

## CONCENTRATION DEPENDENCY OF THE PREHYDRATION EFFECT FOR A GEOSYNTHETIC CLAY LINER

JAE-MYUNG LEE<sup>i)</sup> and CHARLES D. SHACKELFORD<sup>ii)</sup>

### ABSTRACT

Both prehydrated and non-prehydrated specimens of a geosynthetic clay liner (GCL) are permeated with chemical solutions containing 5, 10, 20, 50, and 100 mM calcium chloride ( $\text{CaCl}_2$ ) to determine the concentration dependency of the prehydration effect on the hydraulic conductivity ( $k$ ) of the GCL. The tests for  $k$  are not terminated before chemical equilibrium between the influent and effluent solutions has been established, resulting in testing durations ranging from 14 hrs to 502 days, depending on the  $\text{CaCl}_2$  concentration and whether or not the specimen was prehydrated. The results are presented in terms of the ratio of  $k$  for a non-prehydrated specimen ( $k_{NP}$ ) relative to  $k$  for a separate but identical prehydrated specimen ( $k_P$ ) permeated with the same chemical solution. Prehydration is shown to have little, if any, effect on the hydraulic conductivity of the GCL for specimens permeated with solutions containing from 5 to 50 mM  $\text{CaCl}_2$  (i.e.,  $0.5 \leq k_{NP}/k_P \leq 1.1$ ), whereas the effect of prehydration for specimens of the GCL permeated with the 100 mM  $\text{CaCl}_2$  solution is more significant (i.e.,  $k_{NP}/k_P \approx 3.0$ ). The results provide evidence that the previously reported prehydration effect is concentration dependent, and may be insignificant when the permeant liquid contains relatively low concentrations of inorganic solutes, i.e., provided permeation is continued until chemical equilibrium is achieved.

**Key words:** compatibility, first exposure effect, geosynthetic clay liners, hydraulic conductivity, hydraulic performance, inorganic chemicals, prehydration effect (IGC: D4/D5/H11/M9)

### INTRODUCTION

Several investigators have reported that prehydration of geosynthetic clay liners (GCLs) or soil mixtures containing bentonite with water prior to permeation with actual permeant liquids can have a significant effect on hydraulic conductivity (e.g., Shan and Daniel, 1991; Daniel et al., 1993; Shackelford, 1994; Didier and Comeaga, 1997; Gleason et al., 1997; Kajita, 1997; Petrov and Rowe, 1997; Petrov et al., 1997a; Quaranta et al., 1997; Ruhl and Daniel, 1997; Stern and Shackelford, 1998; Lin and Benson, 2000; Shackelford et al., 2000; Vasko et al., 2001; Ashmawy et al., 2002; Shan and Lai, 2002). The potential impact of this effect, commonly referred to as the prehydration or the first exposure effect, on hydraulic conductivity is illustrated schematically in Fig. 1.

As shown in Fig. 1, permeation of a prehydrated specimen with a chemical solution can result in an increase in the hydraulic conductivity relative to that based on initial permeation with water (i.e.,  $k_P > k_w$ ) due to physicochemical interactions between the soil and the waste liquid, but the resulting hydraulic conductivity is lower than that based on permeation of a separate, but identical, non-prehydrated specimen (i.e.,  $k_P < k_{NP}$ ). As a result, the

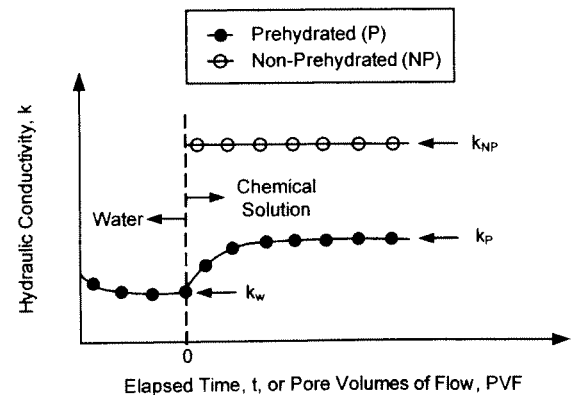


Fig. 1. Schematic illustration of typical results obtained from hydraulic conductivity tests on both prehydrated and non-prehydrated specimens permeated with chemical solutions (after Shackelford, 1994)

hydraulic conductivity of GCLs or soil mixtures containing bentonite permeated directly with chemical solutions typically has been found to be significantly higher than that for the same materials permeated with the same chemical solutions after prehydration with water.

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For example, Shackelford (1994) reported the results of two tests involving permeation of identical specimens of compacted sand-bentonite mixtures containing 16% sodium bentonite by dry weight ( $w/w$ ) with a calcium saturated tailings solution. The first test involved permeation with water resulting in a hydraulic conductivity ( $k_w$ ) of  $3.4 \times 10^{-8}$  cm/s, followed by permeation with the saturated calcium solution resulting in a hydraulic conductivity ( $k_p$ ) of  $1.7 \times 10^{-7}$  cm/s. The second test involved permeation of an identical, but separate, sand-bentonite specimen directly with the calcium solution (i.e., without prehydration), resulting in a hydraulic conductivity ( $k_{NP}$ ) of  $2.3 \times 10^{-5}$  cm/s. Shackelford (1994) noted that, while permeation of the prehydrated specimen resulted in an 5-fold increase in the hydraulic conductivity of the specimen relative to permeation with water (i.e.,  $k_p/k_w = 5$ ), the effect of not prehydrating the specimen was more substantial, resulting in a hydraulic conductivity that is 135 times higher than that for the prehydrated specimen (i.e.,  $k_{NP}/k_p = 135$ ).

However, some results with respect to the potential significance, if any, of prehydration reported in the literature are conflicting. For example, Shan and Daniel (1991) and Rad et al. (1994) found that the hydraulic conductivities of prehydrated GCL specimens permeated with pure ethanol or methanol were similar to the hydraulic conductivities based on permeation with water. In contrast, Petrov et al. (1997b) found that the hydraulic conductivities of prehydrated GCL specimens permeated with pure ethanol were significantly higher (i.e.,  $> 10X$ ) than those based on permeation with water when the tests were continued until chemical equilibrium between the influent and effluent had been achieved. Petrov et al. (1997b) also suggested that the hydraulic conductivities reported by Shan and Daniel (1991) and Rad et al. (1994) probably would have been higher had the tests been continued until chemical equilibrium was achieved.

Gleason et al. (1997) found that the hydraulic conductivity of a prehydrated specimen of calcium bentonite (i.e.,  $7 \times 10^{-9}$  cm/s) permeated with a 250 mM calcium chloride ( $\text{CaCl}_2$ ) solution was approximately half an order-of-magnitude lower than the hydraulic conductivity of a non-prehydrated calcium-bentonite specimen (i.e.,  $3 \times 10^{-8}$  cm/s) permeated with the same solution. However, prehydration of compacted sand-bentonite mixtures containing 5% ( $w/w$ ) sodium bentonite had little effect on the hydraulic conductivity (i.e.,  $10^{-6}$  cm/s) to the same solution. This lack of agreement between the two results may have been due, in part, to the difference in specimen composition (i.e., sand-bentonite mixtures vs. pure bentonite) and/or the types of primary exchangeable cation (i.e., sodium vs. calcium).

In the field, prehydration may occur naturally, such as by precipitation or absorption of water from adjacent soils (Daniel et al., 1993; Bonaparte et al., 1996; Petrov and Rowe, 1997), or may be imposed either by spraying water (Vasko et al., 2001) or, in the case of GCLs, by installing commercially available prehydrated GCLs (e.g., see Shackelford et al., 2000). For laboratory experi-

ments, prehydration typically has been imposed by spraying or soaking the specimens with water (e.g., Daniel et al., 1993; Didier and Comeaga, 1997; Vasko et al., 2001), or more typically by permeating the specimens with water (e.g., Shan and Daniel, 1991; Shackelford, 1994; Bonaparte et al., 1996; Gleason et al., 1997; Petrov and Rowe, 1997; Petrov et al., 1997a,b; Ruhl and Daniel, 1997; Stern and Shackelford, 1998; Shackelford et al., 2000).

The degree of prehydration may be complete or partial, depending on the degree of saturation upon prehydration (e.g., Daniel et al., 1993; Vasko et al., 2001). For example, Daniel et al. (1993) found that the degree of prehydration is not particularly important in terms of the hydraulic conductivity of GCLs permeated with different organic liquids (i.e., benzene, gasoline, methanol, methyl tertiary-butyl ether [MTBE], and trichloroethylene [TCE]), provided the gravimetric water content of the bentonite after prehydration is 100% or greater. However, the tests performed by Daniel et al. (1993) were relatively short-term tests (i.e.,  $< 2$  pore volumes of flow and  $< 2$  months in duration), with no evidence of chemical equilibrium between the influent and effluent having been established. Similarly, Vasko et al. (2001) also found that the degree of prehydration had no apparent effect on the hydraulic conductivity of GCLs permeated with weak to intermediate strength  $\text{CaCl}_2$  solutions (i.e.,  $\leq 25$  mM). However, the hydraulic conductivity of GCL specimens permeated with strong  $\text{CaCl}_2$  solutions (i.e.,  $\geq 100$  mM  $\text{CaCl}_2$ ) decreased from approximately  $1 \times 10^{-4}$  cm/s to  $3 \times 10^{-7}$  cm/s as the prehydration water content increased from 9% (air-dried) to 150%, and no further increase in hydraulic conductivity occurred with further increase in the prehydration water content above 150%. In contrast to the study by Daniel et al. (1993), the tests performed by Vasko et al. (2001) were continued until the pH and electrical conductivity (EC) of the effluent was within  $\pm 10\%$  of that for the influent, as recommended by Shackelford et al. (1999).

