discussion

3.1. Physical Properties

The experimental results for the density and the total surface area of bentonite are listed in Table 1. According to its water content, a clay can be distinguished as three states, namely liquid, plastic and solid. Liquid limit (LL) separates liquid state from plastic state, and Plastic Limit (PL) separates plastic state from solid state The plasticity index (PI) is defined as;

$$PI = LL - PL$$

The liquid limit and plastic limit, and the plasticity index of Kyungju bentonite are presented in Table 1. The results show that Kyungju bentonite has high volume change potential [13]. The measured PI is however about a half of that for a typical Ca-montmorillonite [14] and it implies the presence of considerable amounts of other components in Kyungju bentonite. The PI of domestic bentonite is much lower than that of MX-80 (~480%) [15] and Japan Kunigel V1 (~395%)

Table 1. Summary of the Physical Properties of the Bentonite

Density (g/cm^3)	Specific surface area (m^2/g)	Atterberg limits (%)		
		LL	PL	PI
2.74	347.6	244.5	46.1	198.4

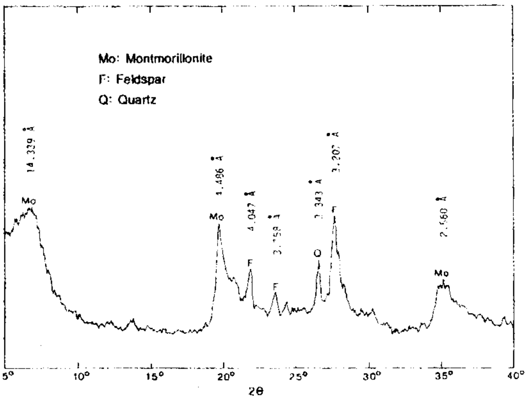

Fig. 1. X-ray Diffraction Patterns of Kyungju Bentonite

[16] that both are Na-bentonites. But it is similar to that of Canada Avonlea Na-bentonite (> 200%) [17] and is higher than that of French Fo-Ca smectite (50 ~ 100%) [18]. The shrinkage of bentonite as a function of water content represents the linear relationship, and the linear shrinkage (LS) was calculated from the following equation:

$$LS (\%) = (1 - L_s/L_m) \times 100$$

where L_s is the length of dry specimen, and L_m is the length of mold. The measured linear shrinkage is 35 %.

3.2. Chemical Properties

The chemical compositions of the bentonite are given in Table 2, As shown in the table, Kyungju bentonite contains approximately 56.8 % SiO_2, 20.0 % Al_2O_3, 6.0 % F_2O_3, and some minor elements. The cation exchange capacity (CEC) and pH of bentonite are 57.6 meq/100 g, and 9.5, respectively. The obtained CEC is similar to those by Choi et al [4], and is slightly higher than that of Fo-Ca smectite [18]. The CEC of MX-80 bentonite is 75 meq/100 g [4].

Table 2. Chemical Composition of the Bentonite

Chemical constituents	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	K_2O	Na_2O
weight %	56.80	19.96	6.03	2.59	0.77	0.93	1.25

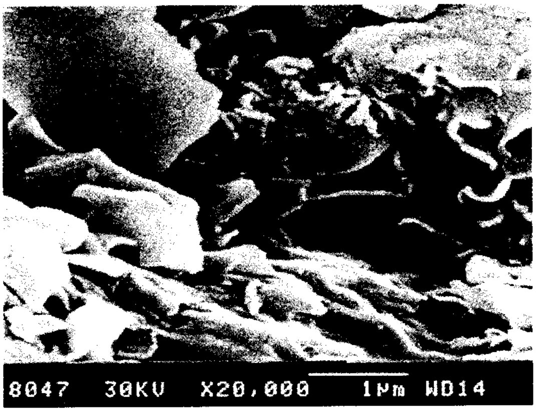

Fig. 2. Scanning Electron Micrograph of Kyungju Bentonite Showing Curled Habit of Montmorillonite Aggregates

3.3. Mineralogical Properties

The XRD patterns of the bentonite are illustrated in Fig. 1. The bentonite contains montmorillonite, feldspar and quartz. The results of SEM analysis and EDX analysis are shown in Fig. 2 and Fig. 3, respectively. These indicate that the bentonite contains Si, Al, Ca, Mg and Fe. From the location of peaks in the XRD pattern and the chemical composition that Ca^{++} is a predominant exchangeable cation, the Kyungju bentonite was classified to be a Ca-bentonite.

