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Factors affecting the chemical compatibility and the barrier performance of GCLs

Des facteurs influant sur la compatibilité chimique et la performance de GCLs comme barrière

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ABSTRACT

The chemical compatibility and the barrier performance of geosynthetic clay liners (GCLs) were examined in this paper. The use of GCLs as hydraulic barriers in liner and cover systems for waste containment facilities is on the rise. GCLs consist of a thin layer of bentonite sandwiched between two geotextiles or glued to a geomembrane. Their extremely low hydraulic conductivity to water is attributed to the bentonite in the GCLs, which is composed primarily of a smectite mineral. However, there have been concerns as to the chemical compatibility of GCLs, since bentonite is very sensitive to chemical effects and this can lead to an increase in hydraulic conductivity. The present study focuses on (1) the difference in performance between powdered and granular types of bentonite, (2) the influence of real waste leachates on the hydraulic conductivity of GCLs, (3) the effect of the overburden pressure on the hydraulic conductivity of GCLs, and (4) the chemical compatibility of modified bentonite and its long-term performance.

RÉSUMÉ

La compatibilité chimique et la performance des géosynthétiques bentonitiques (GCLs) comme barrière ont été examinées dans cet article. GCLs sont utilisés comme barrières dans les systèmes d'ecouvrement pour confinement de déchets. Ils consistent en une couche mince de la bentonite insérée entre deux géotextiles ou mettant sur un géomembrane. La conductibité hydraulique très base contre l'eau est attribuée à la bentonite dans GCL, qui est essentiellement composée en minéral smectique. Cependant, il y a des problèmes sur la compatibilité chimique de GCLs, puisque les bentonites sont très sensbles à des interactions chimiques, qui peuvent augmenter la conductivité hydraulique. Dans cet article, nous étudions (1) la différence à performance entre des bentonites en poudre et granuleuses, (2) l'influence d'eaux de déchets sur la conductivité hydraulique de GCLs, (3) l'effet de pression du dessus à la conductivité hydraulique de GCLs, et (4) la compatibilité chimique de bentonite modifiée et sa performance de long temps.

1 INTRODUCTION

Clay liners that are used as bottom liners in waste containment facilities are required to have low hydraulic conductivity (k) in order to prevent the groundwater from being contaminated. Geosynthetic clay liners (GCLs), which are factory-manufactured clay liners consisting of a thin layer of bentonite clay encased by geotextiles or glued to a geomembrane, have been considered as barrier materials alternate to or combined with compacted clay liners, because they are relatively low in cost, easy to install, require smaller space, and show low hydraulic conductivity to water. For GCLs that do not contain a geomembrane, bentonite is responsible for the low hydraulic conductivity. Since bentonite is sensitive to chemicals, many studies have been conducted on the chemical compatibility of GCLs, and have shown that the hydraulic conductivity is affected by the type and the concentration of chemicals, and by a sequence of permeant liquids (Daniel et al. 1993; Petrov and Rowe 1997; Ruhl and Daniel 1997; Jo et al. 2001; Vasco et al. 2001). In addition to these findings, there are several factors which might affect the hydraulic conductivity, although little has been reported on them. Thus, this study focuses on some of the factors, namely, (1) the difference in performance between powdered and granular types of bentonite, (2) the influence of real waste leachates on the hydraulic conductivity of GCLs, (3) the effect of the overburden pressure on the hydraulic conductivity of GCLs, and (4) the chemical compatibility of modified bentonite and its long-term performance. Although this study neither considers an exact design nor discusses field performances, based on the equivalency to a compacted clay liner thicker than 60 cm and $k < 1 \times 10^{-7}$ cm/s, a hydraulic conductivity of 5×10^{-9} cm/s, which was addressed by Giroud et al. (1997), might be one reference criteria.

