Long-Term Hydraulic Conductivity of a Geosynthetic Clay Liner Permeated with Inorganic Salt Solutions

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**Abstract:** Hydraulic conductivity tests were conducted on a geosynthetic clay liner (GCL) for more than 2.5 years and as many as 686 pore volumes of flow (PVF) using single-species salt solutions (NaCl, KCl, or CaCl\(_2\)) to (1) evaluate how the long-term hydraulic conductivity \(K_L\) is affected by cation concentration and valence and (2) demonstrate the relevance and importance of termination criteria when measuring hydraulic conductivity of GCLs to salt solutions. Permeation with CaCl\(_2\) solutions resulted in an increase in the hydraulic conductivity of 1 order of magnitude or more. The rate at which these changes occurred depended on concentration, with slower changes (years and hundreds of PVF) occurring for weaker solutions. In contrast, permeation with 100 mM NaCl or KCl solutions or de-ionized (DI) water resulted in no appreciable change in hydraulic conductivity, regardless of the duration of permeation or number of pore volumes of flow. Hydraulic conductivities determined in accordance with ASTM D 5084 and D 6766 \((K_{5084} \text{ and } K_{6766})\) equaled \(K_L\) when the permeant solution contained NaCl, KCl, or was a strong \((\geq 50 \text{ mM})\) CaCl\(_2\) solution. In contrast, when the permeant liquid was a weak \((\leq 20 \text{ mM})\) CaCl\(_2\) solution, \(K_{6766}\) and \(K_{5084}\) were 2–13 times lower than \(K_L\). Closer agreement between \(K_{6766}\) and \(K_L\) \((3 \times)\) was obtained for weak CaCl\(_2\) solutions when the electrical conductivity ratio criterion was tightened to ±5%. Hydraulic conductivities obtained after comparable influent and effluent concentrations of the permeant salt (±10%) were approximately 2× lower than \(K_L\) for weak CaCl\(_2\) solutions. Hydraulic conductivities equal to \(K_L\) were obtained from the tests permeated with weak CaCl\(_2\) solutions only when Na was no longer eluted at detectable levels.

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**CE Database subject headings:** Bentonite; Compatibility; Chemical equilibrium; Geosynthetics; Hydraulic conductivity; Inorganic chemicals.

**Introduction**

Geosynthetic clay liners (GCLs) are hydraulic barriers that consist of a thin layer of bentonite \((\approx 5–10 \text{ mm})\) sandwiched between two geotextiles or glued to a geomembrane. The GCLs currently available vary by type of geotextile, the inclusion of a geomembrane or not, the presence and type of reinforcement, and the type and gradation of the bentonite (Koerner 1997). When there is no geomembrane, the hydraulic conductivity \(K\) of a GCL is controlled by the hydraulic conductivity of the bentonite, which is directly related to the physical properties (e.g., water content, void ratio, free swell) and chemical composition (e.g., exchangeable cations) of the bentonite (Egloffstein 1997; Shackelford et al. 2000; Jo et al. 2001).

Several studies have addressed how the hydraulic conductivity of GCLs is affected by permeation with inorganic salt solutions (Gleason et al. 1997; Petrov and Rowe 1997; Quanta et al. 1997; Ruhl and Daniel 1997; Shackelford et al. 2000; Egloffstein 2001; Jo et al. 2001; Vasko et al. 2001; Shan and Lai 2002; Kolstad et al. 2004). These studies have shown that permeation with either strong \((\geq 50 \text{ mM})\) solutions or solutions containing a large fraction of polyvalent cations can cause the hydraulic conductivity of GCLs to increase 1 order of magnitude or more. These studies also have shown that the hydraulic conductivity of GCLs permeated with weak \((\leq 20 \text{ mM})\) solutions tends to be comparable to the hydraulic conductivity obtained with de-ionized (DI) or tap water. However, the durations of most tests employing weak solutions typically have been too short \([<0.5 \text{ year and } 10 \text{ pore volumes of flow (PVF)}]\) to establish chemical equilibrium (Shackelford et al. 2000).

Since most field applications of GCLs are intended to last for several decades (or even hundreds of years) where hundreds of PVF may occur, hydraulic conductivities obtained from short-term tests with a modest PVF may not reflect the long-term hydraulic conductivity, i.e., after chemical equilibrium is established. This observation is especially true when GCLs are permeated with weak solutions containing polyvalent cations (e.g., Ca\(^{2+}\)). A key reason why many hydraulic conductivity tests are terminated prematurely is that the test standards commonly used to evaluate hydraulic conductivity (e.g., ASTM D 5084,
ASTM 2002a) do not require chemical equilibrium to be established prior to termination. In fact, only recently (2002) has an ASTM test standard for hydraulic conductivity been developed that includes a criterion for chemical equilibrium (i.e., ASTM D 6766, ASTM 2002b).

