

## HYDRAULIC CONDUCTIVITY AND SWELL OF NONPREHYDRATED GEOSYNTHETIC CLAY LINERS PERMEATED WITH MULTISPECIES INORGANIC SOLUTIONS

Kolstad, D.C., Benson, C.H. and Edil, T.B., *Journal of Geotechnical and Geo-environmental Engineering*, ASCE, Vol. 130, No. 12, December 2004, pp.1236-1249.

**Introduction.** This article examines the influence of multispecies inorganic solutions on swelling and hydraulic conductivity of non-prehydrated GCLs containing sodium bentonite. This is a continuation of the research done by Benson, Edil and their students in the area of clays, including GCLs (see CETCO TR-326). Multispecies inorganic solutions were mixed using deionized water, monovalent cation salts (NaCl and/or LiCl) and divalent cation salts (CaCl<sub>2</sub> and MgCl<sub>2</sub>). GCL hydraulic conductivity tests were performed using flexible wall permeameters at an average effective stress of 20 kPa (3 psi) and average head pressure of 2 feet. The GCL hydraulic conductivity tests were run per ASTM D6766 until the influent and effluent electrical conductivity deviated less than 10%. Free swell tests were also conducted per ASTM D6141 using the multispecies inorganic solutions as the test liquid.

Ionic strength and the ratio of monovalent cation concentration to the square root of divalent cation concentration (RMD) in the permeant solution were found to influence swell of the bentonite and hydraulic conductivity of GCLs. A regression model was developed relating hydraulic conductivity of the GCL to ionic strength and the RMD of the permeant solution. The results of this model are expressed in equation 3 and graphically in Figures 7 and 10(a) of the article.

A literature search of leachate chemistry data from different waste containment facilities was also conducted and listed in Table 4. Figure 10(b) plots the ionic strength and RMD of these various leachates onto a corresponding isoperm chart. The chart implies that many municipal solid waste (MSW) landfill leachates and mine waste site leachates, as well as some hazardous waste and fly ash leachates, would result in high ( $>10^{-7}$  cm/s) GCL hydraulic conductivities.

**Errata.** An erratum was published by the authors in the July 2006 issue of ASCE *Journal of Geotechnical and Geo-environmental Engineering*. The following corrections were noted:

- The RMD units in the text, tables and figures should be  $M^{1/2}$ , not  $mM^{1/2}$ .
- Equation 3. The correct equation is:

$$\log K_c / \log K_{DI} = 0.965 - 0.976 * I + 0.0797 * RMD + 0.251 * I^2 * RMD$$

- The fly ash leachate point coordinates from Table 4 are plotted incorrectly in Figure 10(b). According to the data, all but one fly ash point should be between the  $10^{-8}$  and  $10^{-9}$  cm/s isoperm lines.

**Data Review.** Most of the MSW leachate chemistry data listed in Kolstad Table 4 is from prior to 1990. MSW leachate chemistry data for cells built prior to 1990 is not representative of modern MSW landfills because: 1) it was common practice to dispose of industrial waste, hazardous waste and various liquid wastes prior to promulgation of current national solid waste regulations, and 2) samples may not have been taken from controlled collection points because many landfills did not have leachate collection and recovery systems (Bonaparte et al., 2002). The difference between these two time periods in MSW leachate chemistry is quite apparent by segregating pre-1990 and post-1990 data. For those references published *prior to* 1990, the average ionic strength was 0.24 M with an average RMD of  $0.22 \text{ M}^{1/2}$  and the maximum ionic strength was 0.62 M with a RMD of  $0.31 \text{ M}^{1/2}$ . For those references published *after* 1990, the average ionic strength is 0.12 M with an RMD of  $0.16 \text{ M}^{1/2}$  and a maximum ionic strength of 0.24 M with a RMD of  $0.18 \text{ M}^{1/2}$  (Table A).

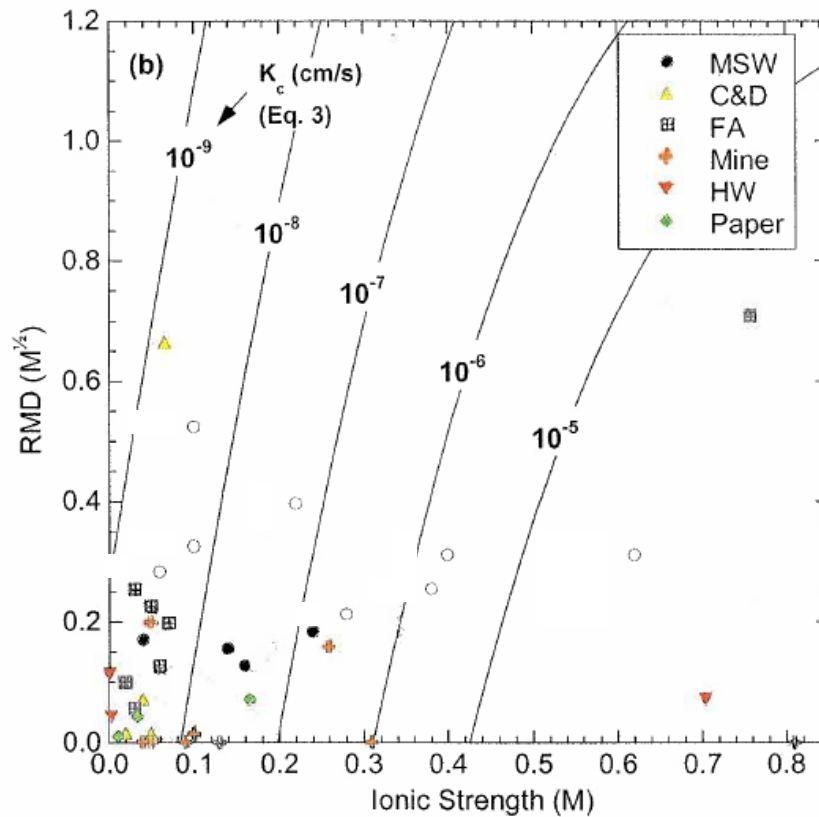
Table A. MSW Landfill Leachate Chemistry Data.

	Average		Maximum	
	Ionic Strength (M)	RMD ( $\text{M}^{1/2}$ )	Ionic Strength (M)	RMD ( $\text{M}^{1/2}$ )
Kolstad – 17 pre-1990 cases	0.24	0.22	0.62	0.31
Kolstad – 5 post-1990 cases	0.12	0.16	0.24	0.18
EPA Study – 26 post-1990 cases	0.06	0.11	0.22	0.22
Combined 31 post-1990 cases	0.07	0.11	0.24	0.18

The segregated Kolstad data shows that modern MSW leachates (post-1990) have a lower ionic strength than older MSW leachates (pre-1990).

Leachate chemistry data collected from 26 post-1990 MSW landfills as part of a major study by USEPA (Bonaparte et al. 2002) is also presented in Table A. Ionic strengths were estimated from specific conductance per Snoeyink and Jenkins (1980). RMDs were estimated from available calcium, magnesium and sodium concentrations. The MSW leachates in the USEPA study had an average estimated ionic strength of 0.06 M and a RMD of  $0.11 \text{ M}^{1/2}$ . Combining post-1990 cases from both Kolstad and USEPA yields an average ionic strength of 0.07 M and RMD of  $0.11 \text{ M}^{1/2}$ .

Modifying Figure 10(b) to correct the fly ash coordinates and to delete the Kolstad MSW data prior to 1990 yields Figure 1.



**Figure 1.** Modified Figure 10(b) from Kolstad et al. 2004 for nonprehydrated GCL permeated at 3 psi average effective stress. When fly ash coordinates are corrected and pre-1990 MSW leachate chemistry data is removed, few leachates cause a significant increase in GCL hydraulic conductivity.

**Data Interpretation.** The GCL tested in Kolstad's research had a hydraulic conductivity of  $9 \times 10^{-10}$  cm/s when permeated with distilled water. Kolstad Equation 3 predicts the hydraulic conductivity of the nonprehydrated GCL permeated with inorganic chemicals. Using the post-1990 MSW leachate chemistry from Table A, the Kolstad model yields an estimated average nonprehydrated GCL long-term hydraulic conductivity of  $6.4 \times 10^{-9}$  cm/s and a maximum of  $1.7 \times 10^{-7}$  cm/s at 3 psi average effective stress.

However, as noted by the authors, the model expressed in Equation 3 does not take into account 1) higher effective stresses which tend to decrease permeability, 2) prehydration from subgrade or 3) increasing pH over time. The model should be adjusted to account for at least the first two important factors. Petrov (1997) developed the following equation for the effect of confining stress on GCL hydraulic conductivity permeated with distilled water:

$$\log K_{DI} = -8.0068 - 0.5429 \log \sigma$$

Where  $K_{DI}$  is in cm/s and  $\sigma$  is in kPa.

Bonaparte et al. found that 74 MSW cells in the USEPA study had maximum heights ranging from 26 to 226 feet, with an average of 90 feet. Assuming a MSW density of 60 pcf (Daniel 1993), the typical effective stress of a MSW liner is 37 psi. Although the expression developed by Petrov is stated to be valid only for effective stress between 3 and 120 kPa (0.4 to 17 psi), the correlation appears to trend linearly. Therefore, it is reasonable to use this expression for 37 psi stress, which is just slightly higher on the logarithmic scale. For an increase in maximum effective stress from 5 psi to 37 psi, the Petrov equation yields a 65% decrease in GCL hydraulic conductivity. Predictions from the Kolstad model, adjusted for post-1990 MSW leachate chemistry data and an effective stress of 37 psi, yield an estimated average nonprehydrated GCL long-term hydraulic conductivity of  $2.3 \times 10^{-9}$  cm/s and a maximum of  $6 \times 10^{-8}$  cm/s for MSW landfills.

Even lower hydraulic conductivity would be expected if the GCL prehydrates from subgrade moisture. Lee and Shackelford (2005) showed that GCL prehydrated with deionized water and then permeated with a 0.1M  $\text{CaCl}_2$  solution ( $I = 0.3$  M) had a hydraulic conductivity 3 times lower compared to a nonprehydrated GCL. But testing at  $\text{CaCl}_2 = 0.05\text{M}$  ( $I = 0.15\text{M}$ ) indicated that the prehydration effect was insignificant. It is possible that prehydration of the GCL will improve long-term hydraulic conductivity at  $I = 0.24\text{M}$ , the upper end of the MSW ionic strength reported. However, more study in this area is needed.