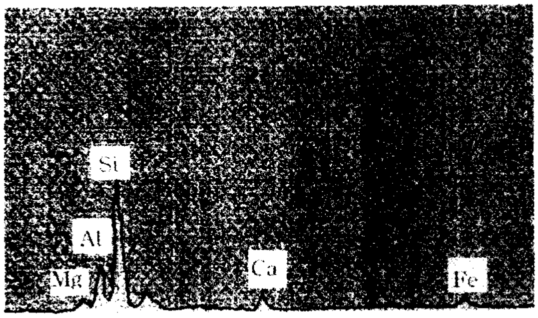

Fig. 3. EDX Pattern of Kyungju Bentonite

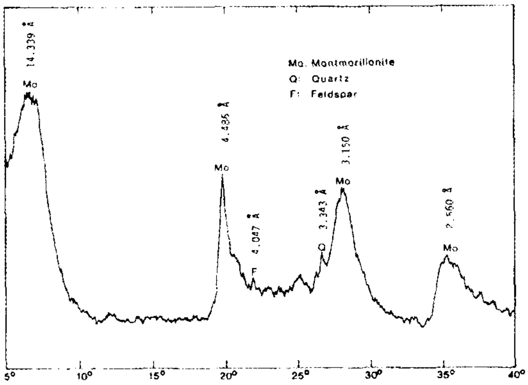

Fig. 4. X-ray Diffraction Pattern of the Montmorillonite Concentrated from Kyungju Bentonite

The XRD patterns indicate that the predominant phyllosilicate present in the Kyungju bentonite is a montmorillonite. To obtain the weight percentage of montmorillonite, the surface area of montmorillonite in the Kyungju bentonite was measured using the following procedures. The bentonite was fully mixed with the distilled water and allowed to settle for a day. The supernatant was moved to the other beaker by pipette, and dried to obtain the clod of montmorillonite. The powder of montmorillonite was obtained by crushing the clod. The XRD patterns of the powder are shown in Fig. 4, and they indicate that the powder is composed mainly of montmorillonite. The surface area of the powder was measured to be 498.0 m^2/g. From the surface areas of Kyungju bentonite and

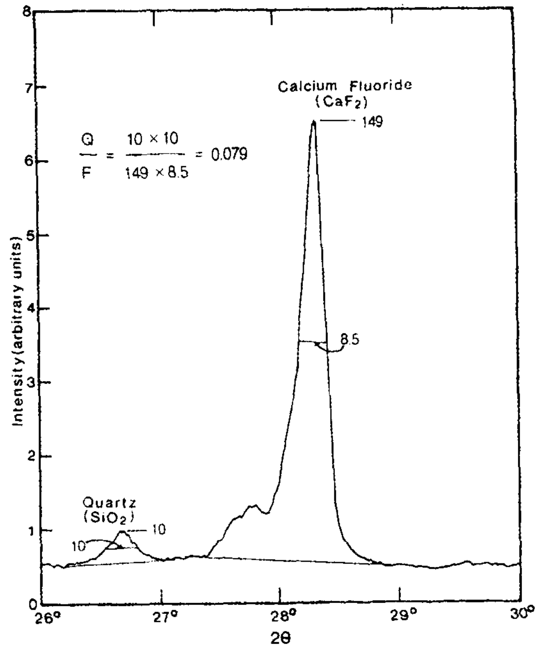

Fig. 5. X-ray Diffraction Patterns of the 3.343 Å Quartz and 3.153 Å Fluorite Lines of a Mixed Sample of the Kyungju Bentonite and Calcium Fluoride Powder Prepared in Proportion of 1 to 0.25 by Weight