This paper discusses findings from a long-term study conducted collaboratively by the Univ. of Wisconsin–Madison (UW) and Colorado State Univ. (CSU) where nonprehydrated GCLs were permeated with inorganic salt solutions for more than 2.5 years and as many as 686 PVF. The objective of the study was to evaluate the long-term temporal behavior of the hydraulic conductivity as a function of concentration and cation valence. This paper describes the hydraulic conductivity of the GCL in the context of the various termination criteria being used for hydraulic conductivity testing and how hydraulic conductivities obtained when satisfying these criteria relate to the long-term hydraulic conductivity at chemical equilibrium.

**Background**

**Effect of Inorganic Solutions on Hydraulic Conductivity of Bentonite**

Sodium bentonite from GCLs is composed primarily of the mineral Na–montmorillonite, which is a member of the smectite family. Na–montmorillonite has a large specific surface, a charge deficiency of 0.5–1.2 esu per unit cell, high cation exchange capacity (≈80–150 meq/100 g), and the ability for interlayer swelling (Mitchell 1993). The prefix Na is used because Na⁺ ions are the predominant cations in the exchange complex of Na–montmorillonite.

The hydraulic conductivity of bentonite changes in accordance with the volume of water bound to the mineral surface, which is affected by concentration, cation valence, and pH of the permeant solution (Mitchell 1993; McBride 1994; Egloffstein 1995; Petrov and Rowe 1997; Shackelford et al. 2000; Jo et al. 2001; Kolstad et al. 2004). The Na⁺ ions initially existing on and between the montmorillonite layers can be replaced by multivalent cations (e.g., Ca²⁺ and Mg²⁺) in the permeant solution, a process referred to as cation exchange (Grim 1968; van Olphen 1991; McBride 1994). The rate at which the ion exchange process occurs is controlled primarily by the rate at which cations diffuse into and out of the interlayer region between the montmorillonite layers (McBride 1994; Jo et al. 2004).

When multivalent cations replace the Na⁺ ions, the volume of water bound to the montmorillonite surface (i.e., immobile water) decreases and the volume of free (mobile) water increases (van Olphen 1991; McBride 1994), resulting in shrinkage of montmorillonite aggregates. Under modest confinement, this change in aggregate size results in larger interaggregate pores (i.e., the pores conducting flow) and higher hydraulic conductivity (Mersi and Olson 1971; Jo et al. 2001). Conversely, if the permeant solution contains only monovalent cations, or if there is no cation exchange, the volume of bound water increases or decreases depending on the cation concentration in the permeant solution. This effect causes expansion or contraction of the montmorillonite aggregates and interaggregate pores, resulting in changes in hydraulic conductivity. A variety of investigators studying the hydraulic conductivity of GCLs to salt solutions have observed these effects (Petrov and Rowe 1997; Shackelford et al. 2000; Egloffstein 2001; Jo et al. 2001; Vasko et al. 2001).

**Long-Term Hydraulic Conductivity Tests with Inorganic Solutions**

Imamura et al. (1996) conducted hydraulic conductivity tests on sand–bentonite mixtures (15% Na–bentonite) for more than 2 years. Specimens were permeated with DI water or a 15 mM Ca(OH)₂ solution. The hydraulic conductivity to DI water (7.0 × 10⁻¹⁰ cm/s) remained stable for 4 year of permeation. Hydraulic conductivities of the three specimens permeated with the 15 mM Ca(OH)₂ solution initially were comparable to that obtained with DI water. However, hydraulic conductivities of these specimens gradually increased by 2 orders of magnitude between 0.5 and 2.2 years of permeation. Imamura et al. (1996) show that the changes in hydraulic conductivity corresponded to exchange of Ca²⁺ for Na⁺ on the exchange complex.