The highest MSW leachate ionic strength was for young MSW leachate (<5 years). After waste placement ceases and the landfill is capped, methanogenic organisms begin to proliferate in a MSW landfill and the pH begins to approach neutrality as the acids are converted into methane and a bicarbonate buffering system is established during the methane fermentation stage (Bonaparte et al 2002). Concentrations of free divalent cations will decrease with increased pH, due to the solubility of divalent cations (Snoeyink and Jenkins 1980). Consequently, after closure, the ionic strength of the MSW leachate will decrease. Therefore, it can be argued that with GCL prehydration and/or leachate aging that the previously estimated average GCL hydraulic conductivity of  $2.3 \times 10^{-9}$  for MSW landfills is quite conservative.

Giroud (1997) has formulated equations for leakage rates through defects in geomembranes. Under typical MSW landfill conditions; these formulas indicate that the leakage rate through geomembrane defects is controlled by the hydraulic conductivity of the underlying clay. Thus, based upon the estimated GCL long-term hydraulic conductivities stated above, a GM/GCL composite MSW liner would be expected to have a lower leakage rate than a GM/CCL composite MSW liner, where the CCL has a hydraulic conductivity of  $10^{-7}$  cm/s. This is supported by the MSW landfill leakage rate data of GM/GCL and GM/CCL composite liners collected in the USEPA study by Bonaparte et al. (see CETCO TR-316).

**Conclusion.** In their conclusions Kolstad et al. state that, "high hydraulic conductivities (i.e.,  $>10^{-7}$  cm/s) are unlikely for nonprehydrated GCLs in base liners in many solid waste containment facilities". Based on the data review compiled above, it is reasonable to refine this conclusion to a) the hydraulic performance of a GCL in a landfill bottom liner will not be significantly affected by a typical modern MSW leachate and that b) high GCL hydraulic conductivities will only occur in certain cases at non-MSW wastes (e.g., hazardous waste, mine waste, fly ash) when the waste leachate contains higher ionic strengths.

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# Hydraulic Conductivity and Swell of Nonprehydrated Geosynthetic Clay Liners Permeated with Multispecies Inorganic Solutions

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**Abstract:** The influence of multispecies inorganic solutions on swelling and hydraulic conductivity of non-prehydrated geosynthetic clay liners (GCLs) containing sodium bentonite was examined. Ionic strength and the relative abundance of monovalent and divalent cations (RMD) in the permeant solution were found to influence swell of the bentonite, and the hydraulic conductivity of GCLs. Swell is directly related to RMD and inversely related to ionic strength, whereas hydraulic conductivity is directly related to ionic strength and inversely related to RMD. RMD has a greater influence for solutions with low ionic strength (e.g., 0.05 M), whereas concentration effects dominate at high ionic strength (e.g., 0.5 M). No discernable effect of cation species of similar valence was observed in the swell or hydraulic conductivity data for test solutions with similar ionic strength and RMD. A strong relationship between hydraulic conductivity and free swell was found, but the relationship must be defined empirically for a particular bentonite. A regression model relating hydraulic conductivity of the GCL to ionic strength and RMD of the permeant solution was developed. Predictions made with the model indicate that high hydraulic conductivities (i.e.,  $>10^{-7}$  cm/s) are not likely for GCLs in base liners in many solid waste containment facilities. However, for wastes with stronger leachates or leachates dominated by polyvalent cations, high hydraulic conductivities may occur.

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**CE Database subject headings:** Hydraulic conductivity; Swelling; Inorganic chemicals; Clay liners; Bentonite.

## Introduction

Geosynthetic clay liners (GCLs) are factory-manufactured clay liners consisting of a layer of bentonite clay encased by geotextiles or glued to a geomembrane. GCLs have become a popular alternative to compacted clay liners in waste containment applications because of their relatively low cost, ease of installation, perceived resistance to environmental distress (e.g. freeze-thaw and wet-dry cycling), smaller air-space requirements, and low hydraulic conductivity to water ( $<10^{-8}$  cm/s). For GCLs that do not contain a geomembrane, bentonite is responsible for the low hydraulic conductivity. Sodium (Na) montmorillonite mineral is the primary component of bentonite, and largely controls the hydraulic conductivity of GCLs (Shackelford et al. 2000).

A variety of studies have shown that the hydraulic conductivity and swelling of bentonite can be affected by inorganic permeant solutions (Alther et al. 1985; Shan and Daniel 1991; Egloffstein 1997, 2001; Quaranta et al. 1997; Ruhl and Daniel 1997;

Petrov and Rowe 1997; Shackelford et al. 2000; Jo et al. 2001; Vasko et al. 2001; Ashmawy et al. 2002; Katsumi et al. 2002, 2003; Shan and Lai 2002). The general conclusion of these studies is that the hydraulic conductivity and swelling of GCLs is sensitive to the concentration of the permeant solution and the cation valence. In general, higher hydraulic conductivity and lower swell are obtained in more concentrated solutions or solutions with a preponderance of divalent cations. However, no systematic study has been made regarding how the concentration and relative proportions of monovalent and polyvalent cations in a multispecies (i.e., more than one cation species) solution affect swelling and hydraulic conductivity of bentonite and GCLs.

Several studies have been conducted in soil science regarding the effect of multispecies solutions on the hydraulic conductivity of montmorillonitic soils (Reeve and Bower 1960; McNeal and Coleman 1966; McNeal et al. 1966; Mustafa and Hamid 1975; Malik et al. 1992). However, these studies have focused on increasing the hydraulic conductivity of montmorillonitic soils for land drainage and agricultural applications rather than maintaining low hydraulic conductivity for containment applications. Moreover, none of these studies has focused on clay soils very rich in montmorillonite, such as the Na-bentonites used for GCLs.

This paper discusses how the ionic strength and relative amounts of monovalent and divalent cations in multispecies solutions affect swelling and hydraulic conductivity of nonprehydrated GCLs containing Na-bentonite. The focus is on applications where inorganic solutes are the primary factor affecting hydraulic conductivity (e.g., conventional solid waste containment facilities for municipal, hazardous, or mining wastes) and where complete prehydration (i.e., prehydration by permeation with distilled, deionized, or potable water) is unlikely. The effects of complete prehydration and organic compounds are discussed

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by others (e.g., Shan and Daniel 1991; Petrov and Rowe 1997; Ruhl and Daniel 1997; Shackelford et al. 2000).

## Background

### *Exchangeable Cations, Mobility of Water, and Hydration of Bentonite*

A weak interlayer bond allows the montmorillonite crystal layers to separate during hydration as water molecules enter the interlayer space (Grim 1968; van Olphen 1977). Consequently, cations on the interlayer surfaces become exchangeable, which renders the physical properties of Na-montmorillonite susceptible to interactions with the permeant liquid. The degree of exchange depends on the valence, relative abundance, and size of the cations. Generally, cations of greater valence and smaller size replace cations of lower valence and larger size. The preference for replacement is the lyotropic series, which is  $\text{Li}^+ < \text{Na}^+ < \text{K}^+ < \text{Rb}^+ < \text{Cs}^+ < \text{Mg}^{2+} < \text{Ca}^{2+} < \text{Ba}^{2+} < \text{Cu}^{2+} < \text{Al}^{3+} < \text{Fe}^{3+}$  (Sposito 1981; 1989; McBride 1994). Because  $\text{Na}^+$  is at the lower end of the lyotropic series, Na-bentonites are prone to cation exchange when permeated with solutions containing divalent or trivalent ions (Sposito 1981).

Water in the pores of bentonite can be considered mobile or immobile. Mobile water is bulk pore water that is free to move under a hydraulic gradient. Immobile water is bound to the external and internal (i.e., interlayer) mineral surfaces by strong electrical forces, and is believed to act as an extension of the solid surface. When the amount of immobile water in the system increases, the hydraulic conductivity of bentonite decreases because the interparticle flow paths for mobile water become more constricted and tortuous. This is especially true in bentonites where swell is constrained (e.g., needle-punched GCLs or GCLs under confining pressure) (Reeve and Ramaddoni 1965; McNeal and Coleman 1966; McNeal et al. 1966; Lagerwerff et al. 1969; Mesri and Olson 1971; Petrov and Rowe 1997; Shackelford et al. 2000; Jo et al. 2001). Changes in the volume of immobile water also cause volume changes in the bentonite (swell occurs as the volume of immobile water increases). Thus swell and hydraulic conductivity are generally inversely related for bentonites (Shackelford et al. 2000; Jo et al. 2001; Ashmawy et al. 2002; Katsumi et al. 2002).

The fraction of the pore water that is immobile is proportional to the number of layers of water molecules hydrating the interlayer surfaces of the montmorillonite particles (McBride 1994). Hydration of montmorillonite in electrolyte solutions occurs in two phases: the crystalline phase and the osmotic phase (Norrish and Quirk 1954; McBride 1994; Zhang et al. 1995; Prost et al. 1998). The crystalline phase occurs first as several molecular layers of water hydrate the interlayer and outer surfaces from the completely dry state. Osmotic hydration occurs when additional water molecules hydrate the interlayer surfaces, resulting in large interlayer distances (McBride 1994). Crystalline hydration generally results in a small expansion of the interlayer space and a limited amount of immobile water, which is manifested at the macroscale as a small amount of swelling (referred to as “crystalline swell”) and higher hydraulic conductivity. Osmotic hydration can result in appreciable expansion of the interlayer space, a large fraction of the pore water being bound, and is responsible for the large amount of swelling (referred to as “osmotic swell”) and low hydraulic conductivity often associated with Na-bentonites.

When the interlayer cations are monovalent, both crystalline and osmotic hydration occur, allowing the interlayer spacings to become large. However, only crystalline swelling occurs when the interlayer cations are divalent or trivalent, limiting expansion of the interlayer region to approximately 1.96 nm (four layers of water molecules). Strong electrostatic attraction between the montmorillonite sheets and the interlayer cations prevent osmotic swelling when the cations are polyvalent, despite the larger hydration energy associated with polyvalent cations (McBride 1994; 1997; Quirk and Marčelja 1997). Thus, appreciable swelling and lower hydraulic conductivity occur when the interlayer cations are monovalent, whereas very little swelling and higher hydraulic conductivity occur when the cations are divalent or trivalent (Norrish and Quirk 1954; McBride 1994; Wu et al. 1994; Egloffstein 1997, 2001; Onikata et al. 1999; Jo et al. 2001; Ashmawy et al. 2002). In monovalent solutions, the volume of swelling and spacing of the interlayer region is inversely proportional to the square root of the concentration of the solution (Norrish and Quirk 1954; McBride 1994; Zhang et al. 1995; Onikata et al. 1999).

