EFFECT OF CHEMICAL SOLUTIONS ON HYDRAULIC BARRIER PERFORMANCE OF CLAY GEOSYNTHETIC BARRIERS

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ABSTRACT: This paper discusses the experimental results on chemical compatibility of clay geosynthetic barriers (CGBs), which are the hydraulic barrier performances of (1) non-prehydrated CGB against multi species chemical solutions, (2) prehydrated CGB against chemical solutions, and (3) chemical-resistant modified bentonite against chemical solutions. The first project, in which $CaCl_2$ and NaCl solutions were used on needle-punched CGB encapsulating powdered bentonites, found that the free swell of bentonite and hydraulic conductivity (*k*) of CGB was significantly affected by the ionic strength, and powdered bentonites are more compatible against chemical solutions than granular bentonites. The second project was conducted since prehydration prior to permeation is expected to provide low *k*. The CGB samples prehydrated in the laboratory were subjected to hydraulic conductivity tests using $CaCl_2$ solutions. The results show that prehydrated CGBs provided only 0-1 order of magnitude lower *k* than non-prehydrated CGBs for $CaCl_2$ solutions, which is probably attributed to the the non-uniform prehydration over the entire area. The last project was arisen from the needs to improve the chemical compatibility of bentonites. Multiswellable bentonite (MSB) was used in the experiment. MSB exhibited higher swelling and lower *k* values for inorganic chemical solutions, and expected to be an excellent alternative barrier material for landfill liner systems.

1 INTRODUCTION

Clay liners that are used as bottom liners in waste landfill facilities to contain the waste leachate generated from the disposed wastes are required to have low hydraulic conductivity and to prevent the groundwater contamination. Clay geosynthetic barriers (CGBs), also referred as to geosynthetic clay liners (GCLs), have been considered barrier materials alternate to or combined with compacted clay liners. CGBs are the factory-manufactured prefabricated barrier material, either bentonite granules sandwiched between two geotextiles (either woven or nonwoven) or bentonite glued to geomembrane. CGBs are expected to be utilized as landfill liner material because bentonite provides extremely low hydraulic conductivity and high containment capacity due to its high swelling capacity. Advantages of CGBs are very thin (usually 5-10 mm thick), and to need less construction quality control. Hydraulic conductivity values of CGBs permeated with pure water have been reported approximately $1x10^{-9}$ cm/s. However, to evaluate the hydraulic performance of CGBs, landfill operators, engineers, and regulators are required to be knowledgeable about clay-chemical interactions, and to choose appropriate methods in evaluating their hydraulic performance, because it is well known that bentonite does not swell against inorganic solutions or nonpolar liquids (Norrish and Quirk 1954). When bentonite is used as a liner material, its chemical compatibility should be investigated according to the given conditions in the field.

This paper presents the experimental results on chemical compatibility of CGBs, which are (1) the hydraulic barrier performance of non-prehydrated CGB against multi species inorganic chemical solutions, (2) the performance of prehydrated CGB against inorganic chemical solutions, and (3) the performance of chemical-resistant modified bentonite against inorganic chemical solutions. The first project was conducted because many previous researches on chemical compatibility of CGBs had been conducted with single-species chemical solutions or waste leachate and little research had been performed with multi-species chemical solutions. Calcium chloride ($CaCl_2$) and sodium chloride (NaCl) solutions having a different *RMD* (ratio of monovalent to divalent) and *I* (ionic strength) values were used for the free swell and hydraulic conductivity tests on needle-punch type CGB encapsulating powdered bentonites. The second project was conducted because prehydration prior to permeation has been known to provide the comparatively low hydraulic conductivity, but only the limited data have been reported on effects of prehydration and its water content on the hydraulic conductivity as well as on effects of base soil conditions on prehydration. In the experiment, after certain period of prehydratoin where the CGBs were placed on the decomposed granite soil compacted at optimum water content simulating base soil, the prehydrated CGB samples were subjected to hydraulic conductivity tests using $CaCl_2$ solutions having different concentrations as permeants with flexible-wall permeameter. The last project was arisen from the needs to improve the chemical compatibility of bentonites based on the above two projects. Multiswellable bentonite (MSB) is bentonite which has been treated with propylene carbonate (PC) to activate the osmotic swelling of bentonite even in chemical solutions, where the natural bentonite does not swell.