2 BARRIER PERFORMANCE AGAINST INORGANIC CHEMICAL SOLUTIONS AND WASTE LEACHATES

To evaluate the barrier performance of GCLs against inorganic chemical solutions, calcium chloride ($CaCl_2$) and sodium chloride (NaCl) solutions with different *RMD* (ratio of monovalent to divalent) and I (ionic strength) values were used for the free swell and the hydraulic conductivity tests on a needle-punch type of GCL. $RMD = c_1/(2\ c_2)^{0.5}$ and $I = 0.5\ \Sigma\ c_i z_i^2$, in which c_i and z_i are the concentration and the valence of the ith ion and c_1 and c_2 are the concentrations of monovalent and divalent cations, respectively. According to the Stern-Guoy model (Mitchell 1993), the thickness of the diffused double layer (DDL) is thought to be related to the hydraulic conductivity and to be inversely proportional to the square root of the ionic strength. To calculate the ionic strength in this study, only the cations are taken into account. This is because the ions which affect the clay properties may be mainly cations. The GCLs used here contained powdered natural sodium bentonite encapsulated between slit-film monofilament woven and staple-fiber non-woven geotextiles. The geotextiles have been bonded by needle-punched fibers thermally fused to the woven geotextiles. The mass of bentonite per area was approximately 4.9 kg/m^2. Free swell tests were conducted according to ASTM D 5890 on the bentonite derived from the GCLs. In the hydraulic conductivity tests, the GCL specimens were set in a flexible-wall permeameter under a cell pressure of 30 kPa and were directly exposed to the chemical solutions for 2 days; then a hydraulic gradient of approximately 80 - 90 was applied.

The hydraulic conductivities of the GCLs permeated with NaCl - $CaCl_2$ solutions, as well as those reported by Kolstad et al. (2004) permeated with LiCl - $CaCl_2$ solutions for a GCL made through the same manufacturing process but containing a granular bentonite, are shown in Figure 1. According to the

results of the swell volume, the two (powdered and ground) types of bentonite have almost the same sensitivity against chemicals (the swell volume is measured with the ground bentonite). The results of the hydraulic conductivity indicate that both GCLs are affected by the chemical solutions, but the powdered bentonite may be more compatible than the granular bentonite, particularly against 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 significantly increased the hydraulic conductivity. In contrast, the pores of the powdered bentonite are small, even when the swelling is limited due to the chemical effect, and they provide low hydraulic conductivity. Katsumi et al. (2004a and 2004b) reported that powdered bentonite is also more chemically compatible for prehydration conditions.
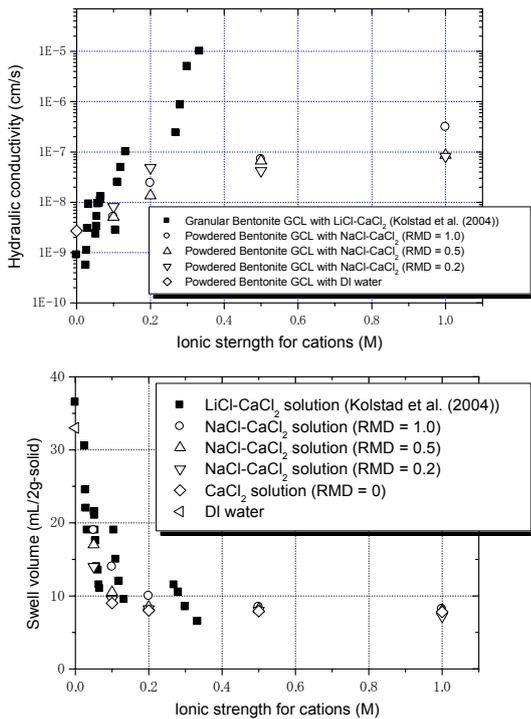

Figure 1 Hydraulic conductivities of GCLs (upper) and the swell volumes of bentonite (lower) versus the ionic strength for cations in the liquids
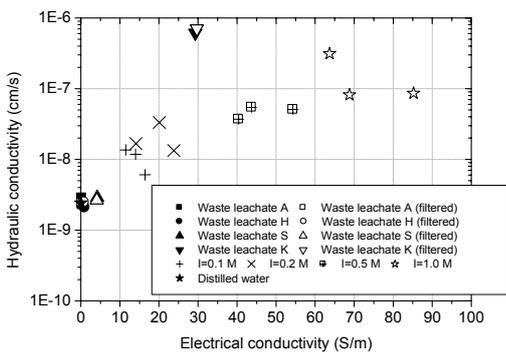

Figure 2 Hydraulic conductivities of the powdered bentonite GCL against waste leachates and the NaCl - CaCl₂ solutions