Due to the potential importance of prehydration on the hydraulic performance of bentonite-based barrier materials, such as GCLs, and the apparent inconsistencies regarding the effect of prehydration on hydraulic conductivity, the primary objective of this study is to evaluate the concentration dependency of the prehydration effect on the hydraulic performance of a GCL permeated with inorganic solutions containing a wide range of  $\text{CaCl}_2$  concentrations. In order to achieve this objective, hydraulic conductivity tests are performed on both prehydrated and non-prehydrated specimens of the same GCL until chemical equilibrium between influent and effluent is established. In some cases when relatively weak solutions were used (i.e., 5, and 10 mM  $\text{CaCl}_2$ ), establishing chemical equilibrium required on the order of a year or more of permeation with the  $\text{CaCl}_2$  solution. The scope of this study is similar to that of Vasko et al. (2001), except the prehydrated specimens in this study are fully prehydrated by permeation with water under confinement (23.5 kPa), whereas the prehydrated speci-

mens tested specific gravity diffusion (1 little (8 kPa; this study method of on the confinement effect for a

## MATERIALS

### Geosynthetic

The geosynthetic consists of a woven geotextile and a GCL was 6 mm thick (average grain size (ASTM D 4753) respectively GCL was 5 mm thick (20% ( $w/w$ ) fine aggregate ( $w/w$ ) of the hydrometer limit and plasticity are 430 and 10, respectively as a high plasticity. The mineral morillonite content is 61.7 meq/100 g, 18.6% calcium, 1.1% potassium, and the chemical composition of bentonite is

### Permeant

The permeant was tap water. Barnstead® electrical conductivity solutions containing calcium chloride (Type IV) and Calcium chloride. Calcium chloride studies involving barrier materials (figures) with the solution materials (Shackelford, 1997; James, 1997; Ruhl, Benson, 2000; Jo et al., 2000).

The  $\text{CaCl}_2$  (powdered, analytical grade, MO) in the

mens tested by Vasko et al. (2001) were prehydrated at specific gravimetric water contents ( $w$ ) either by vapor diffusion ( $w \leq 50\%$ ) or by wicking ( $w > 50\%$ ) with no or little (8 kPa) confinement. Thus, a secondary objective of this study is to evaluate the potential influence of the method of prehydration and the degree of confinement on the concentration dependency of the prehydration effect for a GCL.

## MATERIALS AND METHODS

### *Geosynthetic Clay Liner*

The geosynthetic clay liner (GCL) used in this study consists of a layer of granular sodium bentonite sandwiched between woven and non-woven polypropylene geotextiles held together by needle-punched fibers. The GCL was 6-mm thick in an air-dried condition, and the average gravimetric water content and specific gravity (ASTM D 854) for the bentonite were 4% and 2.74, respectively. The bentonite mass (ASTM D 5993) of the GCL was 5.1 kg/m<sup>2</sup>. The air-dried bentonite consists of 20% ( $w/w$ ) medium sand-sized (425–2000  $\mu\text{m}$ ) and 72% ( $w/w$ ) fine sand-sized granules (75–425  $\mu\text{m}$ ) based on mechanical sieve analyses, whereas approximately 90% ( $w/w$ ) of the particles are clay sized (<5  $\mu\text{m}$ ) based on hydrometer analyses (ASTM D 421, D 422). The liquid limit and plasticity index (ASTM D 4318) of the bentonite are 430 and 393, respectively, and the bentonite classifies as a high plasticity clay (CH) according to ASTM D 2487. The mineralogy of the bentonite is dominated by montmorillonite (77.2%), and the total exchange complex of 61.7 meq/100 g is comprised of 74.2% sodium ( $\text{Na}^+$ ), 18.6% calcium ( $\text{Ca}^{2+}$ ), 6.0% magnesium ( $\text{Mg}^{2+}$ ), and 1.1% potassium ( $\text{K}^+$ ). Further details of the physical and chemical properties and mineralogical composition of the bentonite are given in Lee and Shackelford (2005).

### *Permeant Liquids*

The permeant liquids used in the experiments consist of tap water that is processed by passage through three Barnstead® ion exchange columns in series (pH=5.6, electrical conductivity, EC, at 25°C=0.2 mS/m) and solutions containing 5, 10, 20, 50, and 100 mM calcium chloride ( $\text{CaCl}_2$ ). The processed tap water classifies as Type IV de-ionized water (DIW) per ASTM D 1193. Calcium chloride was chosen primarily because previous studies involving permeation of bentonite-based hydraulic barrier materials (e.g., GCLs, sand-bentonite mixtures) with  $\text{CaCl}_2$  solutions have shown significant effects of the solutions on the hydraulic conductivity of these materials (Alther et al., 1985; Daniel et al., 1993; Shackelford, 1994; Imamura et al., 1996; Gleason et al., 1997; James et al., 1997; Melchior, 1997; Quaranta et al., 1997; Ruhl and Daniel, 1997; Kolstad, 2000; Lin and Benson, 2000; Shackelford et al., 2000; Egloffstein, 2001; Jo et al., 2001; Vasko et al., 2001; Shan and Lai, 2002).

The  $\text{CaCl}_2$  solutions were prepared by dissolving  $\text{CaCl}_2$  (powdered, >96% pure, Sigma-Aldrich Co., St. Louis, MO) in the DIW. Each solution was mixed in a 20-L

carboy, and pH, EC, and solute concentrations of the permeant liquids were monitored with time using a pH meter (Accumet® AB15 meter, Fisher Scientific Co., Pittsburgh, PA), an EC probe (Accumet® AB30 meter, Fisher Scientific Co., Pittsburgh, PA), an ion chromatograph (Dionex® 4000i IC Module, Dionex Co., Sunnyvale, CA) for chloride concentrations, and an inductively coupled plasma-atomic emission spectrometer (IRIS® Advantage/1000 ICAP Spectrometer, Thermo Jarrell Ash Co., Franklin, MA) for calcium concentrations, respectively. Further details on the measured properties of the permeant liquids are given in Lee and Shackelford (2005).

### *Hydraulic Conductivity Tests*

Both prehydrated and non-prehydrated specimens of the GCL with nominal diameters of 102 mm were permeated with each of the  $\text{CaCl}_2$  solutions. For the prehydrated specimens, prehydration was achieved by permeating GCL specimens with the DIW prior to permeation with the  $\text{CaCl}_2$  solutions. The specimens were permeated using the falling-head procedure with flexible-wall permeameters in a temperature-controlled laboratory (23.0±0.5°C). A pictorial view and schematic of the flexible-wall testing apparatus are shown in Fig. 2. The cell pressure was applied by gravity through lines connecting a large water tank located at height above the permeameters and vented to atmosphere to the water contained in the annulus area surrounding the GCL specimen inside the flexible-wall cell (see Fig. 2). The use of this gravity controlled system provided for the sustained application of a constant cell pressure over the long test durations encountered in this study. Inflow and effluent burettes were located above and below the permeameters, respectively, to induce flow from the bottom of the specimen to the top of the specimen (see Fig. 2(b)). A thin layer of Parafilm "M"® laboratory film (American National Can™, Chicago, IL) was stretched and sealed over the tops of both burettes to minimize evaporation, and a pin hole was punched through the film to equilibrate the head space in each burette to atmospheric pressure.

Flow was controlled via valves located in each of the flow lines (see Fig. 2(b)). Prior to permeation, both inflow and outflow lines were saturated by circulating with the permeant liquid and DIW, respectively, through porous stones located on either side of the specimen (not shown). This saturation procedure eliminated air bubbles trapped in the tubes and allowed for the establishment of a known constant head outflow boundary prior to the start of permeation (i.e., since the outflow tube was saturated, the drainage end of the outflow tube represented atmospheric pressure). Backpressure was not used so that effluent could be conveniently collected from the effluent burettes for pH, EC, and solute concentration measurements.

Although complete saturation of the specimens cannot be assured without backpressure saturation, previous studies indicate that the measured hydraulic conductivity

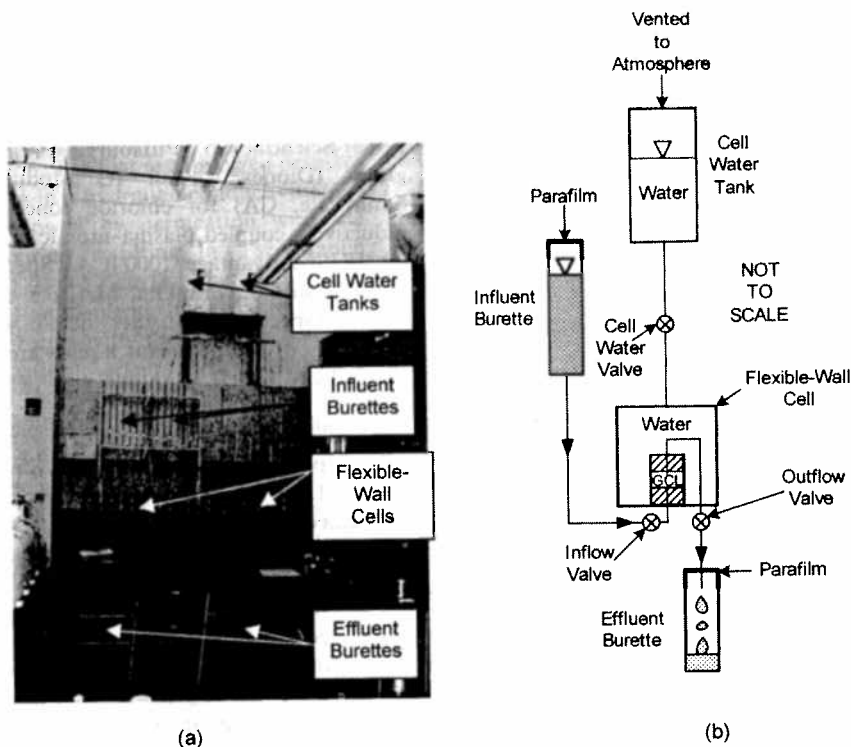


Fig. 2. Flexible-wall testing apparatus used in this study: (a) pictorial view and (b) schematic of set-up

Permeant liquid
5 mM CaCl <sub>2</sub>
10 mM CaCl <sub>2</sub>
20 mM CaCl <sub>2</sub>
50 mM CaCl <sub>2</sub>
100 mM CaCl <sub>2</sub>

a)  $k$  = hydraulic  
 b) Values bas  
 c) Values at c  
 d) Values for  
 e) Based on a

at steady-state (i.e., at chemical equilibrium in this study) likely represents the maximum value of hydraulic conductivity that can be obtained under the imposed test conditions (e.g., Chiu and Shackelford, 1998). In addition, for specimens undergoing prolonged permeation, the degree of saturation is expected to be sufficiently high such that, for all practical purposes, any air existing in the specimen is occluded (discontinuous) and the measured hydraulic conductivity is essentially the same as the saturated hydraulic conductivity. For these reasons, hydraulic conductivity values measured at steady-state in rigid-wall permeameters are assumed to be equal to the saturated hydraulic conductivity values (e.g., Daniel, 1994).

