SWELLING AND HYDRAULIC PROPERTIES OF Ca-BENTONITE FOR THE BUFFER OF A WASTE REPOSITORY

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Abstract

Swelling and hydraulic tests were carried out to provide the information for the selection of buffer material in a radioactive waste repository. Ca-bentonite and de-ionized water were used for the tests. The swelling pressures of compacted bentonite were in the wide range of 0.7 Kg/cm^2 to 190.2 Kg/cm^2, and they largely increased with an increase in the dry density and bentonite content. However, the swelling pressures decreased with increasing the initial water content and beyond about 12 wt.% of the initial water content, leveled off to a nearly constant value. The hydraulic conductivities were lower than 10^{-11} m/s for the compacted bentonite with the dry density higher than 1.4 Mg/m^3. They increased with increasing temperature in the range of 20^0C to 150^0C.

1. INTRODUCTION

A repository for high-level radioactive wastes would be constructed in the bedrock deep below ground surface. The present design concept [1,2] of underground repository includes the use of compacted clay-based materials for the buffer. The buffer material is required to have high swelling capacity and thus low hydraulic conductivity in order to minimize the penetration of ground water from the host environment. Bentonite has been widely favored as a candidate buffer material, because it has high swelling capacity, high sorptive potential and good durability under disposal environment [3,4].

When a compacted bentonite is used for the buffer, the swelling pressure should be low to avoid excessive load to the container and the surrounding rock although the swelling capacity may be high for its successful sealing role as a barrier. It is reported in the literature [5-7] that the swelling pressure was in the wide range of 0.27 Kg/cm^2 to 590.45 Kg/cm^2 depending upon dry density, bentonite content, initial water content, and so on.

The hydraulic conductivity of compacted bentonite may be affected by the temperature change of the buffer, which occurs due to the decay heat from high-level wastes. Pusch [8] observed an increase in the hydraulic conductivity of compacted bentonite up to one order of magnitude heating to 70^0C. Mingarro et al. [9] reported that the hydraulic conductivity was increased up to about one order of magnitude with the increase of temperature from 20^0C to 100^0C for the mixture of bentonite and crushed granite. Peterson and Kelkar [10] and Ouyang and Daemen [11] also investigated the hydraulic conductivity of pure bentonite and bentonite/crushed rock mixture at a temperature up to 90^oC, but the results did not show a large temperature dependence of hydraulic conductivity. The hydraulic conductivity measured at 90^oC was only about two times higher than that at 20^oC. The effect of temperature on the hydraulic conductivity therefore should be fully understood from the viewpoint of the long-term performance of bentonitic buffer for high-level waste repository.

This study is intended to measure the swelling and hydraulic properties of Ca-bentonite which is considered as a candidate buffer material for a high level waste (HLW) repository in Korea. Based upon the experimental data measured, the effect of dry density, bentonite content, and initial water content on the swelling pressure will be analyzed. The effect of temperature on the hydraulic conductivity will be also investigated for various dry densities.

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2. EXPERIMENTALS

2.1. Materials

The bentonite was taken from Jinmyeong mine which is located in Yangnam Kyungsangbuk-do, Korea. It was used for the tests after being air-dried and passed through No. 200 of ASTM (American Society for Testing and Materials) standard sieves. The bentonite was found to be of Ca-type and its mineralogical composition was 70% montmorillonite, 29% feldspar, and 1.0% quartz [12]. The surface area and the cation exchange capacity were 348 m^2/g and 57.6 meq/100g, respectively. The sand that was used in the bentonite-sand mixture to investigate the effect of bentonite content on the swelling pressure was taken from Daekwang Mine Company, and it included quartz as a major mineral and a small amount of feldspar and mica [12]. De-ionized water was used for the swelling and hydraulic tests.