montmorillonite, the weight percentage of montmorillonite was obtained to be 70 %. The weight percentage of quartz was obtained from the XRD pattern with the internal standard of CaF_2 and the calibration curve using the quartz-fluoride intensity by Alexander and Klug [19]. The XRD patterns for the 1 : 0.25 bentonite-CaF_2 mixture are shown in Fig. 5. From the figure, the intensity ratio of SiO_2/CaF_2 is 0.079, and the corresponding weight percentage of quartz is about 1 % from the calibration curve. Because the Kyungju bentonite contains montmorillonite, feldspar and quartz, the weight percentage of feldspa was obtained to be 29 %.

3.4. Hydraulic Properties

The hydraulic conductivities were calculated from the slopes of linear relationships in the flow

Table 3. Hydraulic Conductivities of Compacted Bentonite

Dry density (Mg/m^3)	Thickness (cm)	Hydraulic head (kPa)	Hydraulic gradient (m/m)	Hydraulic conductivity (m/s)
1.4	2.5	900	3749	2.67×10^{-12}
1.4	2.5	900	3749	1.95×10^{-12}
1.4	2.5	900	3749	2.05×10^{-12}
1.6	2.5	2000	8240	3.68×10^{-13}
1.6	2.5	2000	8240	4.15×10^{-13}
1.6	2.5	2000	8240	3.67×10^{-13}
1.6	1.0	2000	20600	4.26×10^{-13}
1.6	1.0	2000	20600	2.29×10^{-13}
1.6	1.0	2000	20600	1.59×10^{-13}
1.8	1.0	2000	20600	2.93×10^{-14}
1.8	1.0	2000	20600	2.72×10^{-14}
1.8	1.0	2000	20600	2.35×10^{-14}
1.8	1.0	2000	20600	9.72×10^{-14}
1.8	1.0	2000	20600	$4.80 \times 10-^{14}$
1.8	1.0	2000	20600	4.30×10^{-14}

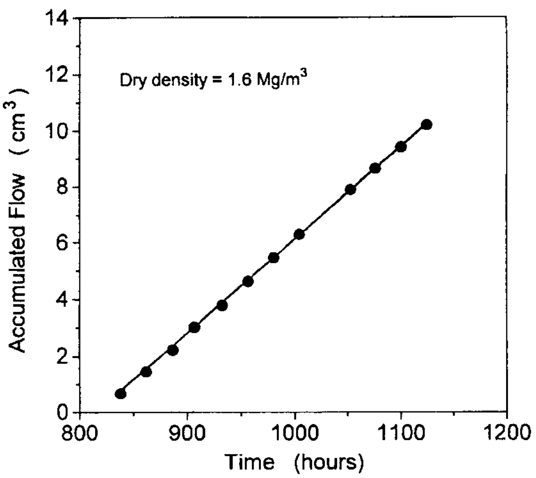

Fig. 6. Typical Flow Rate-time Relationships for the Compacted Bentonite with Dry Density of 1.6 Mg/m^3