The hydraulic conductivity values permeated with the real waste leachates, obtained from four landfill sites (Landfills A, H, S, and K) operated by different municipal governments in Japan, are shown in Figure 2. Both raw and filtered waste leachates were used. The results for the NaCl - CaCl₂ solutions are also plotted in Figure 2. From Figure 1, the hydraulic conductivity was mainly dependent upon the ionic strengths, and was affected little by the *RMD*. It is impossible to obtain the io-

nic strength of waste leachates, because they contain various kinds of organic and inorganic chemicals. Thus, the data are plotted with the electrical conductivity in Figure 2 due to the ease of measurement and a considerable correlation with the ionic strength.
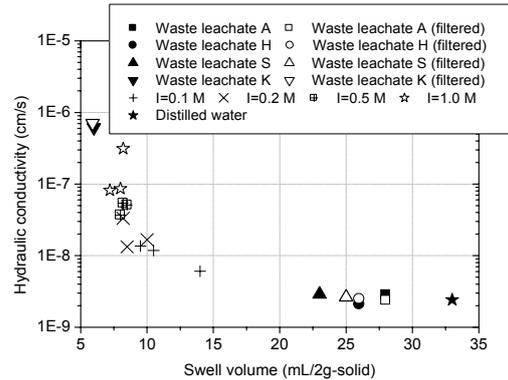

Figure 3 Swell volume of bentonite versus the hydraulic conductivity of a GCL against the waste leachates and the NaCl - CaCl₂ solutions

A strong correlation between the electrical conductivity versus the hydraulic conductivity is clearly obtained for the NaCl - CaCl₂ solutions. For waste leachates at sites A, H, and S, the electrical conductivity values were very low (< 5 S/m), and low hydraulic conductivities equivalent to those for pure water permeation were obtained ($\sim 2 \times 10^{-9}$ cm/s). In contrast, the leachate at site K, having high electrical conductivity (~ 30 S/m), provided more than two orders of magnitude higher hydraulic conductivities ($> 5 \times 10^{-7}$ cm/s) than pure water. These hydraulic conductivity values were more than one order of magnitude higher than those for the NaCl - CaCl₂ solutions for the same level of electrical conductivity. This is probably because electrical conductivity is an index only for inorganic chemicals; it cannot account for the organic chemicals present even if they may affect the clay-chemical interactions. A strong correlation between the swell volumes of bentonite versus the hydraulic conductivity of a GCL was obtained for both the NaCl - CaCl₂ solutions and the waste leachates (Figure 3). The hydraulic conductivity of the GCL against the waste leachates can be roughly predicted from the electrical conductivity of the leachate liquids and the free swell volume of the bentonite, similar to the hypothesis presented by Jo et al. (2001).

3 EFFECTS OF THE OVERBURDEN PRESSURE

Most of the previous studies on the chemical compatibility of GCLs used flexible-wall permeameters under a relatively low cell pressure (20 - 30 kPa) (Daniel et al. 1993; Ruhl and Daniel 1997; Jo et al. 2001; Vasco et al. 2001). However, GCLs used as bottom barriers may be subjected to the overburden pressure of reclaimed wastes, and expectedly may exhibit smaller void ratios and lower hydraulic conductivity levels than those assessed in the laboratory under a low confining pressure. The effect of the overburden pressure may be significant because the bentonite in the GCLs is typically a compressible material. The previous research works on this subject are limited to a case using pure water as the permeant (Shackelford et al. 2000) and a case in which a relatively low confining pressure (< 120 kPa) are applied using NaCl solutions (Petrov and Rowe 1997). In this study, hydraulic conductivity tests were conducted using an oedometer cell under certain vertical pressure levels with NaCl or CaCl₂ solutions. The GCL specimens were exposed to the solutions to hydrate, next they were consolidated with vertical pressure levels (29.4, 78.5, 314, and 1256 kPa) for 24 hours to obtain a degree of consolidation higher than 90%, and then they were subjected to a hydraulic gradient of 80 - 90. Solutions with a CaCl₂ of 0.25 M and an NaCl of 2.0 M were selected because their concentrations are high enough to affect the hydraulic

conductivity and also because almost the same values were obtained for the hydraulic conductivity under a cell pressure of 30 kPa in the flexible-wall permeameter.