For permeation, both the inflow and outflow valves were opened such that the permeant liquid decreased with time in the influent burette under gravity, flowed into the porous stone on the inward (bottom) side of the specimen and upward through the specimen, collected in the porous stone on the outward end (top) of the specimen, and drained under gravity into the effluent burette. Thus, with constant cell and tailwater pressures but decreasing headwater pressure, the effective stress in the specimen varied not only throughout the specimen at a given time, but also with time at a given location within the specimen. The influent burette was filled periodically with fresh permeant liquid, and the effluent burette was emptied periodically as needed for measurement of the chemical properties of the effluent.

All specimens were trimmed and assembled in the permeameters using the procedures described by Daniel et al. (1997) to prevent the possibility of short-circuiting through the geotextiles at the edge of the GCL specimens due to loss of bentonite and pinching of the geotextiles. The specimens initially were exposed to water (prehydrated) or a CaCl<sub>2</sub> solution (non-prehydrated) for at least 48 hrs prior to permeation by opening the inflow valve only, i.e., by keeping the outflow valve closed (see Fig. 2(b)). The thickness of each specimen was measured before, during, and after the hydraulic conductivity test using a caliper, a cathetometer (telescope), and a ruler. The measured thickness at the start of permeation was used to estimate the void (pore) volume for each specimen, whereas the final GCL bulk void ratio was estimated based on the procedure described by Petrov et al. (1997b).

All the specimens were permeated at an average hydraulic gradient of 200 such that the average effective stresses at the bottom (inflow), middle, and top (outflow) of the specimens were 14.7 kPa, 23.5 kPa, and 32.3 kPa, respectively. While a hydraulic gradient of 200 is significantly higher than the maximum gradient (i.e., 30) stipulated in ASTM D 5084 for flexible-wall hydraulic conductivity testing of low-permeability soils ( $\leq 10^{-7}$  cm/s), hydraulic gradients ranging from 50 to 600 are commonly used for measuring the hydraulic conductivity of GCLs (e.g., Shan and Daniel, 1991; Daniel et al., 1993; Didier and Comeaga, 1997; Petrov and Rowe,

1997; Petr and Danie al., 2000). hydraulic extent by a hydraulic : None o equilibriu EC and s establish 2000). Eq when the influent E and ASTM (Cl<sup>-</sup>) and when the s  $\pm 10\%$  o Shackelfo: chemical c tions for actual, lo: meated wi 10 times h these term: Equilib: terminatic Shackelfo decreased dissolution: into the :

Table 1. Summary of test results<sup>a)</sup>

Permeant liquid	Test No.	Pre-hydration	Permeation w/Water <sup>b)</sup>				Permeation w/CaCl <sub>2</sub> solution <sup>c)</sup>		Hydraulic conductivity ratios		
			ASTM D 5084		Final		PVF <sub>NP</sub> or PVF <sub>P</sub> [t <sub>NP</sub> or t <sub>P</sub> (days)]	k <sub>NP</sub> or k <sub>P</sub> (cm/s)	KR1 <sup>d)</sup> (k <sub>P</sub> /k <sub>w</sub> )	KR2 <sup>d)</sup> (k <sub>NP</sub> /k <sub>w</sub> )	FER (k <sub>NP</sub> /k <sub>P</sub> = KR2/KR1)
			PVF <sub>w</sub> [t <sub>w</sub> (days)]	k <sub>w</sub> (cm/s)	PVF <sub>w</sub> [t <sub>w</sub> (days)]	k <sub>w</sub> (cm/s)					
5 mM CaCl <sub>2</sub>	1	No	—	—	—	—	57 [502]	8.6 × 10 <sup>-9</sup>	3.3	3.6	1.1
	2	Yes	1.6 [8.6]	3.5 × 10 <sup>-9</sup>	26 [379]	2.4 × 10 <sup>-9</sup>	58 [358]	7.8 × 10 <sup>-9</sup>			
10 mM CaCl <sub>2</sub>	3	No	—	—	—	—	30 [397]	6.7 × 10 <sup>-9</sup>	2.5	2.7	1.1
	4	Yes	1.3 [26]	2.2 × 10 <sup>-9</sup>	21 [379]	2.5 × 10 <sup>-9</sup>	35 [360]	6.2 × 10 <sup>-9</sup>			
20 mM CaCl <sub>2</sub>	5	No	—	—	—	—	21 [164]	8.1 × 10 <sup>-9</sup>	3.9	3.6	0.9
	6	Yes	1.2 [26]	2.6 × 10 <sup>-9</sup>	17 [379]	2.2 × 10 <sup>-9</sup>	16 [120]	8.8 × 10 <sup>-9</sup>			
50 mM CaCl <sub>2</sub>	7	No	—	—	—	—	9.3 [16]	1.6 × 10 <sup>-8</sup>	14	7.1 <sup>e)</sup>	0.5 <sup>e)</sup>
	8	No	—	—	—	—	8.9 [15]	1.9 × 10 <sup>-8</sup>			
	9	Yes	1.7 [26]	2.3 × 10 <sup>-9</sup>	24 [439]	2.5 × 10 <sup>-9</sup>	11 [24]	3.5 × 10 <sup>-8</sup>			
100 mM CaCl <sub>2</sub>	10	No	—	—	—	—	6.8 [0.80]	3.0 × 10 <sup>-7</sup>	49	148 <sup>e)</sup>	3.0 <sup>e)</sup>
	11	No	—	—	—	—	6.0 [0.59]	3.7 × 10 <sup>-7</sup>			
	12	Yes	1.4 [26]	2.2 × 10 <sup>-9</sup>	26 [406]	2.3 × 10 <sup>-9</sup>	10 [10]	1.1 × 10 <sup>-7</sup>			

<sup>a)</sup>  $k$  = hydraulic conductivity, PVF = pore volumes of flow,  $t$  = elapsed time,  $w$  = water, NP = non-prehydrated, P = prehydrated, respectively.

<sup>b)</sup> Values based on termination criteria in ASTM D 5084 and at the end of permeation with water (i.e., final).

<sup>c)</sup> Values at chemical equilibrium in terms of both electrical conductivity (EC) and solute concentrations.

<sup>d)</sup> Values for KR1 and KR2 are based on final  $k_w$  values.

<sup>e)</sup> Based on average of  $k_{NP}$  values.

1997; Petrov et al., 1997a,b; Quaranta et al., 1997; Ruhl and Daniel, 1997; Lin and Benson, 2000; Shackelford et al., 2000). Shackelford et al. (2000) also show that the hydraulic conductivity of GCLs is affected to a greater extent by average effective stress than by the magnitude of hydraulic gradient.

None of the tests was terminated before chemical equilibrium between effluent and influent in terms of EC and solute concentrations (Cl<sup>-</sup>, Ca<sup>2+</sup>) had been established (e.g., Daniel, 1994; Shackelford et al., 1999, 2000). Equilibrium in EC was considered established when the EC of the effluent was within ±10% of the influent EC in accordance with Shackelford et al. (1999) and ASTM D 6766, and equilibrium for both chloride (Cl<sup>-</sup>) and calcium (Ca<sup>2+</sup>) was considered established when the solute concentrations in the effluent were within ±10% of those in the source solutions (e.g., see Shackelford et al., 1999). Although this requirement for chemical equilibrium resulted in extensive testing durations for some tests, recent results indicate that the actual, long-term hydraulic conductivity of GCLs permeated with inorganic salt solutions may be as much as 10 times higher than the hydraulic conductivity based on these termination criteria (Jo et al., 2005).

Equilibrium in pH also was initially considered as a termination criterion. However, as noted by Lee and Shackelford (2005), the pH of all influent solutions decreased slowly over time due to time-dependent dissolution of carbon dioxide (CO<sub>2</sub>) from the atmosphere into the solutions (e.g., see Shackelford, 1994). As a

result, equilibrium based on pH was not found to be particularly applicable in this study because the pH ratio (i.e., pH<sub>out</sub>/pH<sub>in</sub>) never met the typical acceptable range (i.e., 1.00 ± 0.10) throughout the tests (Shackelford et al., 1999). However, the limited variation (i.e., ~5.9 ± 0.4) in influent pH probably had little, if any, effect on the measured hydraulic conductivity relative to the effects caused by the CaCl<sub>2</sub> solutions (Shackelford, 1994; Ruhl and Daniel, 1997; Shackelford et al., 2000; Jo et al., 2001; Shan and Lai, 2002; Shackelford and Lee, 2005). Thus, although pH of the influent and effluent was measured, equilibrium in pH was not considered as a termination criterion in this study.

The hydraulic conductivity tests performed on non-prehydrated specimens using solutions with relatively high CaCl<sub>2</sub> concentrations (i.e., 50 and 100 mM) were duplicated because the test durations were relatively short, allowing for the re-use of the permeameters. However, because the tests using permeant liquids containing relatively low CaCl<sub>2</sub> concentrations (i.e., 5, 10, and 20 mM) required extensive test durations (>4 months) before chemical equilibrium was established, these tests were not duplicated due to the limited number of permeameters available for testing.