2 BENTONITE-CHEMICAL INTERACTIONS

Smectite group minerals, which are the main component in bentonite, exhibit a high swelling capacity and can provide an extremely low level of hydraulic conductivity against pure water. Many researchers have reported that, even though the hydraulic conductivity is low when the clays are permeated with pure water, permeation with chemical solutions will result in an increase (sometimes significant) in the hydraulic conductivity. These effects were summarized by Mitchell and Madsen (1987), Shackelford (1994), Shackelford et al. (2000), Egloffstein (2001), and Kamon and Katsumi (2001), and categorized into (1) the dissolution of clay particles and chemical compounds, (2) the development of a diffuse double layer (DDL), and (3) the osmotic swelling of smectite clay. The dissolution of soil components results from strong acids and bases. Strong acids promote the dissolution of carbonates, iron oxides, and the alumina octahedral layers of clay minerals. Bases promote the dissolving of the silica tetrahedral layers. These effects can result in an increase in the hydraulic conductivity, although the reprecipitation of dissolved compounds might clog the pores and decrease the hydraulic conductivity (Mitchell and Madsen 1987).

Since the water molecules in a diffuse double layer (DDL) are strongly attracted, they do not contribute to the water flow, and only molecules outside of the DDL can form the flow channel. Thus, the more DDLs develop, the lower the hydraulic conductivity becomes. If the permeant contains cations, the negative charge will attract these cations and fewer water molecules. Therefore, the existence of cations results in fewer DDLs being developed. Polyvalent cations promote the development of fewer DDLs rather than monovalent cations. Higher cation concentrations will also result in the development of fewer DDLs.

The development of a DDLs is also affected by the polarity and the dielectric constant of pore fluids. Mesri and Olson (1971) conducted a series of consolidation tests on kaolinite, illite, and smectite, with several fluids, to investigate the effect of the volume and the shape of voids and the nature of fluids on the hydraulic conductivity. They showed that, when these clays are consolidated with nonpolar fluids (benzen and carbon tetrachloride) instead of water, the hydraulic conductivity is simply dependent on the void ratio regardless of the type of clay. Since these clays differ in particle size, namely, largest for kaolinite and smallest for smectite, the shapes of the flow channels should also be different from clay to clay. Nevertheless, all three types of clay yield the same void ratio versus hydraulic conductivity relationship as the nonpolar fluids. This is because nonpolar fluids do not form a DDL. Mesri and Olson (1971) also showed that the hydraulic conductivity is highest for nonpolar fluids, smaller for polar fluids of low dielectric constant (ethyl alcohol and methyl alcohol), and lowest for water, which is polar and has a high dielectric constant (~80). These results can be explained by the DDL theory.

The permeation of pure organic liquids is not likely to occur in practice when clay liners are applied. Several researchers have reported that no significant increase in hydraulic conductivity occurred when the concentration of organic chemicals was lower than 50% (e.g., Bowders and Daniel 1987, Petrov et al. 1997). This is because dilution with water leads to enough of an increase in the dielectric constant to develop a DDL.

Osmotic swelling is an important phenomenon for smectite clay. When dry smectite is hydrated, water molecules are strongly attracted to the internal and the external clay surfaces during the hydration phase. After this hydration process, if the exchange cations of the smectite clay are monovalent, the region of the interlayer may retain numerous layers of water molecules during the osmotic phase (Norrish and Quirk 1954). This condition can be observed as a significant amount of swelling; it is typically observed when Na-bentonites are hydrated with deionized water. When the smectite is monovalent at the exchange site and is permeated with the solution containing a low concentration of monovalent cations, a large fraction of water is attracted to the clays, less mobile water is available for the water flow, and the hydraulic conductivity is low. When polyvalent cations exist at the exchange sites, the osmotic phase does not occur and less swelling is observed.