### *Hydraulic Conductivity to Single-Species Inorganic Solutions*

Mesri and Olson (1971) studied the mechanisms controlling the hydraulic conductivity of bentonite when the interlayer cation was sodium or calcium. At similar void ratios, the hydraulic conductivity of Na-bentonite was approximately five times lower than that of the Ca-bentonite. Mesri and Olson (1971) attributed the lower hydraulic conductivity of the Na-bentonite to the presence of immobile water, which resulted in smaller and more tortuous flow paths for mobile water.

Petrov and Rowe (1997) investigated how NaCl solutions of varying concentration affected the hydraulic conductivity of a GCL containing Na-bentonite. Tests were conducted with distilled (DI) water and NaCl solutions having concentrations between 0.1–2.0 M. Hydraulic conductivity of the GCL generally increased as the NaCl concentration increased. At 2.0 M, the hydraulic conductivity was as much as 800 times higher than that with distilled water. For concentrations less than 0.1 M, the hydraulic conductivity was comparable to that obtained with distilled water. Prehydration with at least one pore volume of distilled water tempered the sensitivity of hydraulic conductivity to salt concentration. For 2.0 M NaCl, prehydration with distilled water resulted in a hydraulic conductivity 25 times lower than that obtained by direct permeation with 2.0 M NaCl. Tests conducted over a range of confining stresses (3 to 118 kPa) showed that, at a given concentration, the hydraulic conductivity can vary by a factor of 10 to 50 depending on the effective stress.

Jo et al. (2001) investigated how cation valence and concentration of single-species salt solutions affect free swell and hydraulic conductivity of nonprehydrated GCLs containing Na-bentonite. Salt solutions with cation valences of 1, 2, and 3 and concentrations between 0.005 and 1.0 M were used. All tests were conducted until the physical and chemical termination criteria in ASTM D 6766 were achieved. Permeation with salt solutions having concentrations less than 0.1 M (monovalent) or 0.01 M (divalent or trivalent) yielded hydraulic conductivities similar to those with DI water ( $\approx 10^{-9}$  cm/s), regardless of cation valence. For higher concentrations, swell decreased and hydraulic conductivity increased as the concentration or valence increased. Swelling in the presence of monovalent cations followed the order of the hydrated radius ( $r_h$ ) and the lyotropic series, with Li ( $r_h \approx 0.6$  nm) solutions yielding the greatest swell and K ( $r_h$

$\approx 0.3$  nm) solutions yielding the lowest swell at a given concentration. In contrast, hydraulic conductivity to the monovalent solutions was insensitive to cation species. No dependence on species was observed for swell or hydraulic conductivity when the solutions contained divalent or trivalent cations. In addition, solutions with trivalent cations resulted in swell and hydraulic conductivity essentially identical to those obtained with solutions having divalent cations at the same concentration.

Jo et al. (2001) conclude that swell and hydraulic conductivity depend more on valence at intermediate concentrations (0.025 M to 0.1 M), whereas concentration dominates at low (0.005 M) and high (1 M) concentrations. They also conclude that hydraulic conductivity and swelling have a strong inverse relationship, and suggest that swell tests can be used as an indicator of adverse chemical interactions that affect the hydraulic conductivity of GCLs.

### Hydraulic Conductivity to Multispecies Inorganic Solutions

Reeve and Bower (1960) investigated how sodium adsorption ratio (SAR) of the permeant solution and electrolyte concentration affected the hydraulic conductivity of a sodic (sodium rich) soil with a montmorillonitic clay fraction. SAR is a ratio describing the relative amounts of sodium, calcium, and magnesium in the pore water equilibrated with the soil, and can be written as (McBride 1994):

$$SAR = \left[ \frac{Na^+}{[(Ca^{2+} + Mg^{2+})/2]^{1/2}} \right]_e \quad (1)$$

where the cation ( $Na^+$ ,  $Ca^{2+}$ ,  $Mg^{2+}$ ) concentrations are expressed in meq/L (note: 1 meq/L = 1 mN). The soil had a cation exchange capacity (CEC) = 8.9 meq/100 g. The permeant solutions were Salton sea water (SAR = 57) and diluted Salton sea water with SAR = 40, 27.2, 18.2, and 2.2. Reeve and Bower (1960) found that the rate of monovalent for divalent exchange is a function of the divalent cation concentration and SAR of the permeant solution. At a given SAR, solutions with higher ionic strength resulted in more rapid exchange and higher hydraulic conductivity.

McNeal and Coleman (1966) and McNeal et al. (1966) used Na–Ca solutions to investigate how concentration and SAR affect swelling and hydraulic conductivity of Gila clay from New Mexico, USA, which has CEC = 41.2 meq/100 g and consists of 29% montmorillonite. Swelling was quantified as the mass of “bound” solution per mass of clay. Test solutions were prepared with NaCl and  $CaCl_2$  salts at concentrations of 0.8, 0.2, 0.05, 0.012, and 0.003 mN with SAR = 0, 15, 25, 50, 100, and  $\infty$ . Specimens for hydraulic conductivity testing were initially equilibrated by permeation with 10 pore volumes of a 0.8 N solution having the same SAR as the test solution, and then were sequentially permeated with test solutions of decreasing concentration.

McNeal et al. (1966) found no appreciable swell in solutions with SAR = 0 (all divalent) regardless of concentration, which is consistent with the lack of an osmotic swelling phase when the interlayer contains polyvalent cations (Norrish and Quirk 1954). Measurable swelling began at 0.012 N and SAR = 25, and increased as the SAR of the solution increased. Decreases in hydraulic conductivity occurred with decreasing concentration and increasing SAR of the permeant solution. For example, the hydraulic conductivity was  $1.5 \times 10^{-5}$  cm/s for a 0.8 N solution with SAR = 0,  $5.9 \times 10^{-6}$  cm/s for a 0.050 N solution with SAR = 100, and  $1.5 \times 10^{-7}$  cm/s for a 0.012 N solution with SAR =  $\infty$  (all sodium).

McNeal et al. (1966) concluded that salt concentration and SAR affect swelling and hydraulic conductivity of Gila clay in an inverse manner, which was also reported by Jo et al. (2001) for GCLs permeated with single-species solutions. Increasing the concentration or relative abundance of divalent cations (lower SAR) results in less swell and higher hydraulic conductivity. McNeal et al. (1966) postulate that swelling of montmorillonite is the dominant mechanism affecting its hydraulic conductivity because it affects the opening and closing of pores.

Mustafa and Hamid (1975) investigated how electrolyte concentration and SAR of the permeant solution affected the hydraulic conductivity of two montmorillonitic soils, one containing 32% montmorillonite and the other 14% montmorillonite. The hydraulic conductivity of both soils exhibited the same trends with concentration and SAR as reported by McNeal et al. (1966). However, Mustafa and Hamid (1975) indicate that the relationships between swell, hydraulic conductivity, and characteristics of the permeant solution are unique for each soil.

Malik et al. (1992) investigated how mixed Na–Ca solutions of various concentrations affect swelling, dispersion, and flow in two unsaturated clays reported to be montmorillonitic (the montmorillonite content was not reported). NaCl and  $CaCl_2$  solutions with SAR = 0, 5, 15, 25, and 50 and concentrations of 3.1, 12.5, 50, 200, and 500 mM were used. Their results were also similar to those reported by McNeal et al. (1966); swell of both soils increased and the hydraulic conductivity decreased as the concentration decreased or the SAR increased.

## Materials and Methods

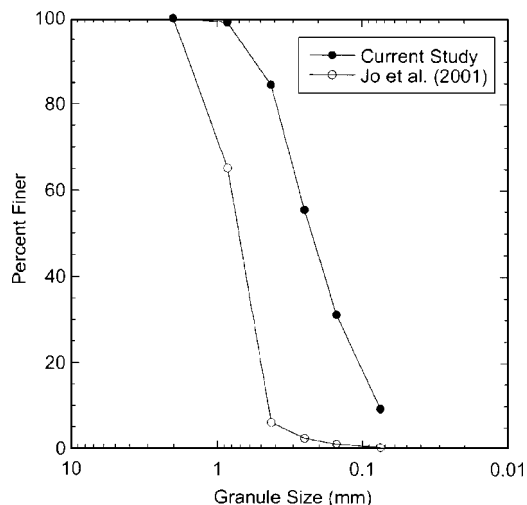
### Geosynthetic Clay Liner

The GCL used in this study contains granular sodium bentonite encapsulated between a 170 g/m<sup>2</sup> slit-film monofilament woven geotextile and a 206 g/m<sup>2</sup> staple-fiber nonwoven geotextile. The geotextiles are bonded by needle-punching fibers that are thermally fused to the geotextiles. The specific gravity of the bentonite is 2.65, and the average mass of bentonite per area is 4.3 kg/m<sup>2</sup>. The initial thickness of the GCL ranges from 5.5 to 6.5 mm, and the average initial gravimetric water content of the bentonite was 9%.

X-ray diffraction showed that the bentonite contains 86% montmorillonite, 3% quartz, 5% tridymite, 3% plagioclase feldspar, 1% K-feldspar, 1% aragonite, 1% illite/mica, and trace amounts of calcite, siderite, clinoptilolite, rutile, and gypsum. The granule size distribution for the GCL (determined by mechanical sieve analysis on the air-dry bentonite) is shown in Fig. 1 along with the granule size distribution for the GCL used by Jo et al. (2001). Both GCLs contain sand-size bentonite granules, but the GCL used in this study has smaller granules.

The CEC and composition of the exchange complex (Ca, Mg, Na, and K) were measured on two samples of bentonite from the GCL using the procedures in *Methods of Soil Analysis* (Spark 1996). Soluble salts were extracted with DI water and exchangeable metals were extracted with ammonium acetate. These replicate measurements yielded CECs of 65.2 and 73.5 meq/100 g and the following exchange complex: Na—56.1 and 40.0 meq/100 g, K—0.6 and 0.8 meq/100 g, Ca—12.0 and 15.7 meq/100 g, Mg—4.0 and 4.8 meq/100 g. Thus, the bentonite used in this study is predominantly Na-montmorillonite.