## RESULTS

All the test results are shown in Figs. 3 to 7 and summarized in Table 1. As indicated by the arrows shown in Figs. 3 to 7, permeation of the prehydrated specimens

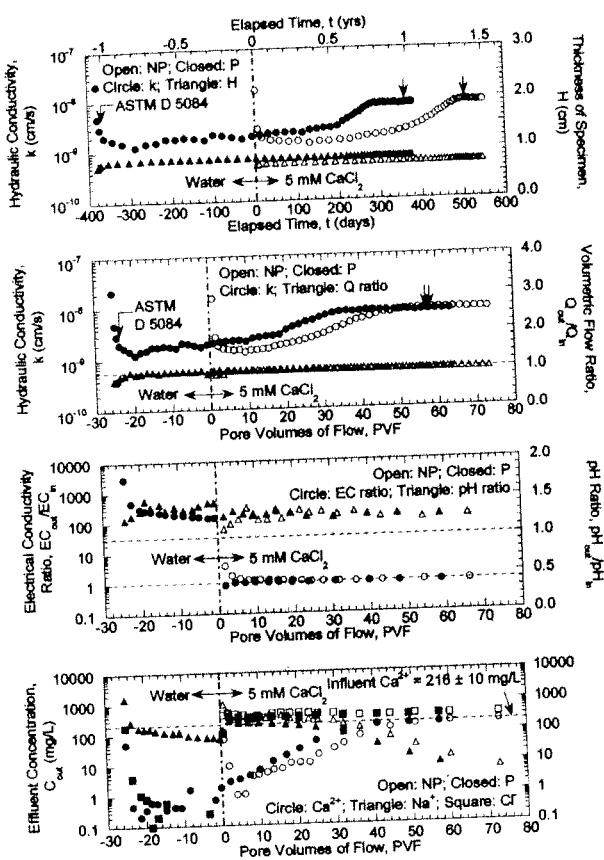


Fig. 3. Test results for both non-prehydrated (NP) and prehydrated (P) specimens permeated with 5 mM CaCl<sub>2</sub> solution (Arrows designate time at compliance with ASTM D 5084 or chemical equilibrium)

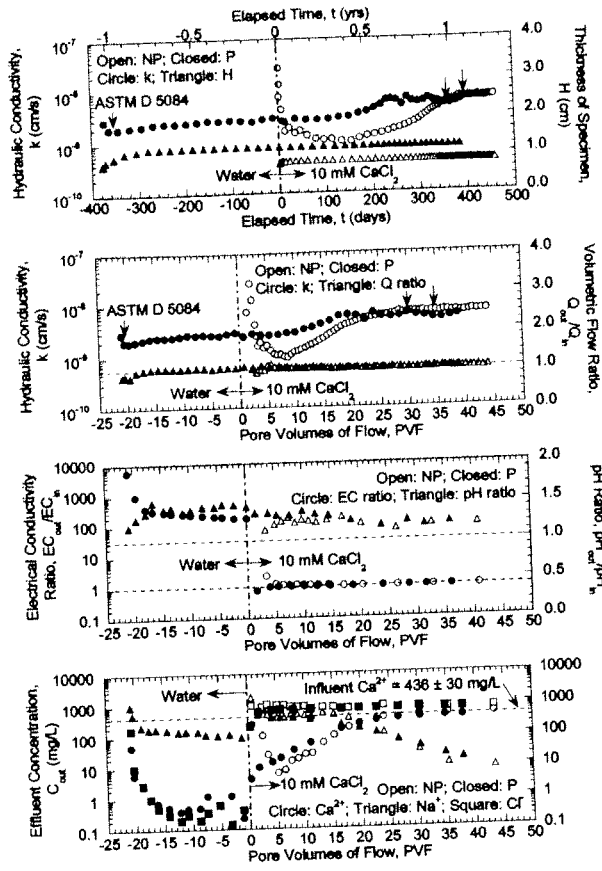


Fig. 4. Test results for both non-prehydrated (NP) and prehydrated (P) specimens permeated with 10 mM CaCl<sub>2</sub> solution (Arrows designate time at compliance with ASTM D 5084 or chemical equilibrium)

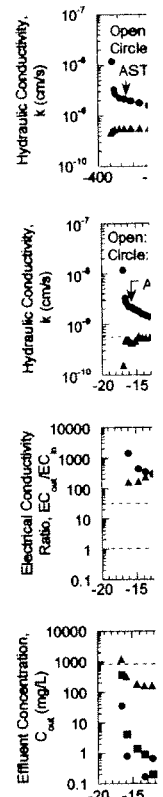


Fig. 5. Test results for both non-prehydrated (NP) and prehydrated (P) specimens permeated with 20 mM CaCl<sub>2</sub> solution (Arrows designate time at compliance with ASTM D 5084 or chemical equilibrium)

with water was extended well beyond the durations based on the termination criteria specified in ASTM D 5084. As a result, two values for the hydraulic conductivity of each specimen based on permeation with water ( $k_w$ ) are given in Table 1, i.e., one  $k_w$  value corresponding to the termination criteria specified in ASTM D 5084 and one  $k_w$  value corresponding to the end of permeation with water.

In general, the majority of the values for  $k_w$  at the end of permeation are within  $\pm 15\%$  of the values based on the termination criteria in ASTM D 5084, with the only exception pertaining to Test No. 2. In addition, the average of the five values for  $k_w$  corresponding to the end of permeation of  $2.4 \times 10^{-9}$  cm/s is very close to the average  $k_w$  value of  $2.6 \times 10^{-9}$  cm/s based on the termination criteria in ASTM D 5084. Finally, except for the  $k_w$  value based on ASTM D 5084 for Test No. 2, all of the  $k_w$  values are in close agreement with the respective average value (i.e.,  $\leq 15.4\%$  difference for the  $k_w$  values based on ASTM D 5084 and  $\leq 8.3\%$  difference based on the final  $k_w$  values). Daniel et al. (1997) also reported similar results and reproducibility based on round-robin tests from 18 different laboratories performed with water for a single type and make of GCL.

Results of the hydraulic conductivity tests for both

non-prehydrated and prehydrated GCL specimens permeated with 5, 10, and 20 mM CaCl<sub>2</sub> solutions are shown in Figs. 3 to 5, respectively. The hydraulic conductivity values for all the non-prehydrated specimens initially decreased to approximately  $10^{-9}$  cm/s after 1 month of permeation ( $< 5$  PVF), and then began to increase after 3 to 6 months (5 to 10 PVF). Finally, the hydraulic conductivity values for all three non-prehydrated specimens increased by approximately 7 times to  $7.8 \times 10^{-9}$  cm/s by the time chemical equilibrium was established, corresponding to 1.4 yrs (57 PVF), 1.1 yrs (31 PVF), and 0.4 yr (21 PVF) for permeation with the 5, 10, and 20 mM CaCl<sub>2</sub> solutions, respectively. After establishment of chemical equilibrium, all of the effluent sodium (Na<sup>+</sup>) concentrations for the non-prehydrated specimens permeated with 5, 10, and 20 mM CaCl<sub>2</sub> solutions were  $\leq 40$  mg/L.

For the prehydrated specimens permeated with 5, 10, and 20 mM CaCl<sub>2</sub> solutions, the hydraulic conductivity values at the end of prehydration (i.e., permeation with water) were approximately  $2.4 \times 10^{-9}$  cm/s for all specimens. However, after switching the influent from water to the CaCl<sub>2</sub> solutions, the hydraulic conductivity values

began to increase with the duration of permeation. The prehydrated specimens permeated with 20 mM CaCl<sub>2</sub> solution had the highest hydraulic conductivity values, which were approximately 7 times greater than those for the non-prehydrated specimens permeated with 20 mM CaCl<sub>2</sub> solution.

In addition to the increase in hydraulic conductivity, the prehydrated specimens permeated with 20 mM CaCl<sub>2</sub> solution had the highest effluent sodium concentrations after 1 yr (20 PVF) of permeation, which were approximately 100 mg/L. These relative differences in hydraulic conductivity and effluent sodium concentrations after 1 yr or more of permeation are attributed, in part, to the presence of fine particles in the influent water, which are likely to be trapped in the GCL pores, resulting in a reduction in hydraulic conductivity and an increase in effluent sodium concentrations.

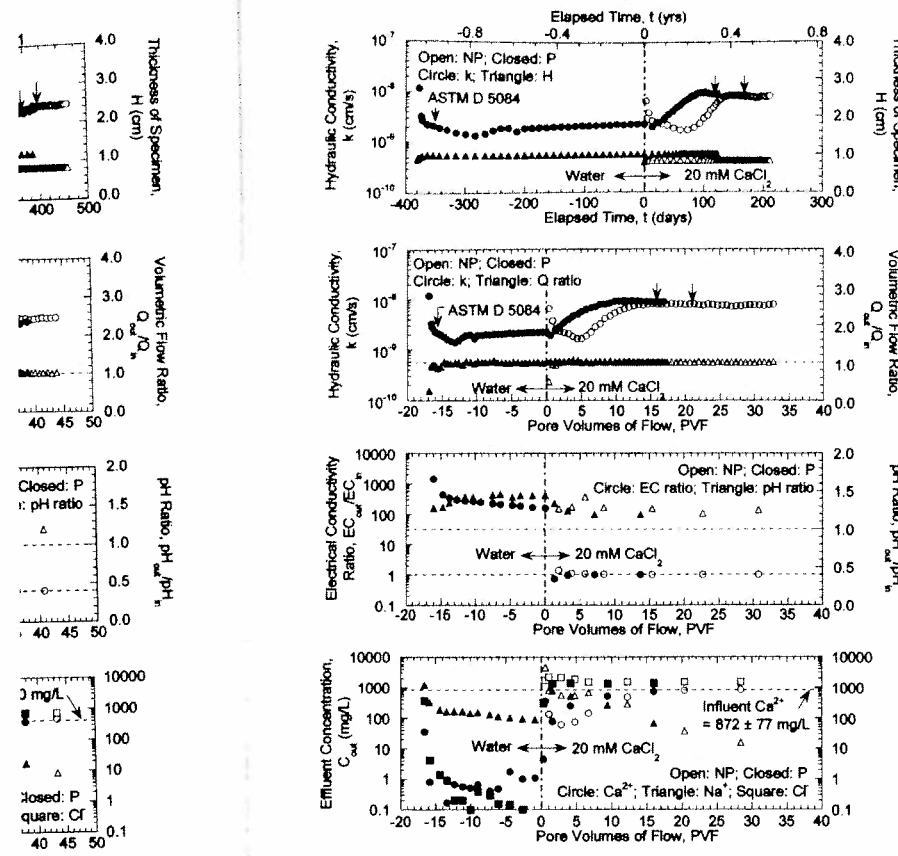


Fig. 5. Test results for both non-prehydrated (NP) and prehydrated (P) specimens permeated with 20 mM CaCl<sub>2</sub> solution (Arrows designate time at compliance with ASTM D 5084 or chemical equilibrium)