3 NONPREHYDRATED (DRY) CGBS WITH INORGANIC SOLUTIONS

Several research projects on the hydraulic conductivity of non-prehydrated CGBs with chemical solutions/liquids were conducted (e.g., Ruhl and Daniel 1997, Petrov and Rowe 1997, Shackelford et al. 2000, Jo et al. 2001, Egloffstein et al. 2002, Shan and Lai 2002). However, most of these researches had been conducted with single-species chemical solutions or waste leachate and little research had been performed with multi-species chemical solutions. In this study, calcium chloride ($CaCl_2$) and sodium chloride (NaCl) solutions having a different *RMD* (ratio of monovalent to divalent) and *I* (ionic strength) values were used for the free swell and hydraulic conductivity tests on needle-punch type CGB. $RMD = c_1/(2\ c_2)^{0.5}$ and $I = 0.5\ \Sigma\ c_i z_i^2$, where where c_i is the concentration of *i* th ion, z_i is the valence of *i* th ion, c_1 is the concentration of monovalent cation, and c_2 is the concentration of divalent cation. In this study, only the cations are considered to calculate the ionic strengths.

The CGBs used in this study contain natural sodium bentonite (Na-Bentonite) encapsulated between a slit-film monofilament woven geotextile and a staple-fiber nonwoven geotextile. The bentonites in CGB used are powdered, while those in CGB that have been reported in many previous studies are granular. The geotextiles are bonded by needle-punched fibers that have been thermally fused to the woven geotextile. The initial thickness of the CGB raged from 5.5 to 6.5 mm and the average initial gravimetric water content of the bentonite was 9 to 11%. The

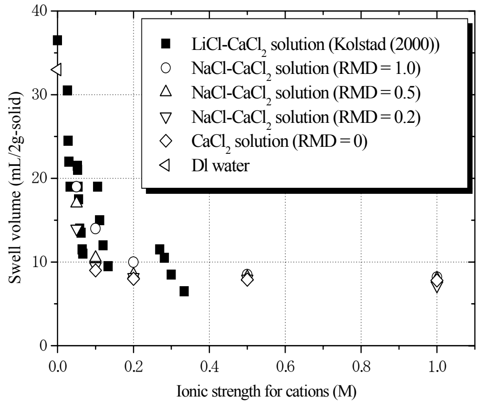


Figure 1 Comparison of swell volume of granular versus powder bentonites in CGBs (data of granular bentonites from Kolstad (2000))

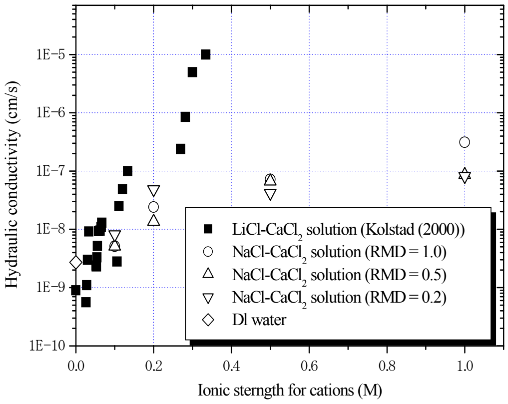


Figure 2 Comparison of hydraulic conductivity of granular versus powder bentonite CGBs (data of granular bentonites from Kolstad (2000))

CGB has a mass of bentonite per area of approximately 5.0 kg/m^2.

Hydraulic conductivity tests were performed using a flexible-wall permeameter with a cell pressure of 20 kPa and a hydraulic gradient of approximately 100.