characteristics of compacted bentonite. The typical flow characteris is presented in Fig. 6, and the calculated hydraulic conductivities are shown in Table 3. The hydraulic conductivities of domestic bentonite are in the range of 2×10^{-14} to 3×10^{-12} m/s in case of the dry density of 1.4 to 1.8 Mg/m^3. The hydraulic conductivity of MX-80 bentonite with the dry density of 1.7 Mg/m^3 is 1.5×10^{-14} m/s [20], and is somewhat lower than that of domestic bentonite. However the hydraulic conductivities for Avonlea bentonite with the dry density of 1.3 to 1.5 Mg/m^3 are in the range of 1×10^{-13} to 1×10^{-12} m/s [21], and those of Fo-Ca smectite with the dry density of 1.4 to 1.75 Mg/m^3 are in the range of 1×10^{-13} to 1×10^{-12} m/s [22]. These values are similar to those of domestic bentonite. As observed in Table 3, the hydraulic conductivities of all compacted bentonites with the dry densities of 1.4, 1.6, and 1.8 Mg/m^3 are very low and less than 10^{-11} m/s. It is reasoned that the high swelling potential of the bentonite contributes significantly to development of low resultant hydraulic conductivities. The hydraulic conductivity decreases with increasing dry density of bentonite. To investigate the relationship between the hydraulic conductivity and the dry density of bentonite, the logarithm of the hydraulic conductivity versus bentonite dry

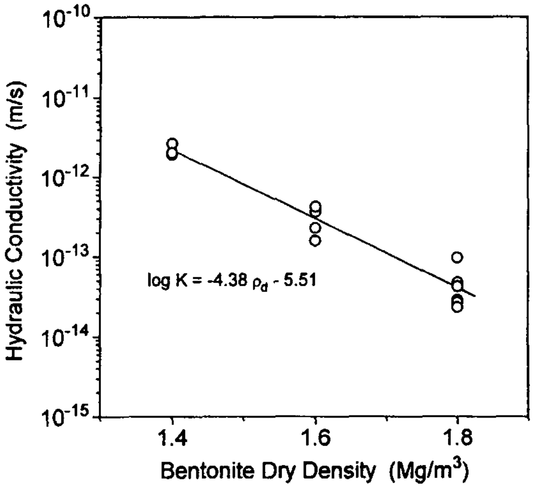

Fig. 7. Hydraulic Conductivity Versus the Dry Density of Compacted Bentonite

density has been plotted and the results are shown in Fig. 7. The relation can be fitted to a straight line, and the expression of this line is:

$$\log K = -4.38\,\rho_d - 5.51 \qquad r^2 = 0.940$$

Gillham and Cherry[23] showed that, if the hydraulic conductivity is less than 10^{-8} m/s when the hydraulic gradient and the porosity are typical values of those of host rock in deep granite, radionuclide migration will be controlled by diffusion. According to the preliminary functional criteria [27] for buffer material, the hydraulic conductivity should be below 1×10^{-12} m/s. The hydraulic conductivity of the compacted bentonite with dry density of 1.6 Mg/m^3 or higher is less than 10^{-12} m/s and therefore may be enough to meet the requirement.

3.5. Swelling Properties

The free swell rate of the bentonite was 89.1 ± 2.8 ml/10g of dry bentonite. Fig. 8 is a typical curve representing the change of swelling pressures of compacted bentonite with water

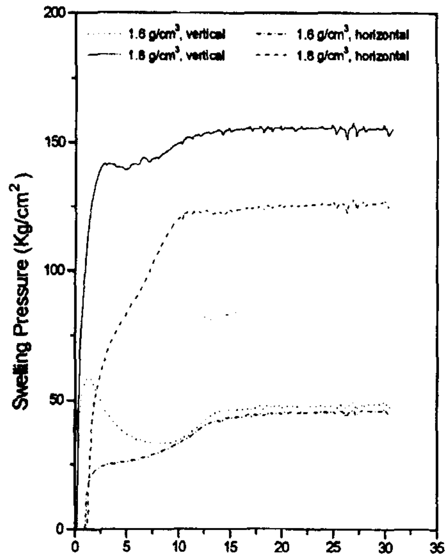

Fig. 8. Change of Swelling Pressure of Compacted Bentonite with Water Contacting Time