2.2. Swelling test

The swelling pressures were measured using the following experimental apparatus. The apparatus consists of four sections namely, a main test section, a pressurized water feeding section, a vacuuming section, and a signal and data processing section. The main test section includes a stainless steel cylindrical cell of which the dimension was 5×10^{-2} m in height and 5×10^{-2} m in diameter, porous metal filters to avoid the loss of bentonite particles, and swelling pressure sensors mounted in both vertical and horizontal directions. The pressurized water feeding section consists of a stainless steel reservoir and a highly pressurized helium gas bomb that is used for feeding the water in the reservoir to compacted sample. The stainless steel reservoir is designed to tolerate the pressure of 30 Kg/cm^2. In the vacuuming section, a vacuum pump was 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 the pressure sensor and then to display their values on digital indicator. The swelling pressures were measured according to Box-Behnken's experimental design which had three levels to each of the following three variables; dry density, bentonite content, and initial water content. The left column of Table I shows experimental conditions designed according to the Box-Behnken's scheme for the measurement of the swelling pressures. These swelling tests start with feeding the pressurized water after the zero-point of pressure sensor was adjusted, and the swelling pressures sent from the sensor were collected in a given time interval using personal computer. The effective swelling pressures are obtained by subtracting applied pressures from the collected swelling pressures. These tests proceed until the swelling pressure reaches steady state.

2.3. Hydraulic test

The hydraulic conductivities in water-saturated bentonites with the dry densities of 1.4 Mg/m^3, 1.6 Mg/m^3 and 1.8 Mg/m^3 were measured within the temperature range of 20°C to 150°C. A modified constant head test was applied using constant helium gas pressure [13,14]. The apparatus used 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 5×10^{-2} m. The compacted samples with pre-determined dry density were placed in the sample chamber, and the water flew from the bottom to the top of the sample chamber. The penetrated water volumes were measured by weighing. The hydraulic conductivity was determined when steady state was reached. The measurement was done on the same bentonite specimen at different temperature, that is, the hydraulic conductivity was measured at 20 °C for a sample placed in the chamber and then at temperatures progressively higher.

TABLE I. BOX-BEHNKEN'S EXPERIMENTAL DESIGN AND MEASURED SWELLING PRESSURES.

ID	Box-Behnken Design no.	Experimental conditions			Measured swelling pressures	
		Dry density (Mg/m^3)	Bentonite content (%)	Water content (%)	Vertical (Kg/cm^2)	Horizontal (Kg/cm^2)
1	13	1.6	65	10	5.84	5.53
2	10	1.6	100	6	50.11	33.76
3	3	1.4	100	10	6.62	9.54
4	9	1.6	100	14	36.68	34.68
5	1	1.8	100	10	143.52	114.59
6	12	1.6	30	6	1.16	1.53
7	8	1.4	65	6	2.81	4.16
8	6	1.8	65	6	21.23	20.24
9	4	1.4	30	10	0.96	0.56
10	11	1.6	30	14	1.10	0.98
11	7	1.4	65	14	1.39	2.12
12	15	1.6	65	10	5.84	5.53
13	2	1.8	30	10	1.36	2.88
14	5	1.8	65	14	27.16	23.06
15	14	1.6	65	10	5.84	5.53

3. RESULTS AND DISCUSSION

3.1. Swelling pressures of compacted bentonite

Fig. 1 is an example representing the change of swelling pressures as a function of elapsed time. As shown in the figure, the swelling pressures develop rapidly until they reach a peak in a few days.

Subsequently, they start to decline gradually to constant values. These tests ended in 30 days, because the time-dependent behavior of the swelling pressure was regarded as reaching steady state. The swelling pressures measured are listed in the right column of Table I together with experimental conditions. Both vertical and horizontal swelling pressures showed similar trends in the time-dependent behavior. The vertical swelling pressures were higher than the horizontal ones, which suggests the anisotropic nature of compacted sample. Based upon the measured experimental data, the parametric modeling of swelling pressure was made to investigate the relationship between the swelling pressure and three variables which expressed which was expressed in a polynomial in a polynomial. The following quadratic equation is used to approximate the experimental data [15].

$$
\begin{aligned}
P_s &= f\,(x_1, x_2, x_3) \\
&= b_o + \sum_{i=0}^{3} b_i x_i + \sum_{i=0}^{3} b_{ii} x_{ii}^{\;2} + \sum_{i=0}^{3} b_{ij} x_i x_j \\
&= b_o + b_1 x_1 + b_2 x_2 + b_3 x_3 + b_{11} x_1^{\;2} + b_{22} x_2^{\;2} + b_{33} x_3^{\;3} \\
&\qquad + b_{12} x_1 x_2 + b_{13} x_1 x_3 + b_{23} x_2 x_3
\end{aligned}
\tag{1}
$$