The difference between granular versus powder was evaluated using the data by Kolstad (2000) on a CGB with granular bentonites, permeated with LiCl-$CaCl_2$ solutions. According to the results of the swell volume shown in Figure 1, the two (powdered and ground) types of bentonite have almost the same sensitivity against chemicals (the swell volume is measured using ground bentonite). Figure 2 illustrates the ionic strength of the cations of the permeants versus the hydraulic conductivity values. The figure indicates that both CGBs are affected by the chemical solutions, but that the powder bentonite may be more compatible than the granular bentonite, particularly with chemically strong (high-concentration) solutions. This is probably because the pores between the granules are not blocked due to a lower level of swelling of the bentonite, especially for aggressive chemical solutions. This results in a significant increase in hydraulic conductivity. In contrast, the pores of the powdered bentonite are small even when the swelling is limited, and result in the lower hydraulic conductivity.

4 EFFECTS OF PREHYDRATION

4.1 *Background*

The water content of the underlain base soil affects the increases in the water content of the CGBs. CGBs underlain by a base soil, with a water content of –4% to +4% of an optimum water content, exhibit a water content ranging from 40 - 100% (Bonaparte et al. 1996). Thus, the chemical compatibility of prehydrated CGBs is also a necessary issue to be addressed.

Values for the hydraulic conductivity, when permeated with sequential liquids, are dominantly affected by the first wetting liquid, and prehydration prior to permeation is known to provide the comparatively low hydraulic conductivity as shown in Figure 3. This is referred to as the “First Exposure Effect” (Shackleford 1994). Prehydration (permeation with water prior to the chemical solutions) versus nonprehydration (direct permeation of the chemical solutions without water permeation) conditions were compared in Figure 4. The figure replots the data obtained by Petrov et al. (1997) and Ruhl and Daniel (1997). The permeant liquids were NaCl, HCl, and NaOH solutions and waste leachate. The levels of ionic strength for these permeants are plotted along the x-axis, while the levels of hydraulic conductivity with the permeant relative to the one permeated with the water are plotted along the y-axis. The non-prehydrated condition has a more significant effect on the hydraulic conductivity and is 1-3 orders of magnitude greater in hydraulic conductivity than the prehydrated condition.

Vasko et al. (2001) conducted hydraulic conductivity tests on CGBs with several different levels of initial (prehydration) water contents permeated with different concentrations of $CaCl_2$ solutions. If the concentration of $CaCl_2$ is 0.025 mol/L, the hydraulic conductivity yields around 1x10^{-7} cm/s regardless of the prehydration water content of the CGB as shown in Figure 5. If the $CaCl_2$ concentration is higher than 0.1 mol/L, the prehydration water content influences the hydraulic conductivity. An increase in the initial water content, from 9% to 200%, results in a decrease in the hydraulic conductivity of two orders of magnitude. However, even a prehydration water content of 200% results in a hydraulic conductivity two orders of magnitude larger than the water permeation, and does not provide hydraulic conductivity lower than 1x10^{-7} cm/s. Daniel et al. (1993) conducted similar experiments in which CGBs with different levels of prehydration water content were permeated with five different organic liquids including benzene and trichloroethylene (TCE). They showed that, if the prehydration water content is greater than 100%, the hydraulic conductivity could be lower than 1x10^{-7} cm/s (and lower than 1x10^{-8} cm/s for benzene and gasoline). The difference between the results of Daniel et al. (1993) and those of Vasko et al. (2001) might be attributed to the type of bentonite and the nature of the liquids employed. Daniel et

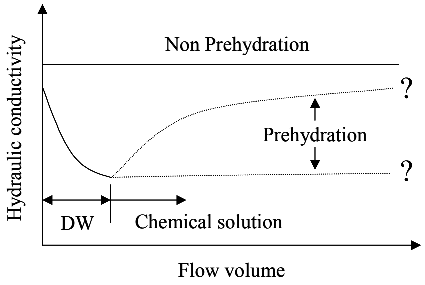


Figure 3 Difference in the hydraulic performance between non-prehydration and prehydration conditions