**Fig. 1.** Granule size distributions for geosynthetic clay liner used in this study and by Jo et al. (2001)

### Permeant Liquids

The multispecies salt solutions were prepared with anhydrous inorganic salts (>96% purity) dissociated in DI water. LiCl and NaCl salts were used to investigate the effects of monovalent cations, and CaCl<sub>2</sub> and MgCl<sub>2</sub> salts were used to investigate the effects of divalent cations. The anionic background (Cl<sup>-</sup>) was held constant for all permeant solutions. Type II DI water was used to prepare the solutions and as the reference solution.

A summary of the solutions used in this study is in Table 1. All of the solutions have near neutral pH (6.6 to 8.5). The parameter RMD in Table 1 represents a ratio of the concentrations of monovalent and divalent cations in the permeant solution. RMD is defined as

$$\text{RMD} = \frac{M_M}{\sqrt{M_D}} \quad (2)$$

where  $M_M$ =total molarity of monovalent cations; and  $M_D$ =total total molarity of divalent cations in the solution. RMD is slightly different from SAR in that RMD characterizes the permeant solution introduced to the soil, whereas SAR generally describes pore water equilibrated with the soil (although SAR has been used to describe solutions by some investigators). RMD also is in terms of molar concentrations (rather than normality), includes all monovalent and divalent cations (SAR is limited to Na, Mg, and Ca) in solution, and does not include a factor of 2 in the denominator (because more than two cations can contribute to  $M_D$ ).

Solutions having ionic strength (I) ranging from 0.05 to 0.5 M and RMD from 0 to ∞ (all divalent to all monovalent) were used as permeant liquids. These solutions were selected to represent the range of ionic strengths and RMDs expected in leachate from modern disposal facilities for municipal solid waste, hazardous wastes, construction and demolition wastes, fly ash, paper sludge, and mine waste. A review of literature pertaining to the composition of leachates from these wastes is included in Kolstad (2000), and is summarized later in this paper. Most of the solutions were Li–Ca mixtures. However, tests were also conducted with Na–Mg and Li–Na–Ca–Mg mixtures to investigate how cation species affected swell and hydraulic conductivity of the GCL.

### Free Swell Tests

Free swell tests were conducted in accordance with ASTM D 5890. Bentonite from the GCL was ground to a fine powder using a mortar and pestle and dry sieved through a No. 200 U.S. standard sieve. The sieved bentonite was air dried for 24 h, and then stored in an airtight container prior to testing. A 100 mL graduated cylinder, accurate to ±0.5 mL, was filled to the 90 mL mark with the test solution. Two grams of sieved bentonite were added to the graduated cylinder in 0.1 g increments. Test solution was then added to the cylinder to reach a final volume of 100 mL by flowing the solution along the cylinder wall so that any particles adhered to the wall would be washed into solution. Swell volume (mL/2 g) was recorded after 24 h, which Jo et al. (2001) report is adequate to establish equilibrium.

### Hydraulic Conductivity Tests

Falling head hydraulic conductivity tests with constant tailwater elevation were conducted on the GCL specimens using flexible-wall permeameters in general accordance with ASTM D 5084 and D 6766. An average hydraulic gradient of 100 and effective stress of 20 kPa were applied. Hydraulic gradients this large are uncommon when testing clay soils, but are common when testing GCLs. Large gradients are acceptable when testing GCLs because the differential in effective stress across a thin specimen is not very sensitive to the hydraulic gradient (Shackelford et al. 2000). Aqueous solutions of the inorganic salts (Table 1) were used as the permeant solutions. Backpressure was not used to permit convenient collection of effluent samples for pH and electrical conductivity (EC) testing.

GCL test specimens were prepared by cutting a sample from a GCL panel using a steel cutting ring (105 mm in diameter) and a sharp utility knife following the method described in Daniel et al. (1997). A small amount of test solution was applied along the inner circumference of the ring using a squirt bottle to prevent bentonite loss when removing the specimen from the trimming ring. Excess geotextile fibers were removed from the edge of the specimen with sharp scissors to eliminate potential preferential flow paths between the GCL and flexible membrane (Petrov et al. 1997). Paste prepared with the test solution and bentonite trimmings was delicately placed along the perimeter of the specimen with a small spatula to minimize the potential for sidewall leakage during permeation.

The initial thickness of the GCL specimen was measured to the nearest 0.1 mm with a caliper. Four measurements were made and the average thickness was recorded. The initial weight of the specimen was measured to the nearest 0.01 g. On completion of the hydraulic conductivity test, the specimen was removed from the permeameter and the final thickness and weight were measured in the same manner.

Sidewall leakage and preferential flow paths along the needle-punched fibers are of concern when permeating GCLs with solutions that alter the hydraulic conductivity of bentonite. When relatively high hydraulic conductivities (>10<sup>-6</sup> cm/s) were obtained, the influent solution was spiked with Rhodamine WT dye (5 mg/L) to stain the flow paths bright red. For all tests that were conducted, the dye tests showed that preferential flow along the needle-punching fibers and the sidewalls did not occur. Jo et al. (2001) report similar findings in their single-species tests on GCLs.

**Table 1.** Summary of Permeant Solutions

Type of solution	Ionic strength (M)	Monovalent concentration $10^{-2}$ (M)	Divalent concentration $10^{-2}$ (M)	RMD ( $\text{mM}^{1/2}$ )	pH	EC (S/m)
Li–Ca	0.05	5.00	0.00	$\infty$	7.4	0.50
		4.35	0.22	0.93	7.6	0.48
		3.33	0.56	0.45	7.7	0.43
		2.00	1.00	0.20	7.2	0.42
		0.00	1.67	0.00	7.8	0.36
Na–Mg		3.33	0.56	0.45	7.4	0.43
Li–Na		3.33	0.56	0.45	7.2	0.43
Ca–Mg		Li(1):Na(3) <sup>a</sup>	Ca(1):Mg(3) <sup>b</sup>			
Li–Ca	0.1	10.0	0.00	$\infty$	6.8	0.88
		8.70	0.44	1.32	7.9	0.87
		8.33	0.56	1.12	8.5	0.88
		7.77	0.77	0.88	8.1	0.87
		6.67	1.11	0.64	7.7	0.86
		5.00	1.67	0.38	7.3	0.80
		2.50	2.50	0.16	7.5	0.77
		1.00	3.00	0.06	7.4	0.72
		0.00	3.33	0.00	7.9	0.70
		8.70	4.35	1.32	6.8	0.87
Na–Mg		6.67	1.11	0.64	6.6	0.86
		1.00	3.00	0.06	7.2	0.73
		8.33	0.56	1.12	7.1	0.88
Li–Na		8.33	0.56	1.12	7.1	0.88
Ca–Mg		Li(3):Na(1) <sup>a</sup>	Ca(3):Mg(1) <sup>b</sup>			
Li–Na		2.50	2.50	0.16	6.5	0.77
Ca–Mg		Li(1):Na(1) <sup>a</sup>	Ca(1):Mg(1) <sup>b</sup>			
Li–Ca	0.2	20.0	0.00	$\infty$	8.1	1.86
		16.7	1.11	1.58	7.2	1.72
		13.3	2.22	0.89	7.1	1.62
		8.00	4.00	0.40	7.2	1.50
		0.00	6.67	0.00	7.2	1.29
Na–Mg		13.3	2.22	0.89	6.7	1.61
Li–Na		8.00	4.00	0.40	7.2	1.50
Ca–Mg		Li(1):Na(3) <sup>a</sup>	Ca(3):Mg(1) <sup>b</sup>			
Li–Ca	0.5	50.0	0.00	$\infty$	8.1	3.45
		38.5	3.85	1.97	7.3	3.46
		31.3	6.25	1.24	8.2	3.29
		20.0	10.0	0.64	8.1	3.03
		0.0	16.7	0.00	7.6	2.74
		31.3	6.25	1.24	7.2	3.30
		38.5	3.85	1.97	6.6	3.46
		Li(3):Na(1) <sup>a</sup>	Ca(1):Mg(3) <sup>b</sup>			
		20.0	10.0	0.64	6.9	3.02
		Li(2):Na(1) <sup>a</sup>	Ca(1):Mg(2) <sup>b</sup>			

Note: RMD=Relative abundance of monovalent and divalent cations; EC=Exchange capacity.

<sup>a</sup>Molar ratio of monovalent cations when two species are present.

<sup>b</sup>Molar ratio of divalent cations when two species are present

The hydraulic conductivity tests were terminated when the termination criteria in ASTM D 5084 and D 6766 were satisfied. The hydraulic conductivity was required to be steady ( $\pm 25\%$  of the mean with no statistically significant trend for at least four values), the ratio of outflow to inflow was between 0.75 and 1.25 for four consecutive values, and the pH and EC of the influent and

effluent deviated less than 10%. A minimum of 2 pore volumes of flow (PVF) was also stipulated, although all tests required more than 2 PVF to satisfy all of the termination criteria (some tests required more than 150 PVF). A pH meter and a portable electrical conductivity probe were used to measure the pH and EC.

**Table 2.** Summary of Free Swell Data

Ionic strength (M)	RMD $\text{mM}^{1/2}$	Free swell ( $\text{mL}/2\text{ g}$ )		
		Li–Ca solutions	Na–Mg solutions	Li–Na–Ca–Mg solutions
0.05	0.93	30.5	—	—
	0.45	24.5	24.5	—
	0.20	22.0	—	—
	0.00	19.0	—	—
0.1	1.32	21.5	21.0	—
	1.12	21.0	—	21.0
	0.88	19.0	—	—
	0.64	17.5	17.5	—
	0.38	14.0	—	—
	0.16	13.5	—	13.0
	0.06	11.5	12.0	—
	0.00	11.0	—	—
0.2	1.67	19.0	—	—
	0.89	15.0	15.5	—
	0.40	12.0	—	12.5
	0.00	9.5	—	—
0.5	1.97	11.5	—	12.0
	1.24	10.5	10.0	—
	0.64	8.5	—	8.5
	0.00	6.5	—	—

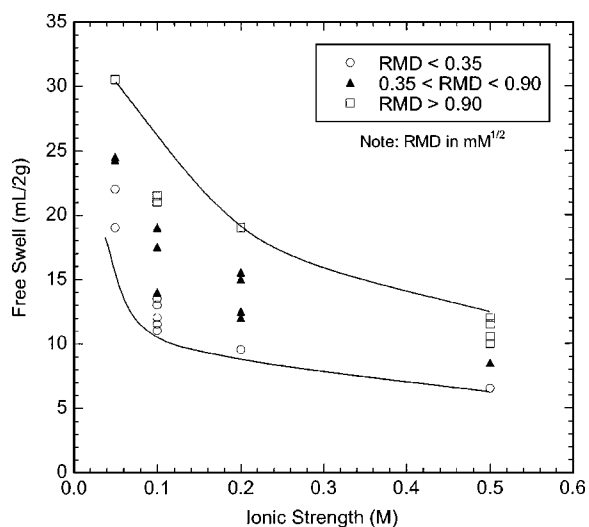
Note: Free Swell in distilled water =  $36.5\text{ mL}/2\text{ g}$ ; RMD = Relative abundance of monovalent and divalent cations.