Shackelford et al. (2000) describe a hydraulic conductivity test performed on a GCL permeated with a 12.5 mM CaCl₂ solution for more than 1.4 years and 50 PVF. This test was continued by the authors, and ultimately was terminated after 4.4 years and 1,100 PVF. The hydraulic conductivity initially was 1.3 × 10⁻⁹ cm/s, and no change in hydraulic conductivity occurred until 5 PVF (220 days). After 5 PVF, the hydraulic conductivity increased gradually to 2.6 × 10⁻⁸ cm/s after approximately 200 PVF (1.6 years). Essentially no further change in hydraulic conductivity occurred during the remainder of the test (i.e., 1,100 PVF and 4.4 years).

Egloffstein (2001) conducted hydraulic conductivity tests on two GCLs having Na–bentonite and different mass per unit area (i.e., 4.7 and 8.0 kg/m²). The GCLs initially were hydrated with DI water, and then were permeated with solutions representing pore water in a natural soil. Egloffstein (2001) did not cite the composition of the permeant solution, but indicated that the Ca concentration in the pore water of natural soils is comparable to that in a 15 mM CaCl₂ solution. The hydraulic conductivity was initially 2.0 × 10⁻⁹ cm/s for the GCL having lower mass per unit area (4.7 kg/m²) and 4.0 × 10⁻⁹ cm/s for the GCL having higher mass per unit area (8.0 kg/m²). No change in the hydraulic conductivity occurred for either specimen during the first year of permeation. The hydraulic conductivities then increased gradually to 3.0 × 10⁻⁸ cm/s after approximately 1.4 years (mass per unit area=4.7 kg/m²) or 1.0 × 10⁻⁸ cm/s after approximately 2.7 years (mass per unit area=8.0 kg/m²). Egloffstein (2001) attributed the increase in hydraulic conductivity to gradual cation exchange of Ca for Na, but measurements describing the exchange complex were not provided.

**Termination Criteria**

Several investigators have discussed termination criteria for hydraulic conductivity tests conducted to evaluate interactions between barrier soils and permeant solutions (Dunn and Mitchell 1984; Peirce and Witter 1986; Bowders 1988; Daniel 1994; Shackelford 1994; Shackelford et al. 2000). These studies generally recommend that tests not be terminated until steady hydraulic conductivity is achieved, the ratio of incremental outflow to inflow (referred to herein as the “flow ratio”) is approximately unity, and at least one or two PVF have passed through the specimen. Peirce and Witter (1986) suggest a statistical method that can be used to determine when steady hydraulic conductivity has been achieved. In addition, Bowders (1988), Daniel (1994), and Shackelford (1994) recommend that chemical equilibrium be established before a test is terminated. Comparable concentration in the influent and effluent (e.g., ±10%) generally is assumed to
D. Shackelford et al. (1999) suggest electrical conductivity (EC) and pH of the effluent and influent can be used as indicators of chemical equilibrium, and recommend that the ratios of effluent-to-influent EC and effluent-to-influent pH fall within 1.0±0.25 for four consecutive hydraulic conductivity measurements. Shackelford et al. (1999) indicate that chemical equilibrium has been established, but consensus on the definition of chemical equilibrium has not been established. Shackelford et al. (1999) suggest electrical conductivity (EC) and pH of the effluent and influent can be used as indicators of chemical equilibrium, and recommend that the ratios of effluent-to-influent EC and effluent-to-influent pH fall within 1.0±0.25 for four consecutive hydraulic conductivity measurements. Steady hydraulic conductivity is defined as no temporal trend evident in a tabulation of the hydraulic conductivity measurements. Steady hydraulic conductivity is defined as no temporal trend evident in a tabulation of the hydraulic conductivity measurements. That is, CaCl₂ solutions are representative of the behavior of solutions containing divalent cations in general.

### Materials and Methods

#### Geosynthetic Clay Liner

The GCL used in this study contained granular sodium bentonite encased between a slit-film monofilament woven geotextile (130 g/m²) and a staple-fiber nonwoven geotextile (268 g/m²). The geotextiles are joined by needle-punching fibers locked in place by thermal burnishing. The initial thickness of the air-dry specimens tested in this study ranged between 5.5 and 6.5 mm.

The mass per unit area of air-dry bentonite (ASTM D 5993, ASTM 2002b) in the GCL was 4.8±0.8 kg/m² (eight tests), the average initial gravimetric water content of the bentonite was 9.0±1.1% (five tests), and the specific gravity of bentonite solids (ASTM D 854, ASTM 2002a) was 2.74±0.04 (nine tests). Results of x-ray diffraction showed that the bentonite consists primarily of montmorillonite (78±5%) (five tests). Mechanical sieve and sedimentation analyses indicated that the dry bentonite consists of sand-size granules ranging in size from 0.05 to 2.0 mm and that the fraction of clay-size particles (<0.002 mm) is approximately 90%. The liquid limit of the bentonite is 479±90 (three tests) and the plasticity index is 441±86 (three tests) (ASTM D 4318, ASTM 2002a), both of which are comparable to Atterberg limits reported by Mesri and Olson (1971) for Na₂-bentonite.