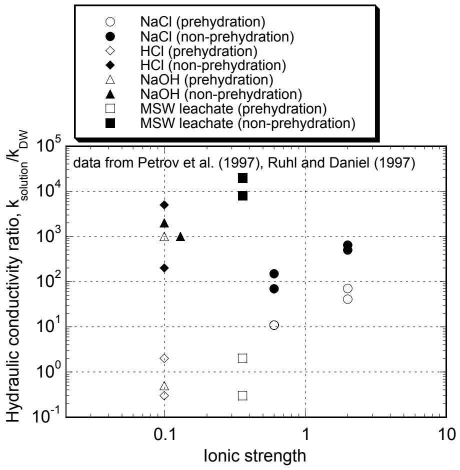


Figure 4 Hydraulic conductivity of CGBs permeated with chemical solutions ($k_{solution}$) relative to hydraulic conductivity with deionized water (k_{DW}) and the ionic strength of the chemical solutions (Katsumi and Kamon 2002)

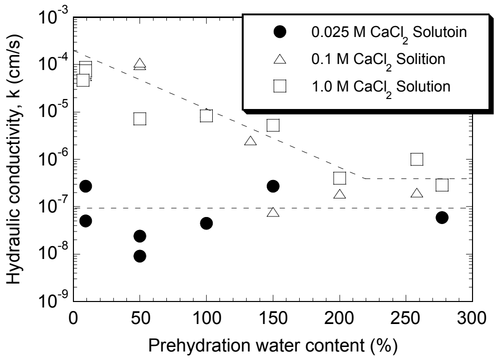


Figure 5 Hydraulic conductivity of CGBs permeated with $CaCl_2$ solutions and prehydration water content (Vasko et al. 2001)

al. (1993) used an organic liquid, which could encounter difficulty when penetrating the pore water, due to its low solubility to water, while the inorganic chemical solutions used by Vasko et al. (2001) are thought to be able to penetrate the pore water with relative ease.

In the experiments presented by Daniel et al. (1993) and Vasko et al. (2001), CGBs were prehydrated with pure water instead of pore water in the actual soil. Also, the experiments by Vasko et al. (2001) put the non-woven textile of CGB at the bottom in order to obtain the uniform prehydration water content over the entire CGB area. There was no reported data on hydraulic conductivity of CGBs that were prehydrated with actual base soil.

4.2 *Experiments*

For the prehydration test, the same CGBs were used as those used in the experiment for non-prehydration conditions presented in the previous chapter. Decomposed granite soil obtained from Otsu-city, Shiga, Japan was used for the base soil. The optimum water content of the decomposed granite soil was 15%. Decomposed granite soil was compacted at 15% or 20% water content. The compacted decomposed granite specimen was put in the mould having a 15 cm height and 10 cm diameter. CGBs cut as a circle shape of 10 cm diameter was placed on the compacted decomposed granite soil. CGB was place with a woven geotextile as a bottom and a non-woven geotextile as a top. This is not consistent with the prehydration test presented by Vasko et al. (2001), who put CGBs with the non-woven geotextile as a bottom to try to obtain the uniform prehydration over the entire area although the authors tried to simulate the actual case in this study. Then, pedestal cap was placed on the CGB and a 5-kPa vertical pressure was applied. The mould containing the base soil and the CGB was put in a container in the constant temperature room (20 degree in Celsius). In the container, pure water was put to supply water continuously in some cases, and not in others. Schematic view of the prehydration tests is shown in Figure 6.