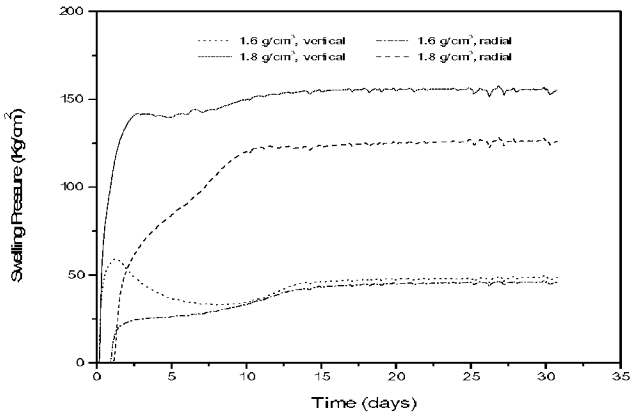


Fig. 1. Development of swelling pressure versus elapsed time for bentonite.

where P_s is the swelling pressure, x_1 the dry density, x_2 the bentonite content, x_3 the initial water content, and b the coefficient of the quadratic equation. The coefficients of the quadratic equation were determined through multiple regression analysis of experimental data, in which the swelling pressures were employed as log P_s to minimize the deviation in the calculation. The multiple regression analysis was conducted using LOTUS 123 program. The polynomial equation determined was given:

$$
\begin{aligned}
\log P_s &= 0.766 + 0.457x_1 + 0.753x_2 - 0.045x_3 + 0.001x_1^{\;2} + 0.006x_2^{\;2} + 0.070x_3^{\;2} \\
&\qquad + 0.296x_1x_2 + 0.103x_1x_3 - 0.028x_2x_3
\end{aligned}
\tag{2}
$$

Correlation coefficient, $r^2 = 0.99$

This equation explains that the dry density and bentonite content give positive and large contribution to the swelling pressure, while the initial water content gives negative and small one.

The effect of dry density, bentonite content, and initial water content on the swelling pressure was analyzed using the polynomial equation (2). Fig. 2 is the swelling pressure versus dry density for various bentonite contents. The swelling pressure was in the range of 0.7 Kg/cm^2 to 190.2 Kg/cm^2 and it increased with increasing the dry density. The dependence of the swelling pressure upon the dry density was more sensitive in higher dry density. This figure also shows that the swelling pressure increases with increasing the bentonite content, which is at a slow gradient up to about 50 wt% of bentonite content. Above 50 wt%, there is a rapid increase in the swelling pressure. Such a behavior becomes distinct at higher dry density. It is suggested for the mixture of bentonite and sand that the swelling pressure is controlled largely by the bentonite content

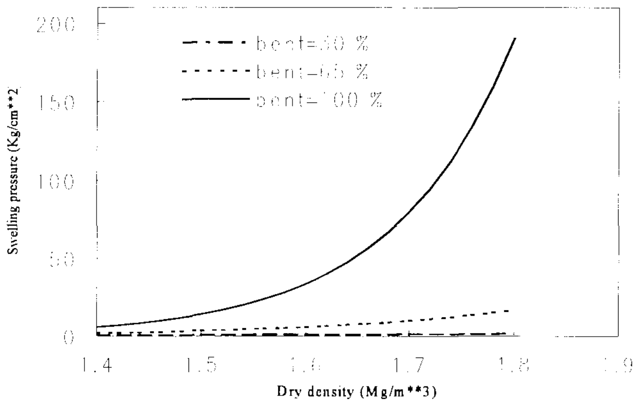


Fig. 2. Effect of dry density and bentonite content on the swelling pressure.

Using the concept of effective bentonite dry density [16], an attempt was made to explain the relationship between swelling pressure and dry density for the compacted samples with various bentonite content. The effective bentonite dry density, ρ_{eff} is defined as the ratio of the mass of bentonite to the volume of bentonite plus void in the mixture :

$$\rho_{eff} = \frac{mass\ of\ bentonite}{volume\ of\ bentonite\ plus\ void\ in\ the\ mixture} = \frac{(1-\omega_s)\,\rho_{md}}{1-\omega_s(\rho_{md}/\rho_s)} \qquad (3)$$

where ω_s is the weight fraction of sand in the mixture, ρ_{md} the dry density of the mixture, and ρ_s the true density of sand. The effective bentonite dry densities that correspond to the dry densities in the Fig. 2 are calculated using the equation (3). The swelling pressures versus the effective bentonite dry densities are plotted in Fig. 3. As shown in the figure, the relationship between swelling pressure and dry density can be explained by the concept of effective clay dry density. This suggests that the swelling pressure for the mixture of bentonite and sand is dominated by the bentonite. The sand acts merely as an inert filler material and serves only to limit the free volume into which the bentonite can be compacted. Therefore, when the effective bentonite dry density of mixture is similar to the dry density of pure bentonite, then the swelling pressure is close to that for the pure bentonite.