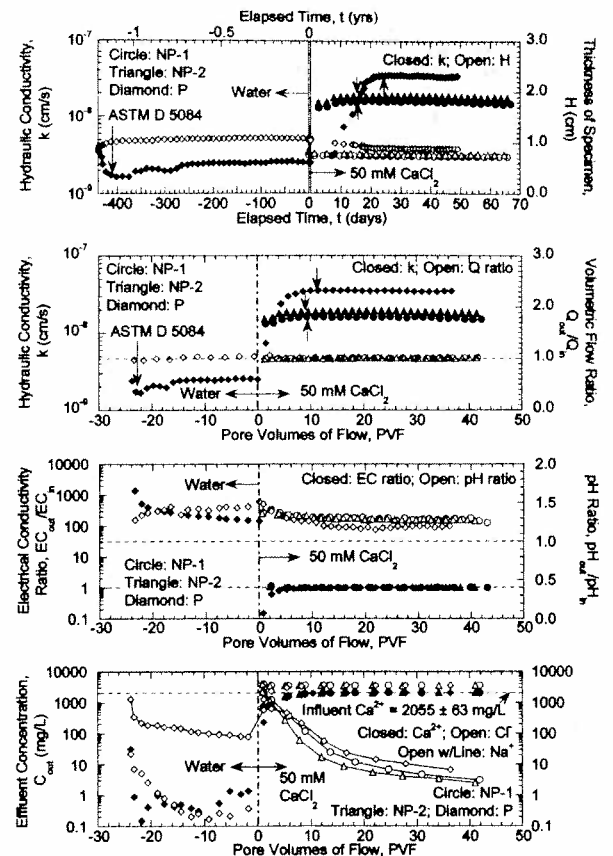


Fig. 6. Test results for both non-prehydrated (NP) and prehydrated (P) specimens permeated with 50 mM CaCl<sub>2</sub> solution (Arrows designate time at compliance with ASTM D 5084 or chemical equilibrium)

and prehydrated solution (Arrows 5084 or chemical

specimens permeations are shown. Hydraulic conductivity values initially after 1 month of permeation increase after 3 months (10 PVF) and 10, and 20 mM CaCl<sub>2</sub> solutions were established, corresponding to 10, and 20 PVF of permeation with water can be attributed, in part, to diffusion-limited leaching of soluble Na<sup>+</sup> from the interlayer region of the bentonite particles to the interparticle and interclod migration pathways resulting from the relatively slow flow rate during

the prehydration period (Coats and Smith, 1964; van Genuchten and Wierenga, 1976; Pusch, 1999). Upon subsequent permeation of these prehydrated specimens with the 5, 10, and 20 mM CaCl<sub>2</sub> solutions, the effluent Na<sup>+</sup> concentrations increased relatively quickly to values ranging from about 100 to 1,000 mg/L, presumably due to cation exchange of the Ca<sup>2+</sup> for the Na<sup>+</sup> on the exchange complex of the bentonite particles. However, by the end of the permeation with the CaCl<sub>2</sub> solutions, the effluent Na<sup>+</sup> concentrations had once again decreased to values ≤ 70 mg/L for all the prehydrated specimens permeated with 5, 10, and 20 mM CaCl<sub>2</sub> solutions.

Results of the hydraulic conductivity tests for both non-prehydrated and prehydrated GCL specimens permeated with 50 and 100 mM CaCl<sub>2</sub> solutions as the permeant liquids are shown in Figs. 6 and 7, respectively. For the duplicate non-prehydrated specimens permeated with 50 and 100 mM CaCl<sub>2</sub> solutions, the hydraulic conductivity values stabilized within 1 month (10 PVF) at approximately 1.8 × 10<sup>-8</sup> and 3.4 × 10<sup>-7</sup> cm/s, respectively. Chemical equilibrium for these specimens also was established within this same time frame. These hydraulic conductivity values at chemical equilibrium are 2.3 times

the prehydration period (Coats and Smith, 1964; van Genuchten and Wierenga, 1976; Pusch, 1999). Upon subsequent permeation of these prehydrated specimens with the 5, 10, and 20 mM CaCl<sub>2</sub> solutions, the effluent Na<sup>+</sup> concentrations increased relatively quickly to values ranging from about 100 to 1,000 mg/L, presumably due to cation exchange of the Ca<sup>2+</sup> for the Na<sup>+</sup> on the exchange complex of the bentonite particles. However, by the end of the permeation with the CaCl<sub>2</sub> solutions, the effluent Na<sup>+</sup> concentrations had once again decreased to values ≤ 70 mg/L for all the prehydrated specimens permeated with 5, 10, and 20 mM CaCl<sub>2</sub> solutions.

Results of the hydraulic conductivity tests for both non-prehydrated and prehydrated GCL specimens permeated with 50 and 100 mM CaCl<sub>2</sub> solutions as the permeant liquids are shown in Figs. 6 and 7, respectively. For the duplicate non-prehydrated specimens permeated with 50 and 100 mM CaCl<sub>2</sub> solutions, the hydraulic conductivity values stabilized within 1 month (10 PVF) at approximately 1.8 × 10<sup>-8</sup> and 3.4 × 10<sup>-7</sup> cm/s, respectively. Chemical equilibrium for these specimens also was established within this same time frame. These hydraulic conductivity values at chemical equilibrium are 2.3 times





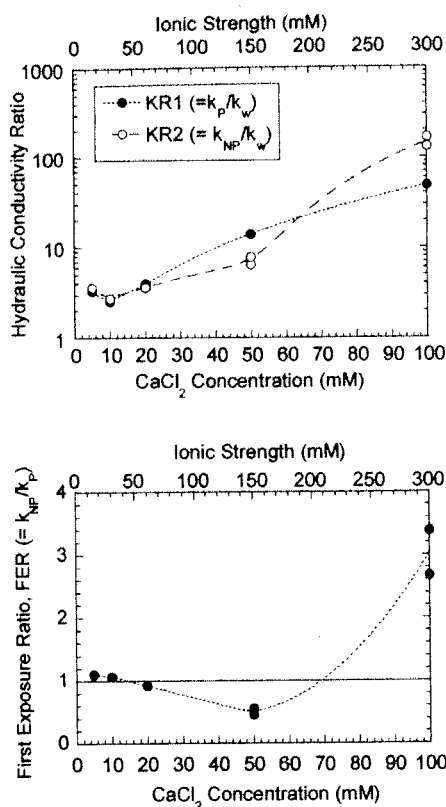


Fig. 9. Hydraulic conductivity ratios relative to that based on permeation with water for both prehydrated (KR1) and non-prehydrated (KR2) specimens and first exposure ratio ( $FER = k_{NP}/k_P = KR2/KR1$ ) as a function of influent  $CaCl_2$  concentration

effect of initial permeation with the chemical solution relative to initial permeation with water on two separate but identical test specimens; and (3) the first exposure ratio,  $FER (= k_{NP}/k_P = KR2/KR1)$ , which represents the ratio of the hydraulic conductivity of the specimen permeated initially with the chemical solution to the hydraulic conductivity of a separate but identical specimen permeated with the same chemical solution after initial permeation with water.

Values for KR1, KR2, and FER as summarized in Table 1 are shown in Fig. 9 as a function of  $CaCl_2$  concentration of the permeant liquid. For both non-prehydrated and prehydrated specimens permeated with 5, 10, and 20 mM  $CaCl_2$  solutions, the KR1 and KR2 values range from 2.5 to 3.9, and the resulting FER is within 10% of unity (i.e.,  $1.0 \pm 0.1$ ). In other words, the specimens permeated with 5, 10, and 20 mM  $CaCl_2$  solutions are 3.2 times on average more permeable than those permeated with water, regardless of whether or not the specimens are prehydrated, but prehydration has little, if any, effect on hydraulic conductivity at chemical equilibrium for solutions containing 5, 10, and 20 mM  $CaCl_2$ .

However, for the specimens permeated with the 50 mM  $CaCl_2$  solution, the KR1 and KR2 values are 14 and 7.1,

respectively, and the resulting FER is 0.5. Thus, since  $FER < 1$ , prehydration also has no effect on hydraulic conductivity based on chemical equilibrium for the 50 mM  $CaCl_2$  solution, although the hydraulic conductivities are 10 times higher on average than those based on permeation with water.

For the specimens permeated with the 100 mM  $CaCl_2$  solution, the KR1 and KR2 values are 49 and 148, respectively, and the resulting FER is 3.0. As a result, the hydraulic conductivity for the prehydrated specimen permeated with the 100 mM  $CaCl_2$  solution is 49 times higher than that based on permeation with water, whereas the hydraulic conductivity for the non-prehydrated specimen permeated directly with the same solution is 148 times higher than that based on permeation with water. Therefore, prehydration has a relatively significant effect on hydraulic conductivity at chemical equilibrium for the tests performed with the 100 mM  $CaCl_2$  solution.

In general, prehydration has little, if any, effect on the hydraulic conductivity of the GCL specimens tested in this study for solutions containing  $\leq 50$  mM  $CaCl_2$  concentration (i.e.,  $0.5 \leq FER \leq 1.1$ ), provided the tests are continued until chemical equilibrium is established. Vasko et al. (2001) report similar results based on their tests in which a GCL was permeated with  $\leq 25$  mM  $CaCl_2$  solutions, even though prehydration was imposed without confinement. However, for the tests performed in this study using the 100 mM  $CaCl_2$  solution as the permeant liquid, prehydration had a relatively significant effect on hydraulic conductivity (i.e.,  $FER \approx 3.0$ ). Nonetheless, this value of FER also is significantly lower than the value of 230 based on the results by Vasko et al. (2001) for tests performed using a 100 mM  $CaCl_2$  solution as the permeant liquid for both non-prehydrated and prehydrated GCL specimens with a confining stress of 8 kPa. This significant difference in the FER between the two studies based on permeation with 100 mM  $CaCl_2$  solutions likely is due, in part, to the differences between the test conditions applied in the study by Vasko et al. (2001) relative to this study, including the confinement method during prehydration (i.e., uniaxial vs. triaxial), the average confining stress applied during prehydration (i.e., 8 kPa vs. 23.5 kPa), and the average effective stress applied during permeation (i.e., 20 kPa vs. 23.5 kPa).