After a certain period of the prehydration, CGBs were subjected to the measurement of water content or hydraulic conductivity values. For the hydraulic conductivity test, CGBs were cut into 6 cm diameter. Flexible-wall permeameter was used. A hydraulic gradient was 80-90 and cell pressure was 30 kPa. The permeants were the calcium chloride solutions having a concentration of 0.2, 0.5, and 1.0 mol/L.

Hydraulic conductivity values obtained are plotted in Figure 7 with the prehydration water content. The results of similar experiments conducted by Vasko et al. (2001) concluded that the increase of prehydration water content may result in the decrease of hydraulic conductivity. However, the experimental results presented in this study do not show the clear tendency as shown by Vasko et al. (2001). This is considered because the prehydration water content obtained in this study was not uniform although the prehydration conducted by Vasko et al. (2001) was tried to be uniform. From the practical viewpoint, woven geotextile side of CGB is put as a bottom and is contacted with the base soil. Therefore, the experimental condition of this study might simulate the actual field rather than the condition presented by Vasko et al. (2001).

Figure 8 illustrates the water content values at different sampled locations of CGBs after prehydration test. The prehydration water content varied from location to location, and was 120% at minimum to 175% at maximum. Vasko et al. (2001) conducted the prehydration test placing the CGB on wet filter paper to induce the water by suction, and when the non-woven geotextile of CGB was put at the bottom to be contacted with the filter paper, the water content increased uniformly over the area. In contrast, when the woven geotextle was contacted with the filter paper, non-uniform prehydration water content was obtained. This is probably because non-woven geotextile attracts the water and then send the water to the soil (bentonite) very homogeneously due to their microscopic structure. Woven geotextile does not attract water because it consists of very thin fibers, and water easily migrates through the holes between the fibers, therefore uniformity of water movement

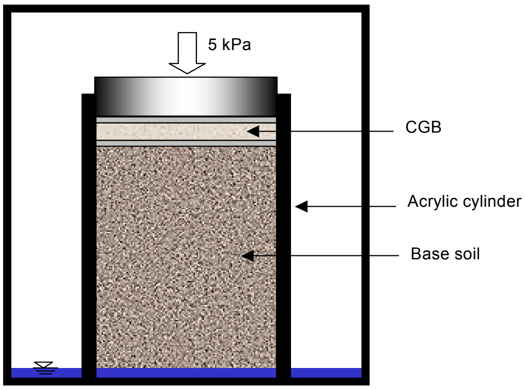


Figure 6 Prehydration test equipment

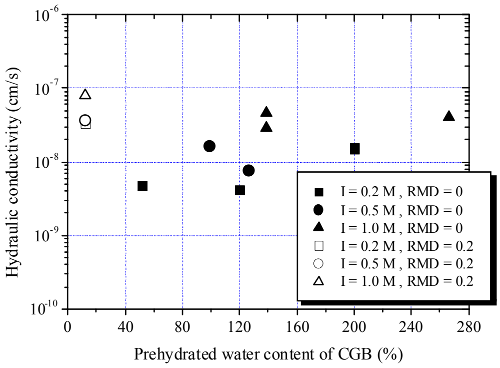
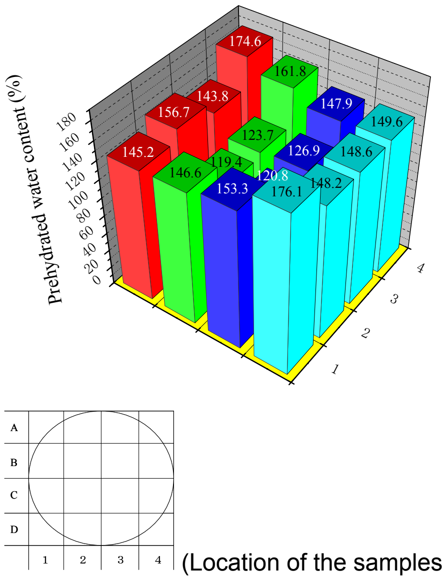


Figure 7 Hydraulic conductivity values versus prehydrated water content (data for non-prehydration from Katsumi et al. (2002))



(Location of the samples)

Figure 8 Water content distribution of CGB from prehydration test (for 7 days with 20% base soil water content) through the geotextile is not assured.