## Results of Free Swell Tests

### Effect of Concentration and Relative Abundance of Monovalent and Divalent Cations

Free swell tests were conducted using solutions with ionic strengths ranging from 0.05 M to 0.5 M and RMD ranging from 0 to  $1.97\text{ mM}^{1/2}$ . The multispecies solutions were prepared with Li and Ca, Na, and Mg, or Li, Na, Ca, and Mg. Results of the tests are summarized in Table 2.

Free swell is shown as a function of ionic strength in Fig. 2 for



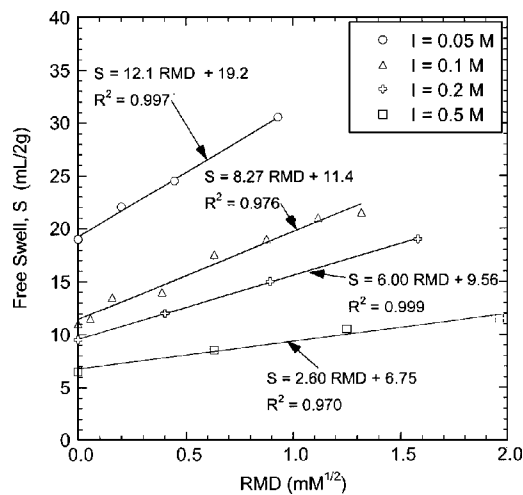
**Fig. 2.** Free swell of geosynthetic clay liner bentonite as a function of ionic strength for low, intermediate, and high relative abundance of monovalent and divalent cation

the Li–Ca solutions. The data are segregated by solutions that are predominantly divalent ( $\text{RMD} < 0.35\text{ mM}^{1/2}$ ), solutions with comparable fractions of monovalent and divalent cations ( $0.35\text{ mM}^{1/2} < \text{RMD} < 0.90\text{ mM}^{1/2}$ ), and solutions that are predominantly monovalent ( $\text{RMD} > 0.90\text{ mM}^{1/2}$ ). Free swell of the bentonite decreases with increasing concentration for each range of RMD. Lower free swell also occurs as the RMD decreases because the presence of more divalent cations suppresses the osmotic component of swelling. RMD also affects the sensitivity to concentration. For the predominantly monovalent solutions ( $\text{RMD} > 0.90\text{ mM}^{1/2}$ ), the free swell decreases  $19\text{ mL}/2\text{ g}$ , on average, as the ionic strength is varied between 0.05 to 0.5 M. For the predominantly divalent solutions ( $\text{RMD} < 0.35\text{ mM}^{1/2}$ ), the free swell decreases  $14\text{ mL}/2\text{ g}$ , on average, over the same range of ionic strengths.

The influence of RMD on swell at constant ionic strength is shown in Fig. 3. The relationships are approximately linear, with trend lines fitted to the data using least-squares linear regression. The slope of each trend line reflects the sensitivity of swell to RMD; the intercept is the free swell when the solution only contains divalent cations. When the ionic strength is lower, the trend lines have a larger slope (e.g., slope = 12.1 for  $I = 0.05\text{ M}$  and 2.6 for  $I = 0.5\text{ M}$ ), which indicates that RMD has a stronger influence on swelling at low ionic strength and less effect at high ionic strength.

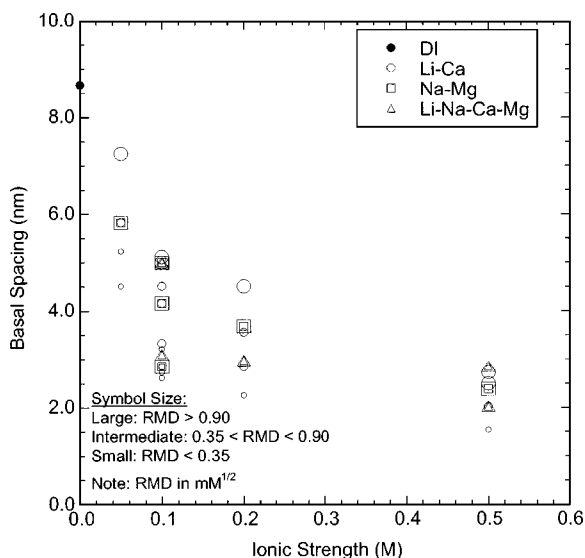
The trends in the free swell tests are consistent with those reported by McNeal et al. (1966) for swelling of Gila clay in mixed Na–Ca solutions. They found a unique relationship between swell and SAR when the concentration was fixed, and that the sensitivity to SAR diminished as the concentration increased. Jo et al. (2001) report similar findings for single species solutions. They found that concentration has a greater effect on free swell for monovalent solutions than divalent solutions.

The sensitivity of free swell to concentration and RMD is

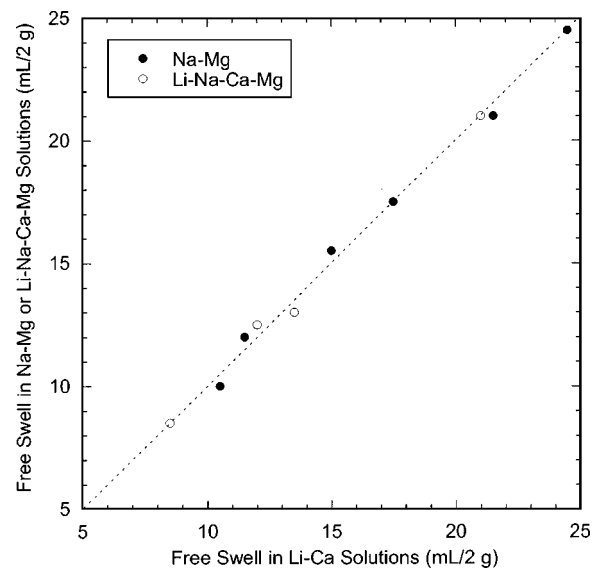


**Fig. 3.** Free swell of GCL bentonite as a function of RMD for ionic strengths of 0.05, 0.1, 0.2 and 0.5 M

caused by expansion and contraction of the interlayer space as a result of crystalline and osmotic swelling. This effect is illustrated in Fig. 4, which shows basal spacing (i.e., sum of interlayer separation distance and thickness of one montmorillonite layer, also referred to as  $d_{001}$ ) as a function of ionic strength. The basal spacing was computed using the method in Smalley (1994), which is based on particle geometry, free swell of the bentonite, the thickness of a montmorillonite layer ( $\approx 0.9$  nm), the basal spacing of Ca-montmorillonite in water ( $\approx 1.96$  nm), and the free swell of Ca-montmorillonite in water ( $\approx 8.0$  mL/2 g). The symbol size in Fig. 4 is proportional to RMD (larger symbols for larger RMD). At high ionic strength (0.5 M), the basal spacing ( $d_{001}$ ) ranges between 1.5 and 2.9 nm, indicating that the swelling is in the crystalline phase ( $d_{001} \leq 1.96$  nm) or the low end of the osmotic phase ( $d_{001} > 1.96$  nm). In contrast, the basal spacing ranges between 4.5 and 8.6 nm at lower concentration ( $I \leq 0.05$  M), which corresponds to crystalline and osmotic swelling ( $d_{001} > 1.96$  nm). Moreover, the smallest symbols (lowest RMD)



**Fig. 4.** Free swell of GCL bentonite as a function of computed basal spacing of montmorillonite



**Fig. 5.** Comparison of free swell of bentonite in Na-Mg and Li-Na-Ca-Mg solutions to free swell in Li-Ca solutions for solutions prepared with the same ionic strength and RMD

often correspond to the lowest  $d_{001}$  for each ionic strength, reflecting suppression of osmotic swelling due to the preponderance of divalent cations.

### Effect of Cation Species

The influence of cation species on free swell is illustrated in Fig. 5 using data from the Li-Ca, Na-Mg, and Li-Na-Ca-Mg solutions. Swell in the Na-Mg and Li-Na-Ca-Mg solutions is essentially equal to the swell in the Li-Ca solutions at the same ionic strength and RMD. No discernable effect of cation species is evident. The tendency of divalent cations to suppress osmotic swelling, combined with the insensitivity of free swell to type of divalent cation species (i.e., as in Jo et al. 2001), probably muted any sensitivity to species for the monovalent cations. The single-species tests by Jo et al. (2001) also show that free swell in monovalent solutions is only slightly sensitive to cations species. Thus, free swell is likely to be insensitive to cation species for most monovalent-divalent cation mixtures.

## Results of Hydraulic Conductivity Tests

### Effect of Concentration and Relative Abundance of Monovalent and Divalent Cations

Hydraulic conductivity tests were conducted using multispecies aqueous solutions listed in Table 1. The ionic strength of the test solutions ranged from 0.05 M to 0.5 M, and the RMD ranged from 0 to  $1.97 \text{ mM}^{1/2}$ . Hydraulic conductivities obtained from these tests are summarized in Table 3.