An increase in the overburden pressure significantly reduced the hydraulic conductivity even for the chemical solutions which have a chemical effect (Figure 4). A powdered bentonite GCL permeated with a NaCl solution of 2.0 M and a granular bentonite GCL with a $CaCl_2$ solution of 0.25 M exhibited the hydraulic conductivity of approximately 2×10^{-8} cm/s for a vertical pressure of 300 kPa, while 1×10^{-9} cm/s was obtained for a powdered bentonite GCL with a $CaCl_2$ solution under a vertical pressure of 300 kPa. These values are low enough that the GCL can be used as a landfill bottom barrier alternate to or at least in combination with compacted clay liners. Shackelford et al. (2000) indicated that the increase in overburden pressure from 20 kPa to 500 kPa led to the decrease in the GCL hydraulic conductivity of only 1/4 to 1/10 for pure water permeation. In contrast, the experimental results presented in this study showed a significant decrease (2 orders of magnitude) in hydraulic conductivity due to the increase in consolidation pressure. The deepest section at the bottom of the landfill will have built-up waste leachate, and therefore, a higher potential for leakage of the leachate through the liners will exist. However, such a section will have a great overburden pressure from the reclaimed wastes, and consequently, the hydraulic conductivity of the GCLs will become low enough that the GCLs may effectively function as a barrier.
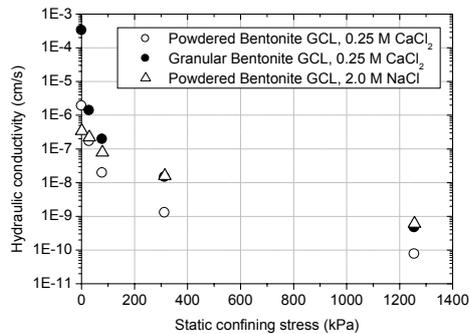

Figure 4 Confining stress versus hydraulic conductivities of GCLs

The effects of the overburden pressure on hydraulic conductivity are dependent on several factors such as the type of permeants and the properties of the bentonite, including whether the bentonite is powdered or granular. Osmotic swelling occurs for the smectite mineral in the bentonite, if the exchangeable cations are monovalent, while only crystalline swelling occurs when polyvalent cations are present or the cation concentration is high. These swelling mechanisms may have an effect on the relations between the hydraulic conductivity and the void space. Figure 5 shows the relationship between the void ratio and the hydraulic conductivity. The data from similar experiments on NaCl solutions reported by Petrov and Rowe (1997) are also plotted. Based on the data by Petrov and Rowe (1997) and Mesri and Olson (1971), Shackelford et al. (2000) have shown that changes in the void ratio have a similar effect on the hydraulic conductivity regardless of the cation concentration or the thickness of the adsorbed layer. The effect of varying the total void space appears to be independent of the effect of changing the thickness of the adsorbed layer, which is influenced by the cation (Na^+) concentration. The fraction of the total void space that conducts the flow is controlled by the thickness of the adsorbed layer. From Figure 5, the effect of the void space on the hydraulic conductivity is independent regardless of whether the bentonite is powdered or granular and also regardless of the concentrations. This is consistent with Shackelford et al. (2000). More importantly, the effect of void space clearly depends on the type of chemicals: NaCl or $CaCl_2$. Powdered bentonite provided one order of magnitude lower hydraulic conductivity values than granular bentonite for the same void ratios. In conclusion, to

evaluate the barrier performance of a GCL in the field, careful consideration should be given to the type of GCL and the type of bentonite (powdered or granular) used, the expected overburden pressure, and the anticipated chemical properties of the waste leachate.
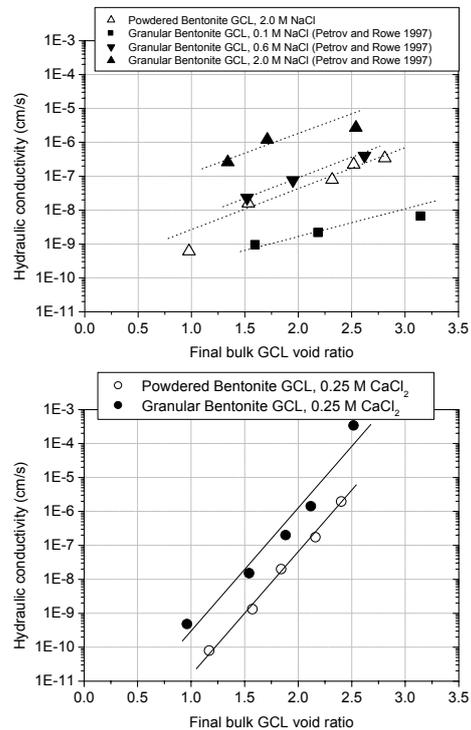

Figure 5 Void ratios versus hydraulic conductivities of GCLs (upper: NaCl solutions and lower: $CaCl_2$ solutions)