From Figure 7, the prehydration might provide only 0-1 order of magnitude lower hydraulic conductivity than the non-prehydration conditions. The increase in prehydration water content does not necessarily decrease the hydraulic conductivity such as the data by Vasko et al. (2001). Therefore, it can be concluded that prehydration does not effectively contribute to maintaining low hydraulic conductivity. Another important point obtained from these results is the magnitude of the hydraulic conductivity values. The hydraulic conductivity values of prehydrated CGBs ranged from $4x10^{-9}$ to $6x10^{-8}$ cm/s regardless of any prehydration water content or $CaCl_2$ concentrations of permeants. These hydraulic conductivity values were significantly lower than those obtained by Vasko et al. (2001). This is because the CGB used in this study contains powdered bentonites. As discussed in the previous chapter on non-prehydration conditions, powdered bentonite CGBs are more compatible against chemical solutions for the non-prehydration condition. Similar to this, powdered bentonites are more chemically compatible for prehydration condition as well.

5 PERFORMANCE OF MODIFIED BENTONITE

In addition to prehydration, the use of a chemically resistant bentonite is considered to be a greatly anticipated countermeasure against chemical attack. Recently, several types of chemically resistant bentonite have been developed to enhance their capability and performance (e.g., Onikata et al. 1996 and 1999, Lo et al. 1997). Multiswellable bentonite (MSB), developed by Onikata et al. (1999), is bentonite which has been mixed with propylene carbonate (PC) to activate the osmotic swelling capacity. Propylene carbonate is placed in the interlayer of the smectite and attracts numerous water molecules. This results in a strong swelling power even if the permeant contains polyvalent cations or a high concentration of monovalents.

Figure 9 shows values for the swell volume (ASTM D 5890) of natural bentonite (NB) and MSB in NaCl and $CaCl_2$ solutions (Katsumi et al. 2001). In NaCl solutions, the swelling power of MSB is clearly greater than that of NB for concentrations lower than 0.6 mol/L, while the difference in swelling power is negligible for concentrations higher than 0.7 mol/L. A larger swelling power for MSB than NB was observed for $CaCl_2$ concentrations lower than 0.5 mol/L. The swelling power of 10 mL/2 g-solid, which is considered the minimum value for the swelling power of this bentonite, was achieved for NB with $CaCl_2$ concentrations higher than only 0.1 mol/L, while the same value was not achieved for MSB when the $CaCl_2$ concentrations were lower than 0.3 mol/L. It can be concluded, therefore, that MSB exhibits higher swelling power than NB for electrolyte solutions which have concentrations lower than a certain level.

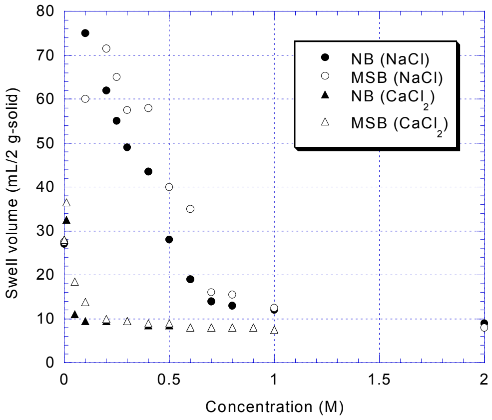


Figure 9 Swell volume of chemical resistant bentonite (multiswellable bentonite: MSB) versus natural bentonite (NB) for NaCl and $CaCl_2$ solutions (Katsumi et al. 2001)