Hydraulic conductivity is shown in Fig. 6 as a function of ionic strength ( $I$ ). As in Fig. 2, the solutions have been characterized as primarily divalent ( $\text{RMD} < 0.35 \text{ mM}^{1/2}$ ), comparable mixtures ( $0.35 \text{ mM}^{1/2} < \text{RMD} < 0.90 \text{ mM}^{1/2}$ ), and primarily monovalent ( $\text{RMD} > 0.90 \text{ mM}^{1/2}$ ). The hydraulic conductivity is sensitive to the composition of the permeant solution, ranging from  $5.6 \times 10^{-10} \text{ cm/s}$  ( $I=0.05$  M and  $\text{RMD}=0.66 \text{ mM}^{1/2}$ ) to  $1.0 \times 10^{-5} \text{ cm/s}$  ( $I=0.5$  M and  $\text{RMD}=0$ ), and varies exponentially



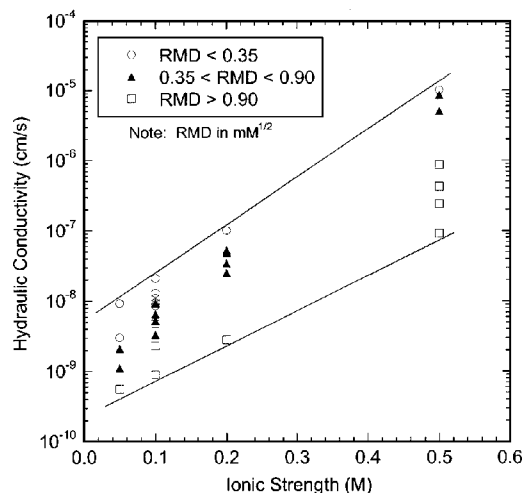
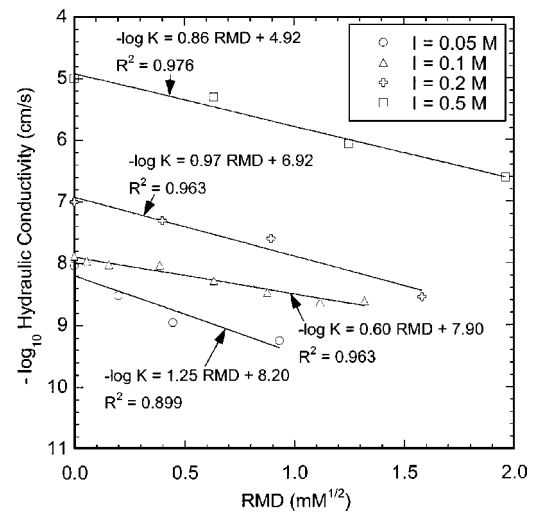
**Table 3.** Summary of Hydraulic Conductivities

Ionic strength (M)	RMD (mM <sup>1/2</sup> )	Hydraulic conductivity (cm/s)		
		Li-Ca solutions	Na-Mg solutions	Li-Na-Ca-Mg solutions
0.05	0.93	$5.6 \times 10^{-10}$	—	—
	0.45	$1.1 \times 10^{-9}$	$2.1 \times 10^{-9}$	—
	0.20	$3.0 \times 10^{-9}$	—	—
	0.00	$9.2 \times 10^{-9}$	—	—
0.1	1.32	$2.4 \times 10^{-9}$	$8.9 \times 10^{-10}$	—
	1.12	$2.3 \times 10^{-9}$	—	$4.8 \times 10^{-9}$
	0.88	$3.3 \times 10^{-9}$	—	—
	0.64	$5.2 \times 10^{-9}$	$6.5 \times 10^{-9}$	—
	0.38	$9.3 \times 10^{-9}$	—	—
	0.16	$9.5 \times 10^{-9}$	—	$2.1 \times 10^{-8}$
	0.06	$1.1 \times 10^{-8}$	$8.5 \times 10^{-9}$	—
	0.00	$1.3 \times 10^{-8}$	—	—
0.2	1.67	$2.8 \times 10^{-9}$	—	—
	0.89	$2.5 \times 10^{-8}$	$3.4 \times 10^{-8}$	—
	0.40	$4.9 \times 10^{-8}$	—	$5.2 \times 10^{-8}$
	0.00	$1.0 \times 10^{-7}$	—	—
0.5	1.97	$2.4 \times 10^{-7}$	—	$9.1 \times 10^{-8}$
	1.24	$8.5 \times 10^{-7}$	$4.2 \times 10^{-7}$	—
	0.64	$5.0 \times 10^{-6}$	—	$8.5 \times 10^{-6}$
	0.00	$1.0 \times 10^{-5}$	—	—

Note: Hydraulic conductivity to distilled water =  $9.0 \times 10^{-10}$  cm/s; RMD = Relative abundance of monovalent and divalent cations.

with ionic strength (linearly on a semilogarithmic graph). The highest hydraulic conductivities at any ionic strength were obtained using the primarily divalent (RMD < 0.35 mM<sup>1/2</sup>) solutions, and the lowest for the primarily monovalent (RMD > 0.90 mM<sup>1/2</sup>) solutions.

The effect of RMD at constant ionic strength is shown in Fig. 7. The base-10 logarithm of hydraulic conductivity ( $\log_{10}K$ ) is approximately linearly related to RMD. The trend lines relating

**Fig. 6.** Hydraulic conductivity of GCL as a function of solution ionic strength for low, intermediate, and high RMD**Fig. 7.** Hydraulic conductivity of GCL as a function of RMD for ionic strengths of 0.05, 0.1, 0.2, and 0.5 M

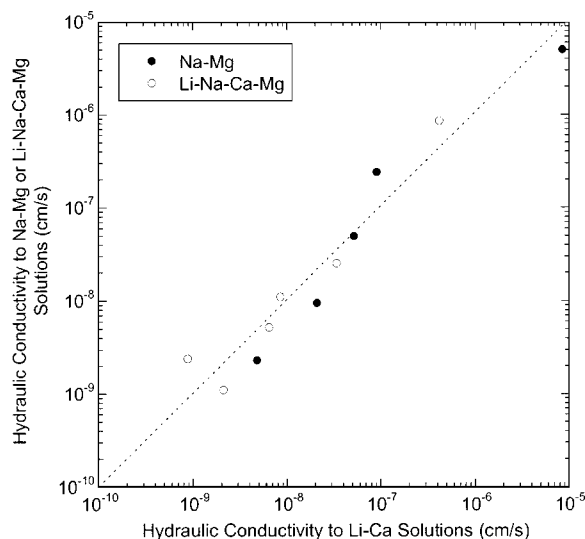
$\log_{10}K$  and RMD in Fig. 7 were fit using least-squares linear regression. Their slope describes the sensitivity of hydraulic conductivity to RMD, and the intercept is the hydraulic conductivity to the divalent solution. The hydraulic conductivity exhibits greater sensitivity to RMD at lower ionic strength (i.e., slope = 1.25 at  $I=0.05$  M and 0.86 at  $I=0.5$  M). The data for the tests conducted at an ionic strength of 0.1 M are an exception to the trend. The reason for this deviation is unknown.

The trends in Figs. 6 and 7 are comparable to the trends reported by McNeal and Coleman (1966) for Gila clay. They found that the hydraulic conductivity increases with increasing concentration and decreasing SAR, and distinct curves relating hydraulic conductivity to SAR exists when the concentration is fixed. McNeal and Coleman (1966) report that SAR has a stronger influence on hydraulic conductivity at low concentrations, and that the effect of SAR diminishes at high concentrations. Jo et al. (2001) also report similar sensitivity to ionic strength and cation valence for single species solutions. At a given ionic strength, the highest hydraulic conductivities were obtained with divalent or trivalent solutions, and the lowest with monovalent solutions.

A diminished effect of ionic strength and RMD probably would have been observed had much lower or much higher ionic strengths been used. For example, DI water is the limiting case for dilute solutions (in this study, the hydraulic conductivity of the GCL to DI water was  $9.0 \times 10^{-10}$  cm/s). In addition, Jo et al. (2001) report that the hydraulic conductivity of the GCL they tested leveled off between  $10^{-5}$  to  $10^{-4}$  cm/s for ionic strengths greater than 1 M. When the ionic strength is high, osmotic swelling becomes negligible, and the basal spacing is reduced to its smallest value in the hydrated state ( $\approx 2$  nm). Once this compressed condition is reached, no further increase in hydraulic conductivity can occur. In fact, a decrease in hydraulic conductivity is possible due to the higher viscosity of concentrated solutions (Fernandez and Quigley 1988).

### Effect of Cation Species

Li-Ca, Na-Mg, and Li-Na-Ca-Mg solutions having various RMD and ionic strengths were used to investigate how differences in cation species affect the hydraulic conductivity of GCLs



**Fig. 8.** Comparison of hydraulic conductivities of GCL obtained with Na-Mg and Li-Na-Ca-Mg solutions to hydraulic conductivities obtained with Li-Ca solutions for solutions prepared with the same ionic strength and RMD

permeated with mixed solutions. Composition of each solution is summarized in Table 1 and the hydraulic conductivities are in Table 3.

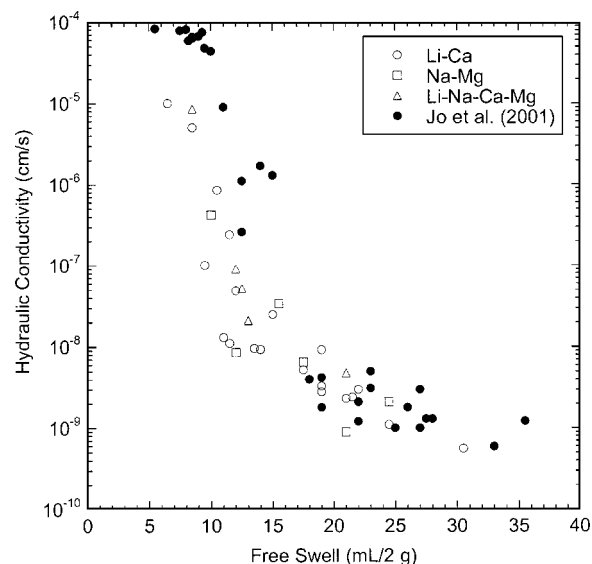
Hydraulic conductivities obtained using the Li-Ca solutions are compared with those obtained from the Na-Mg and Li-Na-Ca-Mg solutions in Fig. 8. At comparable ionic strengths and RMD, essentially the same hydraulic conductivities were obtained with the Na-Mg and Li-Na-Ca-Mg solutions as with the Li-Ca solutions. No discernable effect of cation species is apparent. The insensitivity of hydraulic conductivity to cation species is analogous to the insensitivity of free swell to cation species. Differences in preference of the montmorillonite for Ca over Mg and Na over Li appear to have a small effect compared to the effects of RMD and concentration. In addition, Jo et al. (2001) found that the hydraulic conductivity was insensitive to cation species for a given valence.