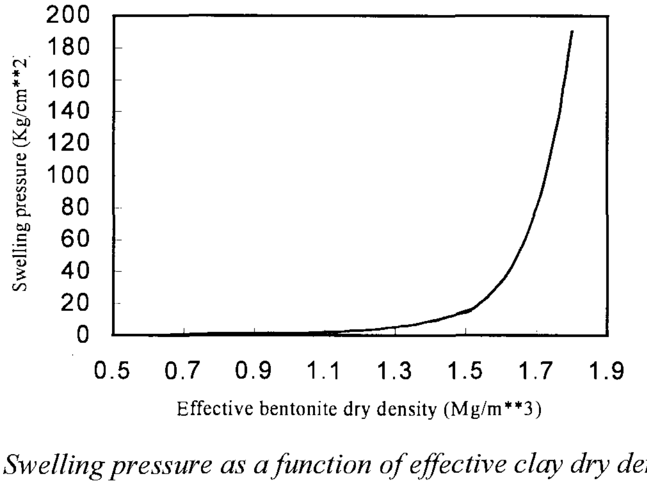
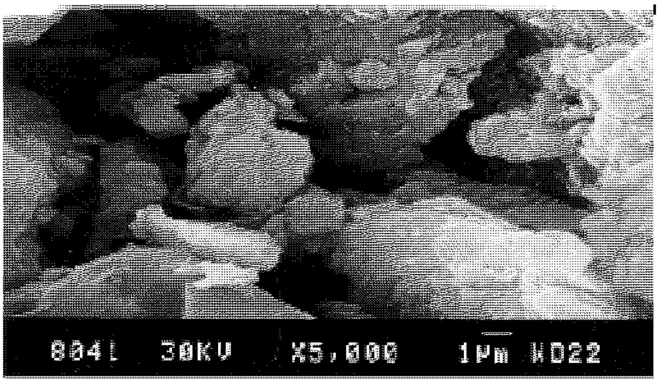
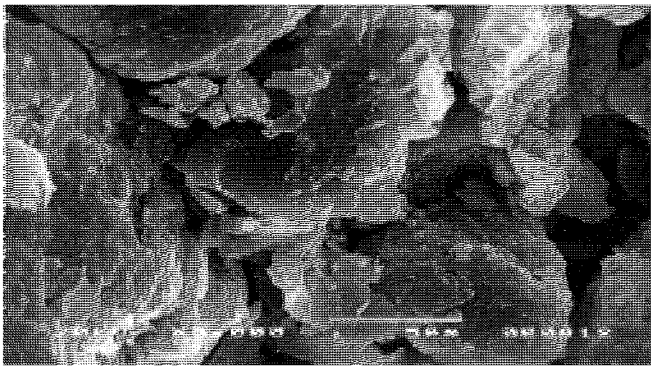


Fig.3. Swelling pressure as a function of effective clay dry density.



(a) dry density = 1.6 Mg/m³



(b) dry density = 1.8 Mg/m³

Fig.4. Results of SEM analysis of the compacted bentonite with the dry densities of 1.6 Mg/m³ and 1.8 Mg/m³.