contacting time. As shown in the figure, the swelling pressures are developed rapidly for a few days until they reach a peak. Subsequently, they start to decline gradually to a constant value. These tests were ended in 30 days when the time-dependent behavior of the swelling pressure was reached at the steady-state. Both vertical and horizontal swelling pressures showed similar trends in the time-dependent behaviour. The vertical swelling pressures were higher than the horizontal ones, suggesting the anisotropic nature of the compacted bentonite. When the dry densities were 1.4 to 1.8 Mg/m^3, the swelling pressures were in the range of 6.6 kg/cm^2 to 143.5 kg/cm^2 and increased with increasing dry density. The dependence of the swelling pressures upon the dry density was more sensitive in higher dry density. According to the preliminary functional criteria [27] for buffer material, the swelling pressure should be less than 200 kg/cm^2 to avoid the excessive external load on the canister. The swelling pressure of the compacted bentonite with dry density of 1.8 Mg/m^3 or lower is less than

Table 4. Unconfined Compressive Strength, Young' s Modulus and Poison' s Ratio for the Compacted Bentonite

Wet density, (Mg/m^3)	Dry density, (Mg/m^3)	Water content, (%)	Unconfined compressive strength, (kg/cm^2)	Young' s modulus, (kg/cm^2)	Poison' s ratio
2.094	1.733	20.83	93.8	9,875	0.35

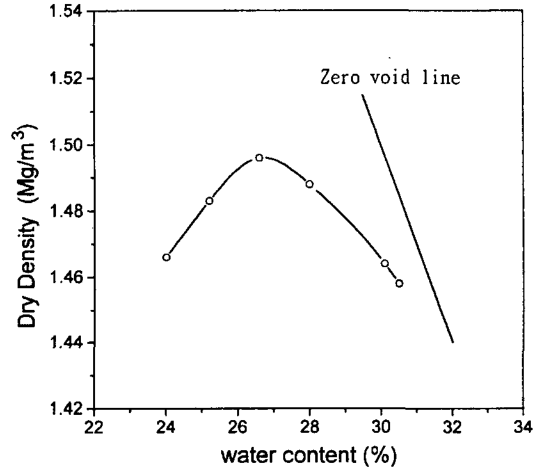

Fig. 9. Moisture-Dry Density Relationship of the Bentonite

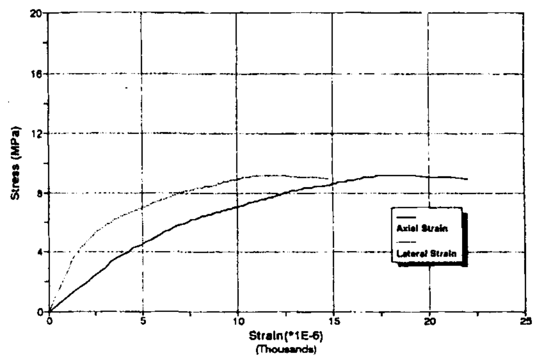

Fig. 10. Stress-strain Relationship of the Bentonite

150 kg/cm^2 and therefore may be enough to meet the requirement. The swelling pressures of MX-80 bentonite and Avonlea bentonite with dry density of 1.75 Mg/m^3 were 130 kg/cm^2 [15] and 160 kg/cm^2 [24], respectively, and those of Fo-Ca smectite with dry density of 1.75 Mg/m^3 were 110 kg/cm^2 [22] and 130 kg/cm^2 [25].

3.6. Mechanical Properties

The dry density-water content relationships are presented in Fig. 9. This compaction curve shows that, as the molding water content is increased, the dry density increases to a maximum value and then decreases. The obtained maximum dry density and the optimum water content are 1.50 Mg/m^3 and 26.6 wt%, respectively. The addition of water increases the amount of free water in the voids. This decreases the intergranular, or interped, shearing resistance and aid compaction. Thus, dry density increases until the system is virtually saturated. The stress-strain relationship for the bentonite specimen is shown in Fig. 10. Young' s module is the tangent modulus corresponding to 40 % of the curve (vertical stress), and Poisson' s ratio is a ratio between the axial strain and the lateral strain. The results are listed in Table 4. The measured Young' s module and unconfined compressive strength are slightly higher than those (6,180 kg/cm^2 and 79.1 kg/cm^2, respectively) for Kunigel V1 Na-bentonite with same density and water content [26]. This may be due to the difference of the type of bentonite. The relationships between void ratio and log pressure for the bentonite are plotted in