#### Concentration Dependency of Prehydration Effect on Test Duration

As shown in Fig. 10, less time is required to achieve chemical equilibrium with increasing  $CaCl_2$  concentration of the permeant liquid regardless of whether or not the specimens are prehydrated. However, the elapsed times at chemical equilibrium (i.e., excluding the time required for prehydration) for the tests performed using the 5, 10, and 20 mM  $CaCl_2$  solutions as permeant liquids are greater for the non-prehydrated specimens relative to the prehydrated specimens, whereas the elapsed times at chemical equilibrium for the tests performed using the 50 and 100 mM  $CaCl_2$  solutions are greater for the

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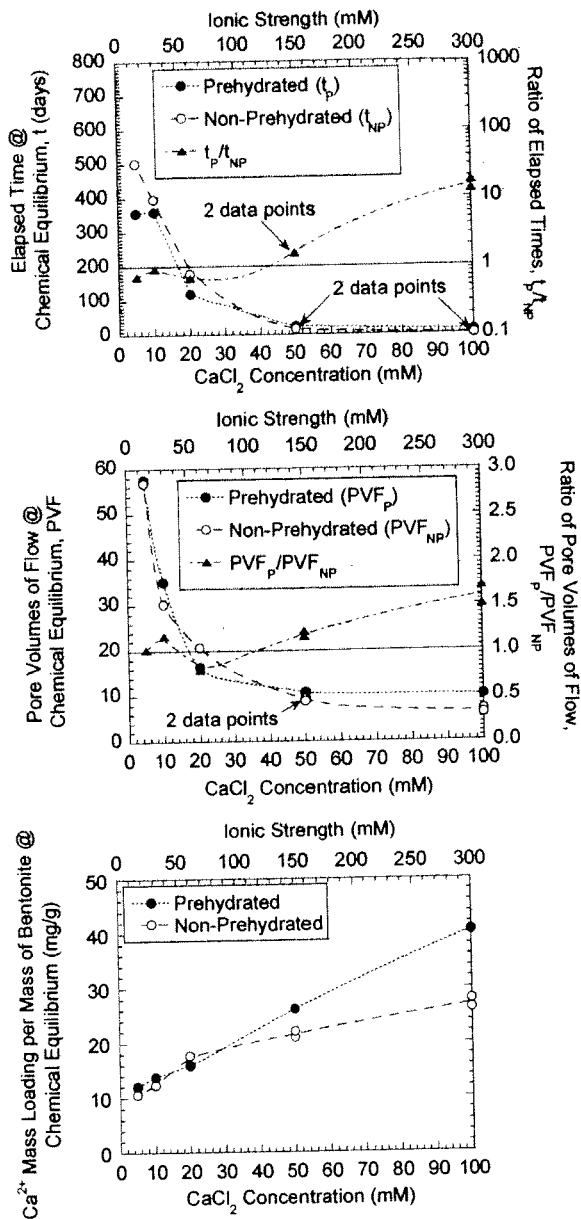


Fig. 10. Elapsed time, elapsed pore volumes of flow, and calcium ( $\text{Ca}^{2+}$ ) mass loading per dry mass of bentonite at chemical equilibrium for both prehydrated and non-prehydrated specimens as a function of influent  $\text{CaCl}_2$  concentration

prehydrated specimens relative to the non-prehydrated specimens. These trends in test durations are a direct consequence of the temporal trends in the hydraulic conductivity values.

For example, as shown in Figs. 3 to 5 for the tests involving permeation with the 5, 10, and 20 mM  $\text{CaCl}_2$  solutions, the hydraulic conductivity of the non-prehydrated specimens is initially substantially higher than that of the prehydrated specimens, but decreases to a value that is lower than that for the prehydrated specimens before eventually increasing to the final value at chemical

equilibrium. However, for the tests involving permeation with the 50 and 100 mM  $\text{CaCl}_2$  solutions (Figs. 6 and 7), the hydraulic conductivity of the non-prehydrated specimens is initially higher than that for the prehydrated specimens, and increases (rather than decreases) with time to the final value at chemical equilibrium.

The fact that the thicknesses of both the prehydrated and non-prehydrated specimens remain essentially constant throughout the durations of permeation with the  $\text{CaCl}_2$  solutions (see Figs. 3 to 7) suggests that these temporal changes in hydraulic conductivity are due to physicochemical interactions rather than to changes in void ratio. For example, as described by Jo et al. (2001), the hydraulic conductivity of bentonite is a function of the degree of exchange that occurs between the original exchange complex and the permeant liquid, and the resulting volume of bound water, and that the degree of exchange will depend on the concentration of the permeating liquid and the duration of permeation. Accordingly, the degrees of  $\text{Ca}^{2+}$  for  $\text{Na}^+$  exchange that occur for the specimens permeated with the 5, 10, and 20 mM  $\text{CaCl}_2$  solutions apparently are the same for both prehydrated and non-prehydrated specimens since the hydraulic conductivity values at chemical equilibrium for these specimens are essentially the same (Table 1 and Fig. 8). In contrast, the degrees of  $\text{Ca}^{2+}$  for  $\text{Na}^+$  exchange between the prehydrated and non-prehydrated specimens permeated with the 50 and 100 mM  $\text{CaCl}_2$  solutions apparently are not the same, since the respective hydraulic conductivity values at chemical equilibrium are not the same.

Similar to the test durations, the number of pore volumes of flow (PVF) required for chemical equilibrium decreases with increasing  $\text{CaCl}_2$  concentration of the permeant liquid regardless of whether or not the specimens are prehydrated, as shown in Fig. 10. However, unlike the test durations, the numbers of PVF required to achieve chemical equilibrium for the prehydrated specimens are relatively close to those for the non-prehydrated specimens (i.e., <30% difference) for the specimens permeated with the 5, 10, and 20 mM  $\text{CaCl}_2$  solutions, whereas the numbers of PVF required to achieve chemical equilibrium for the prehydrated specimens permeated with 50 and 100 mM  $\text{CaCl}_2$  solutions were greater than those for the non-prehydrated specimens by a factor of 1.1 to 1.7 (i.e., 10 to 70% difference). Thus, for the tests performed using permeant liquids containing relatively low concentrations ( $\leq 20$  mM) of  $\text{CaCl}_2$ , the number of PVF required to achieve chemical equilibrium is relatively insensitive to whether or not the specimen is prehydrated. However, the number of PVF required to achieve chemical equilibrium becomes more sensitive to prehydration as the  $\text{CaCl}_2$  concentration in the permeant liquid is increased to 50 and 100 mM, with a greater number of PVF required for chemical equilibrium for the prehydrated specimens relative to the non-prehydrated specimens. However, since the hydraulic conductivity values for the tests using the 50 and 100 mM  $\text{CaCl}_2$  solutions are also greater (Fig. 8), the elapsed test

durations are relatively short.

The concentration of the permeant liquid at chemical equilibrium is the amount of permeant liquid that has permeated during permeation. The amount of permeant liquid that has permeated during permeation is a function of the permeant liquid concentration and the permeant liquid concentration at chemical equilibrium. The amount of permeant liquid that has permeated during permeation is a function of the permeant liquid concentration and the permeant liquid concentration at chemical equilibrium.

As shown in Fig. 10, the degree of exchange between the original exchange complex and the permeant liquid, and the resulting volume of bound water, and that the degree of exchange will depend on the concentration of the permeating liquid and the duration of permeation. Accordingly, the degrees of  $\text{Ca}^{2+}$  for  $\text{Na}^+$  exchange that occur for the specimens permeated with the 5, 10, and 20 mM  $\text{CaCl}_2$  solutions apparently are the same for both prehydrated and non-prehydrated specimens since the hydraulic conductivity values at chemical equilibrium for these specimens are essentially the same (Table 1 and Fig. 8). In contrast, the degrees of  $\text{Ca}^{2+}$  for  $\text{Na}^+$  exchange between the prehydrated and non-prehydrated specimens permeated with the 50 and 100 mM  $\text{CaCl}_2$  solutions apparently are not the same, since the respective hydraulic conductivity values at chemical equilibrium are not the same.

One possible explanation for the difference in test durations relates to the degree of exchange between the original exchange complex and the permeant liquid, and the resulting volume of bound water, and that the degree of exchange will depend on the concentration of the permeating liquid and the duration of permeation. Accordingly, the degrees of  $\text{Ca}^{2+}$  for  $\text{Na}^+$  exchange that occur for the specimens permeated with the 5, 10, and 20 mM  $\text{CaCl}_2$  solutions apparently are the same for both prehydrated and non-prehydrated specimens since the hydraulic conductivity values at chemical equilibrium for these specimens are essentially the same (Table 1 and Fig. 8). In contrast, the degrees of  $\text{Ca}^{2+}$  for  $\text{Na}^+$  exchange between the prehydrated and non-prehydrated specimens permeated with the 50 and 100 mM  $\text{CaCl}_2$  solutions apparently are not the same, since the respective hydraulic conductivity values at chemical equilibrium are not the same.

ng permeation (Figs. 6 and 7), hydrated specimens (decreases) with time. The prehydrated specimens essentially consistent with the tests that these differences are due to changes in Vaso et al. (2001), a function of the original quid, and the degree of permeation. Accordingly that occur 0, and 20 mM for both pre-hydrated specimens since the equilibrium for (Table 1 and  $e_B$  for  $Na^+$  ex-non-prehydrated 100 mM  $CaCl_2$  the respective equilibrium are

number of pore cal equilibrium tration of the not the speci- 10. However, VF required to prehydrated for the non-erence) for the 20 mM  $CaCl_2$  F required to hydrated speci-  $CaCl_2$  solutions hydrated speci- 30% difference). rmeant liquids ( $\leq 20$  mM) of hieve chemical ther or not the umber of PVF becomes more oncentration in 100 mM, with a cal equilibrium to the non- the hydraulic 50 and 100 mM he elapsed test

durations corresponding to this increase in PVF are relatively short (Fig. 10).

The concentration dependency of the effect of pre-hydration on the number of PVF required to achieve chemical equilibrium also can be evaluated on the basis of the amount of exchangeable  $Ca^{2+}$  delivered to a specimen during permeation (i.e.,  $Ca^{2+}$  mass loading), since this amount is a function of the concentration of  $Ca^{2+}$  in the permeant solution and the volume of the solution that has permeated through the specimen at chemical equilibrium. An evaluation based the  $Ca^{2+}$  mass loading is more fundamental than that based on PVF, because differences in the number of PVF can result simply due to differences in the void volumes of the different specimens.