Fig. 3 also shows that there is a slow increase of the swelling pressure up to a threshold dry density between 1.6 Mg/m^3 and 1.8 Mg/m^3. Above the threshold dry density, the swelling pressure rises at a much steeper gradient. Such a behavior of the swelling pressure was also reported in Gray et al.'s experiment with Avonseal bentonite-sand mixture[17], in which the threshold dry density was found to be between 1.6 Mg/m^3 and 1.7 Mg/m^3. The steep increase of the swelling pressure beyond the threshold dry density is supposed to be due to the anisotropic nature of the compacted bentonite. The SEM analysis may offer an explanation for the anisotropy. The typical micrographs of the compacted bentonite, when compacted to 1.6 Mg/m^3 and to 1.8 Mg/m^3, are presented bentonite, when compacted to 1.6 Mg/m^3 and to 1.8 Mg/m^3, are presented in Fig. 4(a) and 4(b), respectively. Fig. 4(a) shows that the mixture at low effective dry density consists of randomly arranged aggregates of bentonite particles (micropeds), while Fig. 4(b) shows that, at the dry densities above the threshold value, interparticle voids are mostly eliminated, the micropeds become fused, and the topography is relatively uniform. The results of SEM analysis suggest that the micropeds of bentonite particles beyond the threshold dry density are arranged in a nearly parallel orientation and thus the corresponding swelling pressures increase steeply. On the other hand, Pusch put out for this phenomenon in his report [18] the explanation that the force of hydration on the surface of particles or their interlamellar layers may be of significance in the swelling pressure of highly compacted bentonite with the densities of 2.0–2.1 Mg/m^3.

Fig. 5 represents the effect of initial water content on the swelling pressure of bentonite with the dry density of 1.6 Mg/m^3. The swelling pressure, which is in the range of from 32.1 Kg/cm^2 to 46.5 Kg/cm^2, decreases with increasing initial water content. From about 12 wt% of initial water content, however, there is little change in the swelling pressure. This is expected to be because the bentonite contacting with the initial water is partially swelled prior to swelling pressure test. Therefore, the lower the initial water content is, the higher is the swelling pressure.

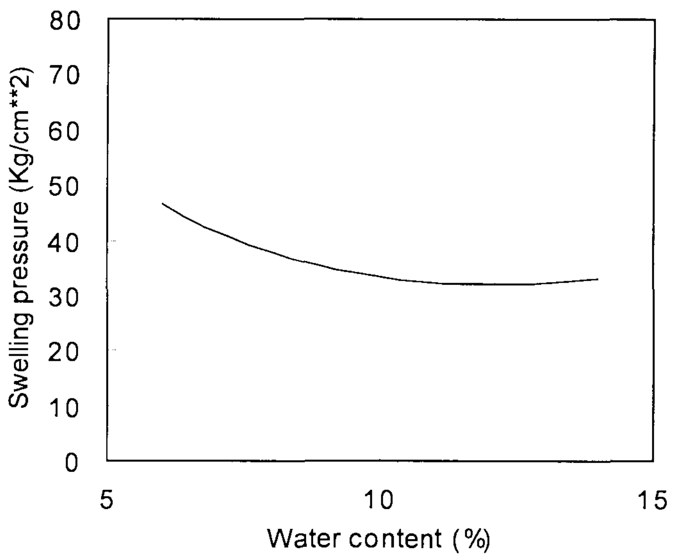


Fig.5. Effect of initial water content on the swelling pressure for bentonite (dry density=1.6 Mg/m³).

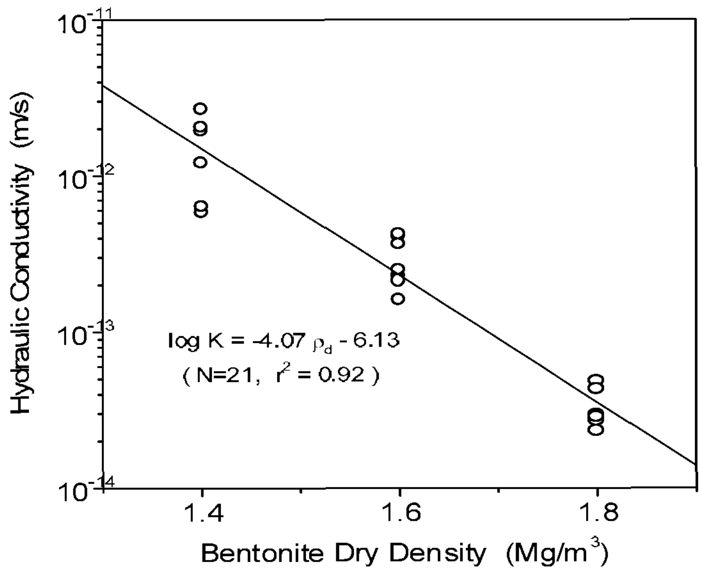


Fig.6. Hydraulic conductivity versus the dry density of compacted bentonite.