The insensitivity to cation species evident in Fig. 8, combined with the insensitivity to cation species observed by Jo et al. (2001) for single-species solutions, suggests that the hydraulic conductivity at fixed RMD is likely to be insensitive to cation species in most monovalent-divalent mixtures. Moreover, Jo et al. (2001) found that permeation with single-species solutions containing divalent and trivalent cations yielded essentially the same hydraulic conductivity at a given concentration. Thus, the insensitivity to cation species may extend to multispecies solutions in general, with ionic strength and RMD being the dominant variables controlling hydraulic conductivity. In this case, the denominator of RMD would include the total normality of the polyvalent (valence  $\geq +2$ ) cations in the solution. While this hypothesis is plausible, more testing is needed to confirm its validity.

## Practical Implications

### Free Swell and Hydraulic Conductivity

Jo et al. (2001) show that a strong relationship exists between free swell of bentonite and the hydraulic conductivity of GCLs ex-



**Fig. 9.** Hydraulic conductivity of GCL as a function of free swell of bentonite. Test data are from this study and from Jo et al. (2001)

posed to single-species solutions. A similar relationship could be expected for multispecies solutions as well because Figs. 2, 3, 6, and 7 show that ionic strength and RMD affect swell and hydraulic conductivity in a consistent and similar manner. McNeal et al. (1966) also report a strong correlation between swelling and hydraulic conductivity for Gila clay permeated with solutions having different ionic strengths and SAR.

Hydraulic conductivity of the GCL specimens permeated with the multispecies solutions is shown in Fig. 9 as a function of free swell along with the single-species data from Jo et al. (2001). A strong relationship exists between hydraulic conductivity and free swell for both data sets. The slight offset in the two data sets at lower swell volumes (and higher hydraulic conductivities) is most likely due to differences in the granule size distributions of the bentonites and not the use of multispecies versus single species solutions. The GCLs used in both studies were essentially identical, except the bentonite in the GCL used in this study has smaller granules than the bentonite in the GCL used by Jo et al. (2001) (Fig. 1). Mesri and Olson (1971) and McNeal et al. (1966) indicate that bentonites with larger “domains” (quasi-crystals) permit larger flow paths and higher hydraulic conductivity. In addition, Katsumi et al. (2002) show that nonprehydrated GCLs containing bentonite with larger granules are more permeable than GCLs with smaller granules when permeated using stronger ( $\geq 0.2$  M) salt solutions. Because the granules do not swell appreciably in strong solutions, bentonites with larger granules have larger intergranular pores, and higher hydraulic conductivity. That is, the hydraulic conductivity of granular bentonite permeated with strong solutions follows a similar relationship with particle size as do granular soils; i.e., the hydraulic conductivity increases as the particle size increases, all factors being equal (e.g., Lambe and Whitman 1969; Terzaghi et al. 1996). In contrast, granule size has no effect on free swell, because the bentonite is crushed to pass the No. 200 sieve prior to free swell testing.

McNeal et al. (1966) conclude that swelling of expansive minerals such as montmorillonite is the dominant mechanism affecting the hydraulic conductivity. The results of this study, as well as those in Jo et al. (2001), support this conclusion. The trends shown in Fig. 9 also indicate that free swell tests can be a relatively simple and quick screening method to evaluate the compat-

ibility of GCLs permeated with inorganic salt solutions containing mixtures of cations. Although not a surrogate for chemical compatibility testing (direct testing is needed to demonstrate that a GCL is compatible with a liquid), free swell testing can be used to identify liquids that are incompatible with GCLs. The data in Fig. 9 also illustrate that the relationship between hydraulic conductivity and free swell is bentonite specific, and needs to be identified empirically.

### Estimating Hydraulic Conductivity

The approximately linear trends shown in Figs. 6 and 7 suggest that a relatively simple empirical model can be used to estimate hydraulic conductivity of GCLs as a function of ionic strength and RMD. A model relating these parameters was developed using stepwise regression (Draper and Smith 1998) using a significance level of 0.05:

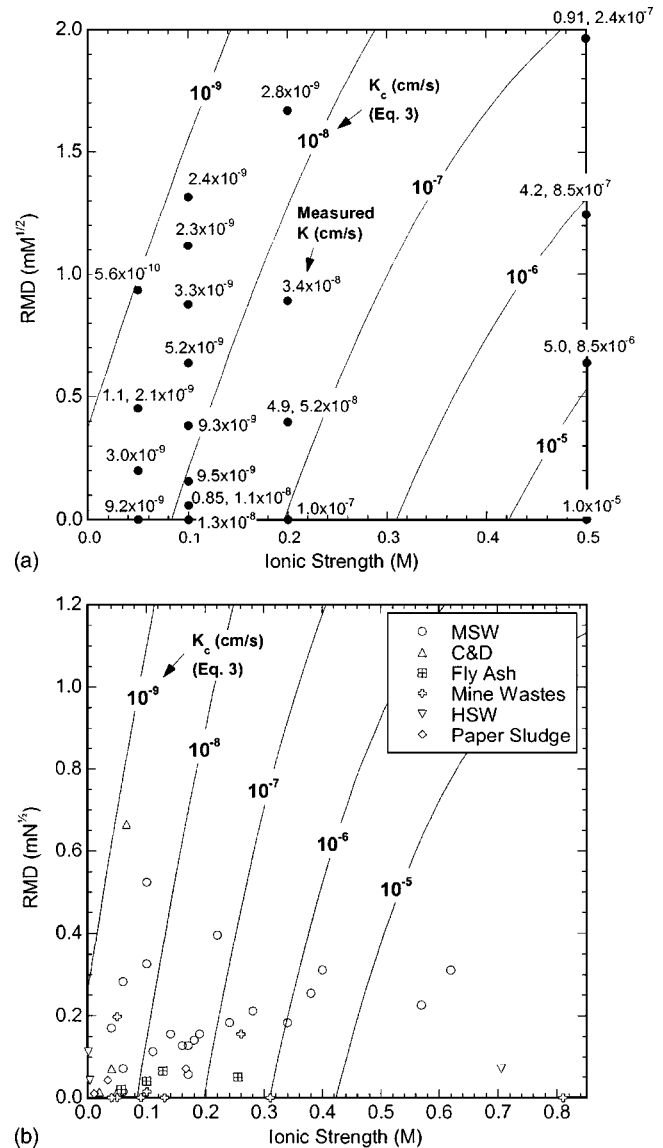
$$\frac{\log K_c}{\log K_{DI}} = 1.085 - 1.097I + 0.0398I^2 \text{ RMD} \quad (3)$$

In Eq. 3,  $K_c$ =hydraulic conductivity to the inorganic chemical solution and  $K_{DI}$ =hydraulic conductivity to deionized water. The  $R^2$  for Eq. (3) is 0.967 and the  $p$  statistic is less than 0.0001. Eq. (3) is linear in both  $I$  and RMD, and the product  $I^2 \times \text{RMD}$  reflects that the sensitivity to RMD varies nonlinearly with ionic strength (e.g., as in Fig. 7). Eq. (3) is valid for  $I=0.05\text{--}0.5$  M and  $\text{RMD} < 2.0 \text{ mM}^{1/2}$ .

Eq. (3) is based on data from the GCL tested in this study under the state of stress that was employed (effective stress = 20 kPa). However, Eq. (3) can be used to estimate how inorganic solutions may affect the hydraulic conductivity of other GCLs provided they employ granular Na-bentonite consisting of approximately 80% montmorillonite. Many of the GCLs used in North America today fit this description. Even if the granule size or montmorillonite content differs from those in this study, the relative effects of ionic strength and RMD should be approximately correct. In addition, Petrov and Rowe (1997) show that the hydraulic conductivity of GCLs exhibits similar sensitivity to effective stress regardless of whether DI water or a salt solution is used as the permeant liquid. Thus, Eq. (3) can be used to estimate the hydraulic conductivity at different effective stresses if the hydraulic conductivity to DI water at these stresses is known.

A comparison of  $K_c$  predicted with Eq. (3) and the measured hydraulic conductivity is shown in Fig. 10(a). The contour lines in Fig. 10 correspond to Eq. (3), whereas the data points correspond to the  $I$  and RMD for the tests conducted in this study. Eq. (3) captures the data reasonably well. Hydraulic conductivities predicted with Eq. (3) are also shown as contours in Fig. 10(b) along with points corresponding to  $I$  and RMD for actual leachates from a variety of wastes and solid waste disposal facilities reviewed by Kolstad (2000). The ionic strength and RMD of each leachate is summarized in Table 4, along with the data source (literature and regulatory agency reports) and the type of containment facility. The points and contour lines in Fig. 10(b) illustrate what hydraulic conductivity likely would have been had the GCL used in this study been tested with these leachates.

Of the 50 points shown in Fig. 10(b), 37 fall below  $10^{-7} \text{ cm/s}$  (74%) and 24 fall below  $10^{-8} \text{ cm/s}$  (48%). Thus, GCLs with high hydraulic conductivities ( $>10^{-7} \text{ cm/s}$ ) should not be common in bottom liners where leachates similar to those in Table 4 are likely to be found. Moreover, many of the points in Fig. 10(b) associated with high hydraulic conductivities correspond to "young" (landfill age  $<5$  yr) municipal solid waste (MSW)



**Fig. 10.** Contours of hydraulic conductivity as a function of RMD and ionic strength predicted with Eq. (3) along with (a) measured hydraulic conductivities as solid circles and (b) points corresponding to ionic strength and RMD of various leachates. Data from Williams (1975) ( $I=1.87$  M and  $\text{RMD}=0$ ) and Kolstad (2000) ( $I=1.37$  M and  $\text{RMD}=2.52 \text{ mM}^{1/2}$ ) are off the scale in (b).

leachates (Table 4). The composition of MSW leachate changes over time, and thus high hydraulic conductivities may not be realized because of the relatively long time required for a GCL and leachate to reach equilibrium under field conditions (Jo 2003). However, some of the points for mine waste, paper sludge, and fly ash disposal facilities are associated with high hydraulic conductivities, and the composition of leachates from these wastes can be persistent.