4 LONG-TERM BARRIER PERFORMANCE OF CHEMICAL-RESISTANT BENTONITE

The use of chemically-resistant bentonite is considered to be another greatly anticipated measure against chemical attack. Multiswellable bentonite (MSB), developed by Onikata et al. (1996), is bentonite mixed with propylene carbonate (PC) to activate the osmotic swelling capacity. PC is placed in the interlayer of the smectite and functions to attract numerous water molecules. This results in a strong swelling power even when the permeant contains polyvalent cations or a high concentration of monovalents. One to two orders of magnitude lower hydraulic conductivities have been reported for MSB than for natural bentonite for NaCl and $CaCl_2$ solutions (Katsumi et al. 2001 and 2004a). Their experiments were limited to the results of tests conducted over only a few months. To evaluate the long-term stability and performance, the results of a three-year hydraulic conductivity test on MSB are herein presented following their reports. MSB granules were placed between the top and the bottom pedestals, with a diameter of 6 cm and a thickness of approximately 1 cm in the flexible-wall permeameter. This condition is to simulate the performance in cases where the bentonite is used as a GCL, as well as simply to evaluate the chemical compatibility of the bentonite. Each specimen had a dry density of approximately 0.79 g/cm^3. Typical termination criteria for the hydraulic conductivity test are (1) equality of the inflow and outflow rates (<25%) and (2) measurement of a steady hydraulic conductivity. In addition, for the chemical compatibility test, (3) permeation of a minimum of two pore volumes of flow should be obtained, and (4) a similarity in the chemical compositions between effluent and influent should be ensured (Shackelford et al. 2000). For the fourth criteria, the electrical conductivities of influents and effluents were checked.

The hydraulic conductivity tests for NaCl solutions of 0.5, 0.7, and 1.0 M achieved chemical equilibrium after a permeation of 2 - 3 years; effluents and influents had almost the same electrical conductivity for these solutions. The case with a NaCl of 0.2 M has not yet achieved the termination criterion on chemical equilibrium, and the hydraulic conductivity test is still being continued. The data on the hydraulic conductivities were scattered due to the difficulty in accurately measuring the thickness using a cathetometer (Figure 6). If the data on the influent or the effluent volumes instead of the hydraulic conductivities were plotted, little scatter would be obtained. The hydraulic conductivities for NaCl solutions of 0.2 - 1.0 M are competitive to the one for pure water ($\sim 1 \times 10^{-9}$ cm/s) (Figure 7). Important issues for the long-term performance may be the stability of the PC and the clay-chemical interactions. The PC will deteriorate to polypropylene glycol. If this deterioration occurs only after osmotic swelling occurs, however, it will not affect the swelling (Onikata et al. 1996). Although a chemical equilibrium has not yet been achieved for a NaCl of 0.2 M, the hydraulic conductivity will be kept low for a longer permeation, because an increase in the hydraulic conductivity was not obtained for the higher concentrations (0.5, 0.7, and 1.0 M) for which a chemical equilibrium was achieved. In conclusion, once swelling occurs and low hydraulic conductivity is achieved in the early stage of permeation, the long-term barrier performance may be attained for MSB.
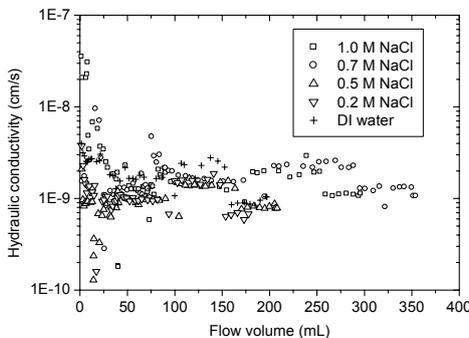

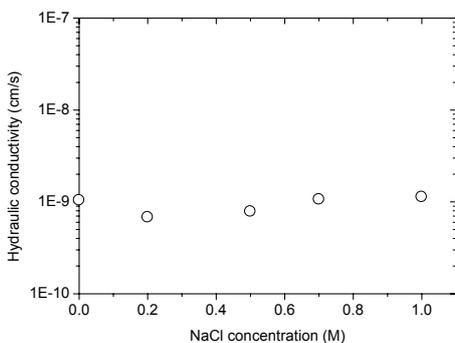Figure 6 Hydraulic conductivity values of MSB permeated with NaCl solutions