3.2. Hydraulic conductivities of compacted bentonite

The hydraulic conductivities were calculated from the slopes of linear relationships in the flow characteristics of compacted bentonite. The hydraulic conductivities of domestic bentonite at 20°C 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/m³. The hydraulic conductivity of MX-80 bentonite with the dry density of 1.7 Mg/m³ is 1.5×10^{-14} m/s [19], 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/m³ are in the range of 1×10^{-13} to 1×10^{-12} m/s [14], and those of Ca-smectite with the dry density of 1.4 to 1.75 Mg/m³ are in the range of 1×10^{-13} to 1×10^{-12} m/s [20]. These values are similar to those of domestic bentonite. The hydraulic conductivities of all compacted bentonites with the dry densities of 1.4, 1.6, and 1.8 Mg/m³ are very low and less than 10^{-11} m/sec. 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

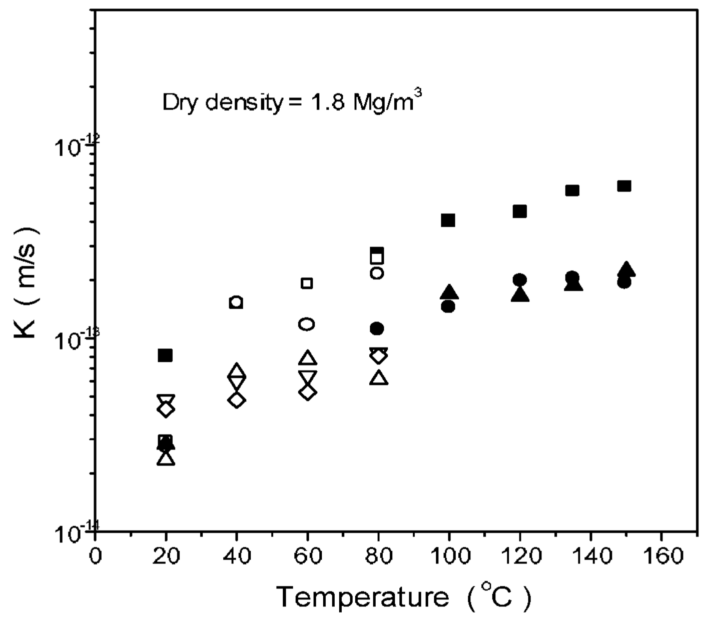


Fig. 7. Influence of temperature elevation on the hydraulic conductivity of compacted bentonite.

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 density has been plotted and the results are shown in Fig. 6. The relation can be fitted to a straight line, and the expression of this line is:

$$\log K = -4.07\,\rho_d - 6.13 \qquad r^2 = 0.92 \qquad (4)$$

Gillham and Cherry[21] 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. The hydraulic conductivities of compacted bentonite increased with increasing temperature. The hydraulic conductivities for bentonites with dry densities of 1.4 Mg/m^3 to 1.8 Mg/m^3 at the temperature of 150°C increased up to about one order higher than those at 20°C. The hydraulic conductivities for bentonite with the dry density of 1.8 Mg/m^3 as a function of temperature are presented in Fig. 7. The change of hydraulic conductivities in the bentonites at elevated temperatures is attributable to the change of the permeability factor, the viscosity factor and the density factor, and these three factors are competitive with one another. Of the three factors, the change in viscosity is most sensitive to the increase in temperature, and contributes greatly to the increase of hydraulic conductivity with increasing temperature [22].

4. CONCLUSIONS

The swelling pressures are in the range of 0.7 Kg/cm^2 to 190.2 Kg/cm^2 under given experimental conditions. The swelling pressures increase with an increase in the dry density, and they also increase with increasing the bentonite content. On the contrary, they decrease with initial water content and, beyond about 12wt% of initial water content, leveled off to a nearly constant value. The relationship of swelling pressure versus dry density for various bentonite contents can be explained by the concept of effective bentonite dry density. The hydraulic conductivities of the compacted domestic Ca-bentonite with its dry density higher than 1.4 g/cm^3 are lower than 10^{-11} m/s, which may be enough to meet the requirement of the preliminary functional criteria for buffer material. The temperature increase in compacted bentonite increases hydraulic conductivity, which is most sensitive to the change in viscosity.

The experimental results obtained will be useful to the selection of a candidate buffer material for a high-level waste repository in Korea.

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