### Effect of Prehydration

The results of this study pertain specifically to nonprehydrated GCLs. Different results may have been obtained had the GCLs been completely prehydrated by permeation with DI or potable water for several pore volumes of flow. Comparisons between hydraulic conductivities of nonprehydrated and completely prehy-

**Table 4.** Summary of Ionic Strength and RMD of Various Leachates

Source	Leachate type	Ionic strength (M)	RMD (mM <sup>1/2</sup> )
Ehrig (1983)	Young MSW leachate (<5 yr)	0.28	0.21
Pohland (1980)		0.38	0.25
Tchbanoglous et al. (1993)		0.14	0.16
Chian and DeWalle (1976)		0.17	0.13
Cheremisinoff (1983)		0.22	0.40
Alker et al. (1995)		0.16	0.13
Chian and DeWalle (1975)		0.57	0.23
Chian and DeWalle (1975)		0.62	0.31
Chian and DeWalle (1975)		0.34	0.18
Farquhar (1989)		0.40	0.31
Shams et al. (1994)		0.24	0.18
Ehrig (1983)	Intermediate MSW leachate (5–10 yr)	0.10	0.52
Pohland (1980)		0.10	0.33
Chian and DeWalle (1975)		0.17	0.06
Chian and DeWalle (1975)		0.11	0.11
Farquhar (1989)		0.18	0.14
Chian and DeWalle (1975)	Old MSW leachate (>10 yr)	0.06	0.01
Chian and DeWalle (1975)		0.06	0.28
Farquhar (1989)		0.06	0.07
Alker et al. (1995)		0.04	0.17
Kmet and McGinley (1982)	MSW leachate	0.19	0.16
Ruhl and Daniel (1997)		0.04	0.17
Kolstad (2000)	C and D leachate	0.05	0.01
Kolstad (2000)		0.02	0.01
WMNA (1993)		0.04	0.07
Weber et al. (2002)		0.066	0.66
Kolstad (2000)	Fly ash leachate	0.03	0.25
Kolstad (2000)		0.03	0.06
Kolstad (2000)		0.02	0.10
Kolstad (2000)		0.05	0.23
Kolstad (2000)		0.06	0.13
Kolstad (2000)		0.07	0.20
Kolstad (2000)		0.37	2.52
Kolstad (2000)		0.76	0.71
Al et al. (1994)	Mine process water	0.05	0.00
Shackelford (1998)		0.04	0.00
Jordan et al. (1998)		0.05	0.20



**Table 4.** (Continued.)

Source	Leachate type	Ionic strength (M)	RMD ( $\text{mM}^{1/2}$ )
Williams (1975)	Acidic mine waste drainage	1.87	0.00
Christensen and Laake (1996)		0.13	0.00
Christensen and Laake (1996)		0.09	0.00
Al et al. (1994)		0.81	0.00
Shackelford (1998)		0.31	0.00
Williams (1975)	Pyritic tailings leachate	0.26	0.16
Pettit and Scharer (1999)	Ur rock drainage	0.10	0.01
Kolstad (2000)	Hazardous waste leachate	0.001	0.11
		0.70	0.07
		0.003	0.04
Kolstad (2000)	Paper mill landfill leachate	0.17	0.07
		0.01	0.01
		0.03	0.04

Note: RMD=Relative abundance of monovalent and divalent cations; MSW=Municipal solid waste.

drated GCLs that have been permeated long enough to establish chemical equilibrium between the bentonite solid and the solution show that prehydration by permeation with DI water results in hydraulic conductivities an order of magnitude lower than those obtained without prehydration, even if cation exchange between the mineral surface and the permeant liquid is complete (Petrov and Rowe 1997; Jo et al. 2004).

Although the effect of complete prehydration is significant, complete prehydration is unlikely to occur in the field. Most GCLs in field applications hydrate as water is drawn to the bentonite from an underlying subgrade via vapor diffusion or gradients in matric potential (Daniel et al. 1993, 1998; Katsumi et al. 2003). The prehydration afforded by these processes does not appear to have the same effect as complete prehydration by direct permeation. Vasko et al. (2001) found that the hydraulic conductivity of GCLs prehydrated with DI water via capillary wetting and vapor diffusion and permeated with  $\text{CaCl}_2$  solutions had essentially the same hydraulic conductivity as nonprehydrated GCLs unless the solution was very strong (concentration  $>0.1$  M). Comparable findings are reported by Katsumi et al. (2003). These observations suggest that hydraulic conductivities reported in this study are likely to be more representative of most field conditions than hydraulic conductivities of completely prehydrated GCLs.

## Summary and Conclusions

This study dealt with the influence of multispecies inorganic salt solutions on swelling and hydraulic conductivity of nonprehydrated GCLs. Free swell and hydraulic conductivity tests were conducted on nonprehydrated specimens of a commercially available GCL using DI water and aqueous solutions of  $\text{LiCl}$ ,  $\text{NaCl}$ ,  $\text{CaCl}_2$ , and  $\text{MgCl}_2$  salts. The relative amounts of monovalent and

divalent cations in solution were quantified with the parameter RMD, which is the ratio of the total molarity of monovalent cations to the square root of the total molarity of divalent cations.

Results of the free swell tests show that swell is directly related to RMD and inversely related to ionic strength. RMD has a strong effect on swell in weaker solutions, and a modest effect in strong solutions. Similar findings were obtained from the hydraulic conductivity tests. Hydraulic conductivity was found to be directly related to ionic strength and inversely related to RMD, with RMD having a greater effect on hydraulic conductivity in weaker solutions. Tests were also conducted to determine if cation species affects swell or hydraulic conductivity. No discernable effect of cation species was evident in the free swell or hydraulic conductivity for tests conducted at a given ionic strength and RMD.

A strong relationship between hydraulic conductivity and free swell was found that is analogous to the relationship reported by Jo et al. (2001) for tests conducted using single-species salt solutions. However, the hydraulic conductivity-free swell relationship is not unique, and must be defined empirically for a particular bentonite if free swell tests are to be used for chemical compatibility screening.

The hydraulic conductivity data were also used to develop a regression model relating hydraulic conductivity of the GCL to ionic strength and RMD of the permeant solution. Predictions made with the model indicate that high hydraulic conductivities (i.e.,  $>10^{-7}$  cm/s) are unlikely for nonprehydrated GCLs in base liners in many solid waste containment facilities. However, for some wastes that transmit stronger leachates or leachates that are dominated by polyvalent cations (e.g., fly ash, paper sludge, and mine wastes), high hydraulic conductivities may be realized provided adequate time exists for the bentonite and leachate to reach chemical equilibrium.

## Acknowledgments

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# Errata for “Hydraulic Conductivity and Swell of Nonprehydrated Geosynthetic Clay Liners Permeated with Multispecies Inorganic Solutions” by D. Kolstad, C. Benson, and T. Edil

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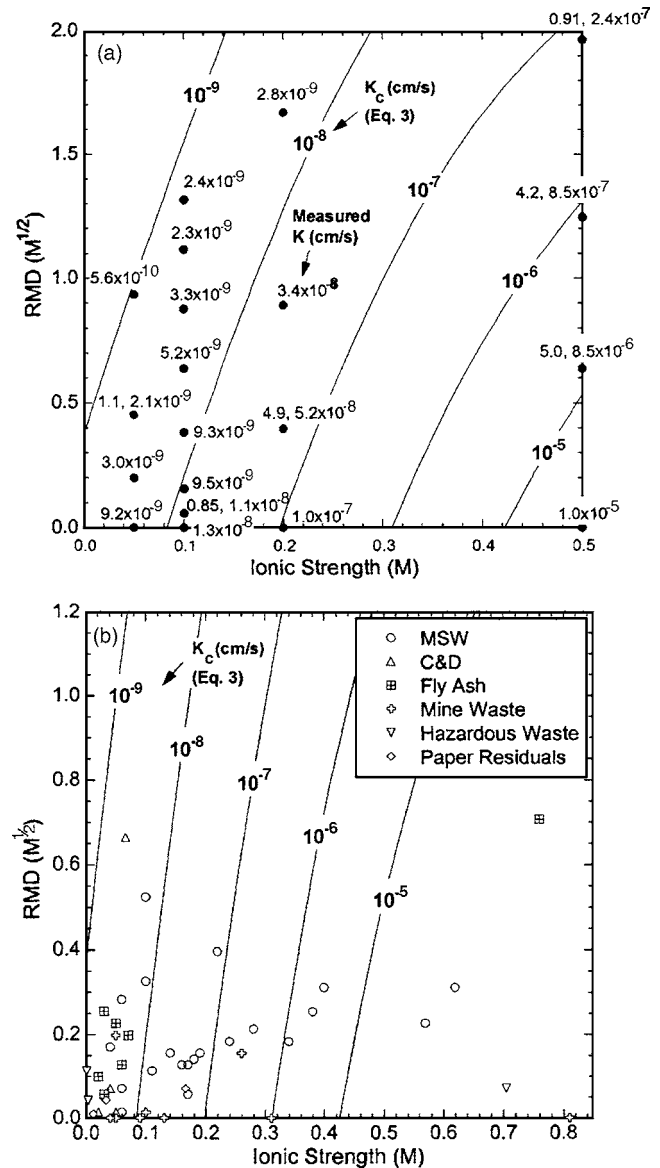
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The following corrections should be noted.

The units for RMD in the text and graphs in this paper should be  $M^{1/2}$  rather than the units of  $mM^{1/2}$  shown in the published version. Eq. (3) also contained typographical errors. The correct version is

$$\frac{\log K_c}{\log K_{DI}} = 0.965 - 0.976I + 0.0797RMD + 0.251I^2RMD \quad (3)$$

In addition, the plotting positions for the fly ash leachates were reversed in Fig. 10(b) (i.e., the fly ash leachate data were plotted as  $I$  versus RMD instead of RMD versus  $I$ ). A correct version of Fig. 10(b) is presented here. These errors do not affect any of the conclusions or inferences in the paper.



**Fig. 10.** Contours of hydraulic conductivity as a function of RMD and ionic strength predicted with Eq. (3) along with (a) measured hydraulic conductivities as solid circles and (b) points corresponding to ionic strength and RMD of various leachates. Data from Williams (1975) ( $I=1.87$  M and  $RMD=0$ ) and Kolstad (2000) ( $I=1.37$  M and  $RMD=2.52M^{1/2}$ ) are off the scale in (b).