Figure 7 NaCl concentration versus the hydraulic conductivity of MSB

5 CONCLUSIONS

The main conclusions obtained from this study are as follows: (1) Powdered bentonite is more compatible against chemical solutions, in particular with high cation concentrations, than granular bentonite. (2) The hydraulic performance against waste leachates may be predicted from the electrical conductivity of the liquid and the free swell volume of the bentonite against the liquid. (3) The effects of the overburden pressure on the hydraulic conductivity permeated with NaCl and CaCl$_2$ solutions were significant and depended on the type of chemicals. Hydraulic conductivities of 1×10^{-9} to 2×10^{-8} cm/s were obtained under an overburden pressure of 300 kPa. (4) Multiswellable bentonite

(MSB) provided a better barrier performance, and it is expected to have long-term stability from the results of hydraulic conductivity tests conducted over a period of 3 years. It is believed that these results will provide useful information when GCLs are used as landfill bottom barriers.

REFERENCES

Daniel, D.E., Shan, H.-Y., and Anderson, J.D. 1993. Effects of partial wetting on the performance of the bentonite component of a geosynthetic clay liner, *Geosynthetics '93,* IFAI, pp. 1482-1496.

Giroud, J.P., Badu-Tweneboah, K., and Soderman, K.L. 1997. Comparison of leachate flow through compacted clay liners and geosynthetic clay liners in landfill liner systems, *Geosynthetics International,* Vol. 4, Nos. 3-4, pp. 391-431.

Jo, H.Y., Katsumi, T., Benson, C.H., and Edil, T.B. 2001. Hydraulic conductivity and swelling of non-prehydrated GCLs permeated with single species salt solutions, *Journal of Geotechnical and Geoenvironmental Engineering,* ASCE, Vol.127, No.7, pp.557-567.

Katsumi, T., Onikata, M., Hasegawa, S., Lin, L., Kondo, M., and Kamon, M. 2001. Chemical compatibility of modified bentonite permeated with inorganic solutions, *Geoenvironmental Engineering,* R.N. Yong and H.R. Thomas (eds.), Thomas Telford, pp. 419-424.

Katsumi, T., Ogawa, A., Fukagawa, R. 2004a. Effect of chemical solutions on hydraulic barrier performance of clay geosynthetic barriers, *Proceedings of the Third European Geosynthetics Conference,* pp. 701-706.

Katsumi, T., Ogawa, A., and Fukagawa, R. 2004b. Effect of prehydration on hydraulic performance of geosynthetic clay liners permeated with inorganic chemical solutions, *Proceedings of the Third Asian Regional Conference on Geosynthetics,* pp. 937-944.

Kolstad, D.C., Benson, C.H., and Edil, T.B. 2004. Hydraulic conductivity and swell of nonprehydrated geosynthetic clay liners permeated with multispecies inorganic solutions, *Journal of Geotechnical and Geoenvironmental Engineering,* ASCE, Vol.130, No.12, pp.1236-1249.

Mesri, G. and Olson, R. 1971. Mechanisms controlling the permeability of clays, *Clays and Clay Minerals,* Vol. 19, pp. 151-158.

Mitchell, J.K. 1993. *Fundamentals of Soil Behavior, 2nd Edition,* John Wiley & Sons, Inc., New York, 437 p.

Onikata, M., Kondo, M., and Kamon, M. 1996. Development and characterization of a multiswellable bentonite, *Environmental Geotechnics,* M. Kamon (ed.), Balkema, pp. 587-590.

Petrov, R.J. and Rowe, R.K. 1997. Geosynthetic clay liner (GCL) – chemical compatibility by hydraulic conductivity testing and factors impacting its performance, *Canadian Geotechnical Journal,* Vol. 34, pp. 863-885.

Ruhl, J.L. and Daniel, D.E. 1997. Geosynthetic clay liners permeated with chemical solutions and leachates, *Journal of Geotechnical and Geoenvironmental Engineering,* ASCE, Vol.123, No.4, pp.369-381.

Shackelford, C.D., Benson, C.H., Katsumi, T., Edil, T.B., and Lin, L. 2000. Evaluating the hydraulic conductivity of GCLs permeated with non-standard liquids, *Geotextiles and Geomembranes,* Elsevier, Vol. 18, Nos. 2-3, pp. 133-161.

Vasko, S.M., Jo, H.Y., Benson, C.H., Edil, T.B., and Katsumi, T. 2001. Hydraulic conductivity of partially prehydrated geosynthetic clay liners permeated with aqueous calcium chloride solutions, *Geosynthetics Conference 2001,* IFAI, pp. 685-699.