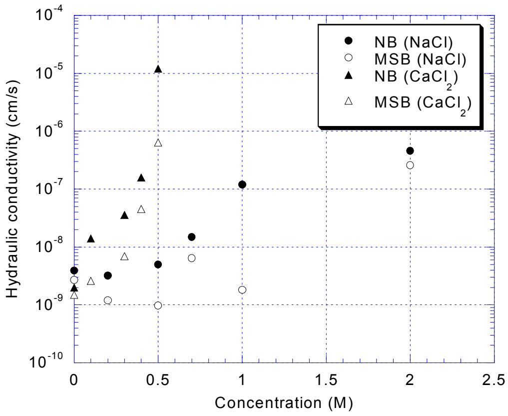


Figure 10Hydraulic conductivity of chemical resistant bentonite (multiswellable bentonite: MSB) versus natural bentonite (NB) for NaCl and $CaCl_2$ solutions (Katsumi et al. 2001)

Figure 10 shows values for the hydraulic conductivity of natural bentonite (NB) and MSB permeated with NaCl and $CaCl_2$ solutions as pore liquids (Katsumi et al. 2001). Hydraulic conductivity tests were performed using flexible-wall permeameters, according to ASTM D 5084. Specimens with a diameter of 6 cm or 10 cm were prepared, and NB or MSB granules were placed between the top and the bottom pedestals to achieve a thickness of approximately 1 cm. This condition is supposed to simulate the performance in cases where these bentonites are used as CGBs, as well as simply to evaluate the chemical compatibility of the bentonites. Each specimen had a dry density of $0.79 g/cm^3$. Under a cell pressure of 20-30 kPa, the bentonite specimens were exposed to the permeant liquid for longer than 24 hours. Then, a hydraulic gradient of 80-90 was applied to the specimens. MSB exhibits higher swell volumes than NB for all concentration levels, and is particularly excellent for concentrations lower than 0.5 mol/L. Most values for the hydraulic conductivity of MSB are 1-2 orders in magnitude lower than the values for that of NB for the same concentration levels. A hydraulic conductivity level lower than $1x10^{-7}$ cm/s can be achieved for a $CaCl_2$ concentration that is lower than 0.5 mol/L. This means that the applicability of MSB as a landfill barrier material can be encouraged. Although the experiments were conducted to evaluate only the applicability to CGBs, MSB may also be advantageous for the application other barriers such as compacted clay liners and bentonite slurry walls from the view of chemical compatibility.

6 CONCLUSION

The main results obtained from this study are as follows:

Powdered bentonites in GCL are more compatible than granular bentonites against chemicals, in particular aggressive (poly-valent and high concentration) solutions. This is probably because the pores between the granules are not blocked due to a lower level of swelling of the bentonite, especially for aggressive chemical solutions. Further research is needed to summarize the effect of multi-species of chemical solutions.

Prehydration of CGBs provided only 0-1 orders of magnitude lower hydraulic conductivity than non-prehydrated CGBs for the chemical solutions, although CGBs were prehydrated to 200% water content at maximum when the water was supplied from the bottom of the base soil. CGBs were not uniformly prehydrated over the area when they were put with the woven geotextile at the bottom, which contributes to the undesirable hydraulic performance. This suction-induced prehydration does not provide the enough prehydration compared to the prehydration by means of direct permeation of water, and therefore their hydraulic conductivity values are higher than the values with deionized water.

From free swell and hydraulic conductivity tests conducted on multiswellable bentonite (MSB), a chemically-resistant modified bentonite, and natural bentonite (NB) with NaCl and $CaCl_2$ solutions, MSB exhibits higher swelling and lower hydraulic conductivity values for NaCl at all levels of concentration of 0-2 mol/L, and for 0-0.5 mol/L $CaCl_2$ solutions. In particular, the hydraulic conductivity values of MSB are always 1-2 orders of magnitude lower than those of NB. Thus, MSB is expected to be an excellent alternative barrier material for landfill liner systems.

7 ACKNOWLEDGEMENTS

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