The cation exchange capacity of the bentonite (determined by the ammonium acetate method, Rhodes 1982a) ranges between 53 and 75 meq/100 g (seven tests). The pH and EC of the bentonite (measured on pastes prepared with DI water, Rhodes 1982b) are 8.9±0.1 (four tests) and 300±16 mS/m (four tests). The exchange complex (determined by the ammonium acetate method, Thomas 1982) is dominated by Na⁺ and has the following composition (eight tests): Na—43.8±16.8 meq/100 g, Ca—16.5±5.4 meq/100 g, Mg—6.0±3.4 meq/100 g, and K—0.8±0.4 meq/100 g.

### Permeant Solutions

Chemical properties of the permeant solutions used in this study are shown in Table 1. De-ionized water was used as a reference solution. NaCl and KCl solutions were used to investigate the effect of hydrated radius (rₜ) of monovalent cations (i.e., rₜ ≈ 0.46 nm for K⁺ and ≈ 0.68 nm for Na⁺, Mitchell 1993). CaCl₂ solutions were used to investigate the effect of cation concentration and valence. Only CaCl₂ solutions were used to represent solutions containing divalent cations because Jo et al. (2001) and Kolstad et al. (2004) show that, at a given concentration, species of divalent cation has no effect on free swell or hydraulic conductivity. That is, CaCl₂ solutions are representative of the behavior of solutions containing divalent cations in general.

### Table 1. Properties of Permeant Solutions

<table>
<thead>
<tr>
<th>Compound used</th>
<th>Compound source and purity</th>
<th>Concentration (mM)ᵃ</th>
<th>Meanᵈ</th>
<th>σᵉ</th>
<th>Mean</th>
<th>σᵉ</th>
<th>Mean</th>
<th>σᵉ</th>
</tr>
</thead>
<tbody>
<tr>
<td>NaCl</td>
<td>SIGMA Co. 99.6%</td>
<td>100</td>
<td>2,261</td>
<td>14.9</td>
<td>6.2</td>
<td>0.2</td>
<td>1,160</td>
<td>13.2</td>
</tr>
<tr>
<td>KCl</td>
<td>SIGMA Co. 99.9%</td>
<td>100</td>
<td>3,883</td>
<td>10.2</td>
<td>6.3</td>
<td>0.2</td>
<td>1,291</td>
<td>8.5</td>
</tr>
<tr>
<td>CaCl₂</td>
<td>SIGMA Co. 96.0%</td>
<td>5</td>
<td>209</td>
<td>2.8</td>
<td>6.7</td>
<td>0.7</td>
<td>119</td>
<td>2.1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>10</td>
<td>412</td>
<td>4.8</td>
<td>6.6</td>
<td>0.7</td>
<td>235</td>
<td>5.3</td>
</tr>
<tr>
<td></td>
<td></td>
<td>20</td>
<td>848</td>
<td>17.7</td>
<td>6.5</td>
<td>0.6</td>
<td>437</td>
<td>7.7</td>
</tr>
<tr>
<td></td>
<td></td>
<td>50</td>
<td>1,985</td>
<td>8.1</td>
<td>6.5</td>
<td>0.7</td>
<td>960</td>
<td>8.4</td>
</tr>
<tr>
<td></td>
<td></td>
<td>100</td>
<td>3,981</td>
<td>10.7</td>
<td>6.2</td>
<td>0.7</td>
<td>1,890</td>
<td>9.0</td>
</tr>
<tr>
<td></td>
<td></td>
<td>500</td>
<td>19,400</td>
<td>9.0</td>
<td>6.9</td>
<td>0.7</td>
<td>9,720</td>
<td>9.0</td>
</tr>
<tr>
<td>DI water</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>0.04 (UW)</td>
<td>0.2 (CSU)</td>
</tr>
</tbody>
</table>

ᵃMeasured by atomic absorption spectrometer.
ᵇMeasured by pH meter.
ᶜMeasured by electrical conductivity probe.
ᵈTemporal mean over duration of study.
ᵉStandard deviation over duration of study.
ᶠTesting period too short to report temporal mean of cation concentration, pH, and EC of stock solutions.
The solutions were prepared by dissolving powdered salts in DI water. The DI water used at UW classified as Type II water per ASTM D 1193 (ASTM 2002c), whereas the DI water at CSU classified as Type IV water. These DI waters differ slightly in terms of EC (EC < 0.1 mS/m for Type II, < 0.5 mS/m for Type IV). However, as shown subsequently, comparable hydraulic conductivities were obtained from the tests conducted at UW and CSU using DI water. Thus, the subtle differences in type of DI water had no measurable effect on hydraulic conductivity.