As shown in Fig. 10, the amount of  $Ca^{2+}$  delivered to the specimens per dry mass of bentonite at chemical equilibrium increases with increasing  $CaCl_2$  concentration in the permeant liquid, regardless of whether or not the specimens are prehydrated. However, the amounts of  $Ca^{2+}$  delivered to the specimens per dry mass of bentonite at chemical equilibrium for both prehydrated and non-prehydrated specimens are essentially the same for the tests performed using the 5, 10, and 20 mM  $CaCl_2$  solutions, whereas significantly greater  $Ca^{2+}$  mass loadings per dry mass of bentonite were required at chemical equilibrium for the prehydrated specimens permeated with the 50 and 100 mM  $CaCl_2$  solutions relative to the non-prehydrated specimens. Since the hydraulic conductivity of the prehydrated specimen permeated with the 50 mM  $CaCl_2$  solution is greater than that for the non-prehydrated specimen permeated with the 50 mM  $CaCl_2$  solution (see Fig. 8), a direct correlation between the amount of  $Ca^{2+}$  per dry mass of bentonite delivered to the GCL and the magnitude of the resulting hydraulic conductivity exists for the specimens permeated with the 50 mM  $CaCl_2$  solutions. However, for the specimens permeated with the 100 mM  $CaCl_2$  solutions, the opposite is true; i.e., a greater  $Ca^{2+}$  mass loading per dry mass of bentonite for the prehydrated specimen correlates with a lower hydraulic conductivity relative to that for the non-prehydrated specimen.

One possible reason for this apparent discrepancy relates to the exposure of the specimens to the initial permeant liquid for 48 hrs prior to permeation, whereby the prehydrated specimens were initially exposed to DIW and the non-prehydrated specimen were initially exposed to the  $CaCl_2$  solution used as the permeant liquid. As a result, any  $Ca^{2+}$  that infused into the specimens prior to permeation of the non-prehydrated specimens, e.g., due to suction within the specimen (i.e., via advection) and/or diffusion from the inflow porous stone, is not included in the  $Ca^{2+}$  mass loadings per dry mass of bentonite based on permeation. Thus, the total  $Ca^{2+}$  mass loading (i.e., due to pre-permeation exposure and permeation) for the non-prehydrated specimen permeated with the 100 mM  $CaCl_2$  solution actually may have been greater than that for the prehydrated specimen permeated with the 100 mM  $CaCl_2$  solution, resulting in the expected correlation between  $Ca^{2+}$  mass loading and hydraulic

conductivity for the non-prehydrated and prehydrated specimens permeated with the 100 mM  $CaCl_2$  solutions. Although this same argument pertains to the specimens permeated with solutions containing the lower  $CaCl_2$  concentrations, the amounts of  $Ca^{2+}$  infused into the non-prehydrated specimens during the 48-hr exposure period for these specimens are expected to have been correspondingly lower (e.g., due to lower concentration gradients between  $Ca^{2+}$  in the porous stone and the  $Ca^{2+}$  in the pores of the bentonite). Of course, since no quantitative measure of the mass of  $Ca^{2+}$  infused into the specimens during the 48-hr pre-permeation exposure period is available, no definitive conclusion regarding this possible explanation is possible at this time.

#### *Concentration Dependency of Prehydration Effect on Final Specimen Properties*

The final bentonite gravimetric water content ( $w_b$ ), bentonite void ratio ( $e_b$ ), and GCL bulk void ratio ( $e_B$ ) for both prehydrated and non-prehydrated GCL specimens are shown in Fig. 11. As the  $CaCl_2$  concentration in the permeant liquid increases from 5 to 100 mM,  $w_b$  for the prehydrated specimen (i.e.,  $w_{b,P}$ ) decreases from 122 to 102%, whereas  $w_b$  for the non-prehydrated specimen (i.e.,  $w_{b,NP}$ ) decreases from 97 to 78%. Also,  $e_b$  for the prehydrated specimen (i.e.,  $e_{b,P}$ ) decreases from 3.3 to 2.8, whereas  $e_b$  for the non-prehydrated specimen (i.e.,  $e_{b,NP}$ ) decreases from 2.7 to 2.1. Finally,  $e_B$  for the prehydrated specimen (i.e.,  $e_{B,P}$ ) decreases from 2.8 to 2.4, whereas  $e_B$  for the non-prehydrated specimen (i.e.,  $e_{B,NP}$ ) decreases from 2.3 to 1.8 for the same range in  $CaCl_2$  concentrations.

However, the overall trends in  $w_b$ ,  $e_b$ , and  $e_B$  as a function of influent  $CaCl_2$  concentration of the permeant liquid are similar for both prehydrated and non-prehydrated specimens, such that the lower the influent  $CaCl_2$  concentration, the greater the change in  $w_b$ ,  $e_b$ , and  $e_B$ . In general, the values of  $w_b$ ,  $e_b$ , and  $e_B$  for the prehydrated specimens (i.e.,  $w_{b,P}$ ,  $e_{b,P}$ , and  $e_{B,P}$ ) are always greater by 22 to 34% than those of the non-prehydrated specimens (i.e.,  $w_{b,NP}$ ,  $e_{b,NP}$ , and  $e_{B,NP}$ ). These relative differences are consistent with the measured thickness of specimens during permeation with  $CaCl_2$  solutions, i.e., greater thicknesses for the prehydrated specimens than for the non-prehydrated specimens (see Figs. 3 to 7). Vasko et al. (2001) reported similar results for final bentonite void ratios of non-prehydrated and prehydrated GCL specimens.

The hydraulic conductivity values are correlated with the GCL bulk void ratios ( $e_B$ ) in Fig. 12. The trends in the data based on the results of the tests with the prehydrated and non-prehydrated specimens are essentially the same in that a higher  $CaCl_2$  solution results in a higher hydraulic conductivity and a lower  $e_B$  due to compression of the adsorbed layers of the bentonite particles. However,  $e_B$  for prehydrated specimens (i.e.,  $e_{B,P}$ ) is always higher than  $e_B$  for non-prehydrated specimens (i.e.,  $e_{B,NP}$ ) at a given hydraulic conductivity value, indicating a thicker adsorbed layer of hydrated cations resulting from initial

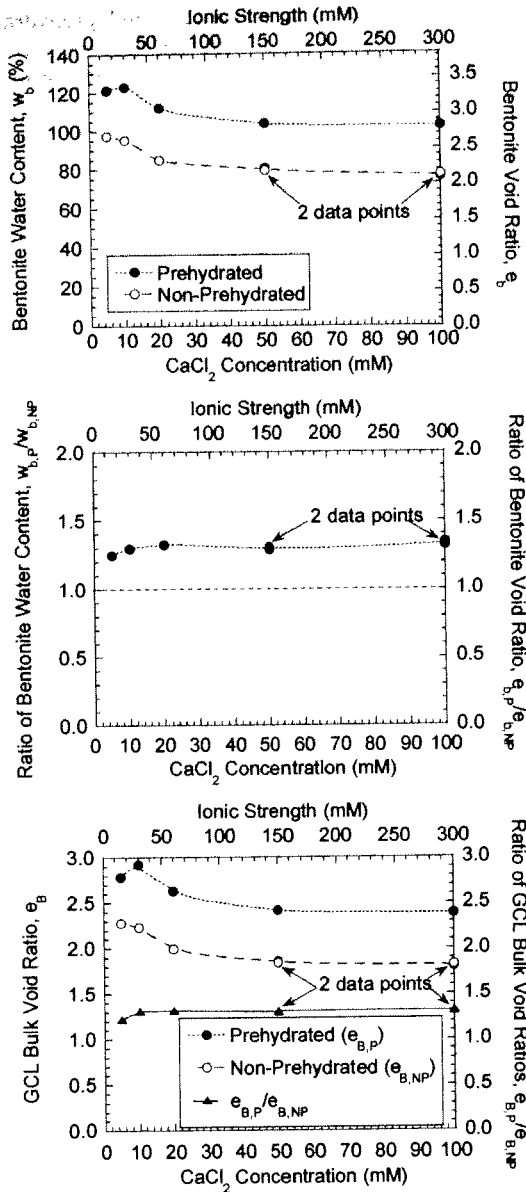


Fig. 11. Final bentonite gravimetric water content and void ratio and GCL bulk void ratio after permeation for both prehydrated and non-prehydrated specimens as a function of influent CaCl<sub>2</sub> concentration

hydration with water (e.g., Shackelford et al., 2000; Vasko et al., 2001). Thus, the effect of concentration of CaCl<sub>2</sub> in the permeant liquid on the hydraulic conductivity of the GCL specimens is as great as, if not greater than, that due to  $e_b$ . This observation is in agreement with Shackelford et al. (2000), who showed that the increase in hydraulic conductivity of prehydrated GCL specimens due to the salt (NaCl) concentration of the permeant liquid was approximately the same order as the effect of  $e_b$ , whereas the effect of salt concentration was more influential than the effect of  $e_b$  on the hydraulic conductivity of non-prehydrated GCL specimens.

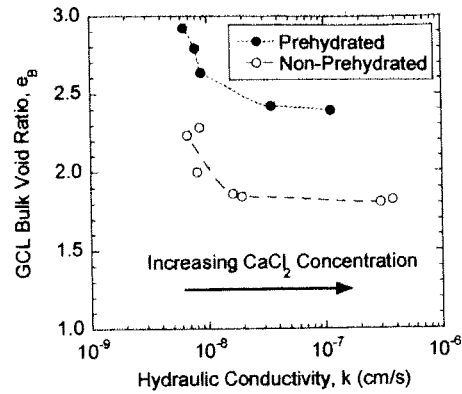


Fig. 12. Final GCL bulk void ratio after permeation versus hydraulic conductivity for both prehydrated and non-prehydrated specimens based on permeation with 5 to 100 mM CaCl<sub>2</sub> solutions

PRACTICAL RELEVANCE

All of the test results in this study are based on permeation of GCLs with single-salt solutions (i.e., CaCl<sub>2</sub>) containing a single, divalent cation (Ca<sup>2+</sup>). Thus, the question arises as to how relevant are the results in terms of actual practical applications wherein the containment liquids (e.g., leachate) include a multitude of different ionic species at different concentrations? This question has recently been addressed by Kolstad (2000) and Kolstad et al. (2004) in terms of the correlation between hydraulic conductivity of non-prehydrated GCLs and two parameters characterizing the chemistry of multi-species inorganic solutions, viz. the ionic strength,  $I$ , and the ratio of the concentrations of the monovalent to the divalent cations in the permeant solution, RMD, where:

$$I = \frac{1}{2} \sum_{i=1}^N c_i z_i^2 \tag{1}$$

and

$$RMD = \frac{M_M}{\sqrt{M_D}} \tag{2}$$

where  $c$  = the molar concentration of each species (both anions and cations) in solution,  $z$  = the valence or charge of the species,  $N$  = the number of ions in solution,  $M_M$  = the total molarity of monovalent cations, and  $M_D$  = the total molarity of divalent cations. The resulting correlation for GCLs based on numerous leachates from municipal solid wastes (MSW), construction and demolition (C&D) waste, fly ash, and mine waste is shown in Fig. 13. As indicated in Fig. 13, the predicted hydraulic conductivity of GCLs increases with increasing  $I$  for a given RMD as well as decreasing RMD for a given  $I$ . Thus, based on the correlations shown in Fig. 13, all solutions with same  $I$  and same RMD should result in the same hydraulic conductivity regardless of the composition of the solution.