Table 5. Consolidation Characteristics of the Compacted Bentonite

Pressure kN/m^2	Settlement mm	Void ratio	Coefficient of compressibility m^2/MN	Coefficient of volume change m^2/MN	Coefficient of consolidation m^2/yr	Compression index	Remarks
200	0.010	0.5658	3.9×10^{-6}	0.00249	0.115		Loading
400	0.046	0.5630	1.4×10^{-5}	0.00889	0.030	0.0056	〃
800	0.142	0.5555	1.9×10^{-5}	0.01212	0.022	0.0151	〃
1600	0.476	0.5293	3.3×10^{-5}	0.02142	0.018	0.0524	〃

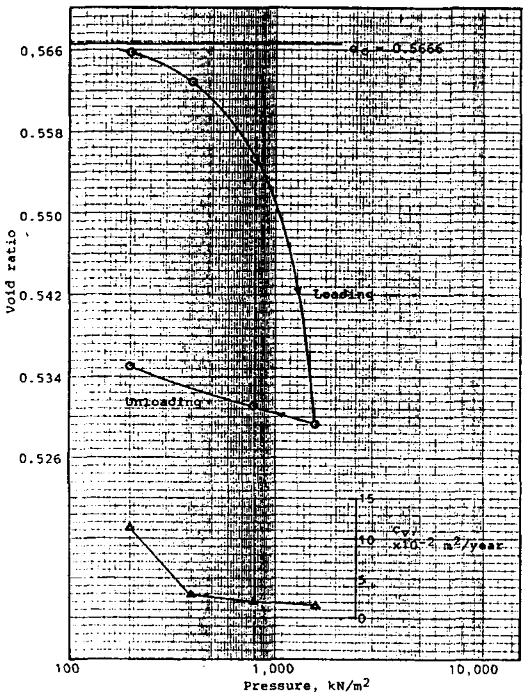

Fig. 11. Voids Ratio Versus Log Pressure Curve for the Bentonite

Fig. 11, and the major consolidation properties obtained from the relationship are presented in Table 5. The coefficient of volume change and the coefficient of consolidation are in the range of 0.00249 to 0.02142 m^2/MN and 0.018 to 0.115 m^2/year, respectively.

4. Summary

The physicochemical, mineralogical, hydraulic, swelling and mechanical properties of a domestic bentonite for use as a buffer material in a high-level waste repository have been measured. The Kyungju bentonite is identified to be a Ca-bentonite containing about 70 wt% of montmorillonite. The hydraulic conductivity of the compacted Ca-bentonite with its dry density higher than 1.4 Mg/m^3 is lower than 10^{-11} m/s. Thus the radionuclides will be migrated by diffusion through the buffer. When the dry densities are 1.4 to 1.8 Mg/m^3, the swelling pressures are in the range of 6.6 kg/cm^2 to 143.5 kg/cm^2. The unconfined compressive strength is about 94 kg/cm^2, and The coefficient of volume change and the coefficient of consolidation are in the range of 0.00249 to 0.02142 m^2/MN and 0.018 to 0.115 m^2/year, respectively. The major properties of Kyungju bentonite are comparable with those of bentonites suggested as a candidate buffer material for high-level waste repository in several foreign countries, and may be enough to meet the requirement of the preliminary functional criteria for buffer material. These results indicate the possibiltity of use of domestic bentonite as a buffer material. However, to determine the suitability of domestic bentonite for the buffer material, the design concept of engineered barriers in the repository should be fixed, because the requirements of mechanical properties depend strongly on the design concept. Also, more experimental works on the mechanical properties such as shear and the thermal properties such as thermal conductivity are also required.

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