To provide an example of the accuracy of the predictions of hydraulic conductivity based on Fig. 13, values of the measured hydraulic conductivity,  $k_m$ , for both the

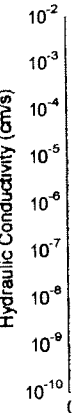


Fig. 13. Correlation of hydraulic conductivity of GCLs with ionic strength and RMD for a wide variety of C&D = const

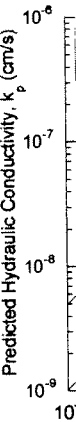


Fig. 14. Measured hydraulic conductivity of specimens tested

non-prehydrated GCLs in this study (Table 1) are close to the predicted values. The predicted hydraulic conductivity of GCLs increases with increasing ionic strength for a given RMD as well as decreasing RMD for a given ionic strength. Thus, based on the correlations shown in Fig. 13, all solutions with same ionic strength and same RMD should result in the same hydraulic conductivity regardless of the composition of the solution. Although measured hydraulic conductivity of GCLs with RMD > 0, RMD < 0, and RMD = 0 are shown, the hydraulic conductivity of GCLs with RMD > 0 is generally higher than that of GCLs with RMD < 0 and RMD = 0.

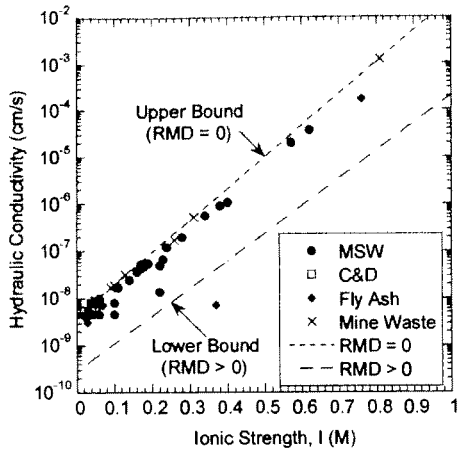


Fig. 13. Correlation between hydraulic conductivity of non-prehydrated GCLs and the ionic strength ( $I$ ) and ratio of concentrations of monovalent-to-divalent cations ( $RMD$ ) in solution for a wide variety of actual leachates (MSW = municipal solid waste, C&D = construction and demolition waste) (from Kolstad, 2000)

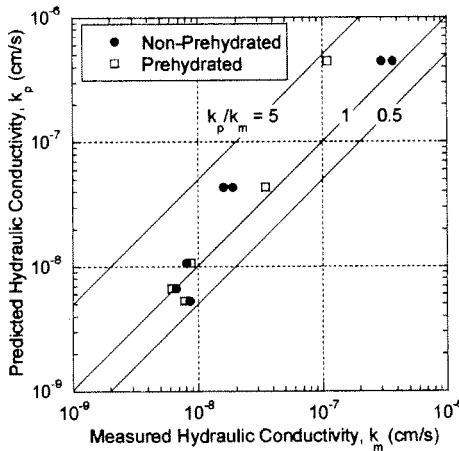


Fig. 14. Measured versus predicted hydraulic conductivities for specimens tested in this study

non-prehydrated and the prehydrated GCL specimens in this study (Table 1) are compared in Fig. 14 with the predicted values,  $k_p$ , using Fig. 13 based on the various ionic strengths for the  $CaCl_2$  solutions and  $RMD = 0$  (i.e., no monovalent cations in solution). As shown in Fig. 14, the predicted hydraulic conductivities are reasonably close to the measured hydraulic conductivities (i.e.,  $5 < k_p/k_m < 0.5$ ), regardless of whether or not the GCL specimens were prehydrated. Thus, the results of this study using only  $CaCl_2$  solutions should be relevant to any practical situation involving leachates with the same  $I$  and  $RMD (=0)$  as the solutions used in this study. Although most real leachates are expected to have  $RMD > 0$ ,  $RMD = 0$  represents the upper bound for hydraulic conductivity (see Fig. 13) such that the assumption of  $RMD = 0$  should be conservative.

SUMMARY AND CONCLUSIONS

The concentration dependency of the prehydration effect on a GCL was evaluated in this study in terms of the hydraulic conductivity ( $k$ ), test duration, and specimen properties resulting from permeating both non-prehydrated and prehydrated specimens with  $CaCl_2$  solutions until chemical equilibrium between influent and effluent was established. This requirement for chemical equilibrium resulted in test durations ranging from 14 hrs to 502 days.

In all cases, permeation with the  $CaCl_2$  solutions resulted in  $k$  values that were greater than the  $k$  to water ( $k_w$ ) of approximately  $2.4 \times 10^{-9}$  cm/s, with the  $k$  of prehydrated specimens ( $k_p$ ) ranging from 2.5 to 49 times greater than  $k_w$ , and the  $k$  of non-prehydrated specimens ( $k_{NP}$ ) ranging from 2.7 to 148 times greater than  $k_w$ . However, prehydration was shown to have little, if any, effect on  $k$  of the GCL when the specimens were permeated with solutions containing from 5 to 50 mM  $CaCl_2$ . In contrast, prehydration had a relatively significant effect on the  $k$  of the GCL permeated with the 100 mM  $CaCl_2$  solution, with  $k_{NP}$  being approximately 3 times greater than  $k_p$ . Thus, the results of this study indicate that the previously reported prehydration effect is concentration dependent, and may be insignificant in terms of  $k$  when the permeant liquid contains relatively low concentrations of inorganic solutes ( $\leq 50$  mM  $CaCl_2$  in this study), i.e., provided permeation is continued until chemical equilibrium is achieved.

In terms of test duration, as the  $CaCl_2$  concentration of the permeant liquid increased for both prehydrated and non-prehydrated specimens, less elapsed time and pore volumes of flow (PVF) were required to establish chemical equilibrium. However, for the tests performed with 5, 10, and 20 mM  $CaCl_2$  solutions, greater test durations (i.e., based on permeation with the  $CaCl_2$  solutions) were required for the non-prehydrated specimens relative to the prehydrated specimens, while the numbers of PVF required to achieve chemical equilibrium for the prehydrated specimens were relatively close to those required for the non-prehydrated specimens. The amounts of  $Ca^{2+}$  delivered to the specimens ( $Ca^{2+}$  mass loadings) per dry mass of bentonite for the tests performed using the 5, 10, and 20 mM  $CaCl_2$  solutions also were essentially the same for the prehydrated and non-prehydrated specimens. However, for the specimens permeated with the 50 and 100 mM  $CaCl_2$  solutions, establishment of chemical equilibrium required shorter test durations and less PVF for the non-prehydrated specimens relative to the prehydrated specimens. Also, significantly greater  $Ca^{2+}$  mass loadings per dry mass of bentonite at chemical equilibrium were required for the prehydrated specimens permeated with the 50 and 100 mM  $CaCl_2$  solutions relative to the non-prehydrated specimens. As a result, the prehydration effect in terms of the test duration, number of PVF, and  $Ca^{2+}$  mass loading per dry mass of bentonite that are required to achieve chemical equilibrium is also shown to be concentration dependent.

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However, the prehydration effect is relatively insignificant in terms of test duration for tests performed using relatively high concentrations of inorganic solutes (50 and 100 mM  $\text{CaCl}_2$  in this study), i.e., because the overall test durations are relatively short. In contrast, the prehydration effect is relatively insignificant in terms of the number of PVF and the  $\text{Ca}^{2+}$  mass loadings per dry mass of bentonite required at chemical equilibrium for the tests performed using the relatively low concentrations of inorganic solutes ( $\leq 20$  mM  $\text{CaCl}_2$  in this study), i.e., provided permeation is continued until chemical equilibrium is achieved.

In terms of final specimen properties, the gravimetric water content ( $w_b$ ), bentonite void ratio ( $e_b$ ), and GCL bulk void ratio ( $e_{bB}$ ) for the prehydrated specimens were always greater than those for the non-prehydrated specimens regardless of the influent  $\text{CaCl}_2$  concentration. However, the overall trend in  $w_b$ ,  $e_b$ , and  $e_{bB}$  for both prehydrated and non-prehydrated GCL specimens as a function of influent  $\text{CaCl}_2$  concentration is similar for both prehydrated and non-prehydrated specimens, such that the lower the influent  $\text{CaCl}_2$  concentration, the greater the change in  $w_b$ ,  $e_b$ , and  $e_{bB}$ . These trends are consistent with greater initial swelling of the bentonite in the presence of de-ionized water (DIW) relative to a  $\text{CaCl}_2$  solution, and greater compression of the adsorbed layers of hydrated cations associated with the bentonite particles with increasing concentration of  $\text{CaCl}_2$  in solution.

Finally, the correlations between  $k$  and the GCL bulk void ratios ( $e_{bB}$ ) for the prehydrated and non-prehydrated specimens are essentially the same in that permeation with a higher  $\text{CaCl}_2$  solution results in a higher hydraulic conductivity and a lower  $e_{bB}$  due to compression of the adsorbed layers of the bentonite particles. However,  $e_{bB}$  for prehydrated specimens (i.e.,  $e_{bB,P}$ ) is always higher than  $e_{bB}$  for non-prehydrated specimens (i.e.,  $e_{bB,NP}$ ) at a given hydraulic conductivity value, indicating a larger void space for the prehydrated specimens with a thicker adsorbed layer resulting from initial hydration with water.

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