The CaCl₂ solutions had concentrations ranging between 5 and 500 mM and the KCl and NaCl solutions had a concentration of 100 mM. The CaCl₂ solutions were selected to bracket the range of concentrations typically found in solid waste facilities and surface impoundments used for storing aqueous solutions (Kolstad et al. 2004). The NaCl and KCl solutions were used because short-term tests (0.2 year and 10 PVF) conducted by Jo et al. (2001) with these solutions yielded hydraulic conductivities comparable to those obtained with DI water when terminated in accordance with the pH and EC criteria in ASTM D 6766. Tests were conducted with the 100 mM CaCl₂ solutions for direct comparison with 100 mM NaCl and KCl solutions to assess the effects of valence. The 500 mM CaCl₂ solution was used as a very strong solution. Jo et al. (2001) show that very strong (≥ 100 mM) CaCl₂ solutions result in rapid equilibrium and high hydraulic conductivity (10⁻⁵–10⁻⁶ cm/s) of nonprehydrated GCLs.

A 20 L carboy of each solution was prepared at the onset of the study to provide a consistent permeant solution for use throughout the testing program. To evaluate whether the chemical properties of the permeant solutions changed during the study, pH, EC, and salt concentrations were measured using a pH meter (Accumet pH meter 50, Fisher Scientific Co.), an electrical conductivity probe (Con 5 series, Cole–Parmer Instrument Co.), and by flame atomic absorption (FAA) SpectrAA 800, Varian Corp.). Typical results from these tests are shown in Fig. 1 as a function of time. The pH of the permeant solutions decreased with time, probably due to dissolution of atmospheric CO₂. However, the EC and cation concentrations remained constant. For all solutions, the coefficient of variation was no more than 2.2% for EC and 2.1% for concentration.

The change in pH that occurred during the study period probably is unimportant because the pH remained near neutral (5.8–7.3). In this pH range, the solubility of the salts that were used is essentially constant (Stumm and Morgan 1996), as is evident by the uniform EC and concentration records. Shackelford (1994), Ruhl and Daniel (1997), and Jo et al. (2001) also report that pH only affects the hydraulic conductivity of clays when the pH is < 2 or >12, and Shan and Lai (2002) found that the hydraulic conductivity of GCLs permeated with HCl solutions having pH 5 was comparable to the hydraulic conductivity of GCLs permeated with DI water having pH 7 (3.0×10⁻⁹ cm/s).

**Hydraulic Conductivity Tests**

The hydraulic conductivity tests were conducted in flexible-wall permeameters using the falling headwater-constant tailwater procedure (Method B in ASTM D 5084). No backpressure was applied so that effluent samples for chemical analyses (i.e., pH, EC, and salt concentrations) could be conveniently collected. However, gravimetric calculations made at the end of testing indicated that the specimens were saturated, at least within the accuracy with which degree of saturation can be computed (±5%). The cell and influent pressures were applied using gravity reservoirs to eliminate external effects due to stress fluctuations (e.g., pressure changes due to thermal and temporal effects on pressure regulators and compressed gas supplies). Conventional latex membranes were used. Examination at the end of the tests indicated that long-term testing had no discernible effect on the membranes.

GCL specimens were prepared following the procedure described in Jo et al. (2001), which consists of trimming the specimen from a GCL panel using a cutting ring and sharp razor knife. Permeant solution was applied lightly around the edge of the specimen during trimming to prevent loss of bentonite. After placement in the permeameter, specimens were hydrated with the permeant solution for 48 h under an effective confining stress of approximately 20 kPa without application of a hydraulic gradient. No prehydration fluid such as DI or tap water was used so that field conditions could be replicated as reasonably as possible. Although some prehydration can occur in the field due to capillary conduction or vapor-phase diffusion, Vasko et al. (2001) show that this type of prehydration has little effect on changes in hydraulic conductivity caused by permeation with salt solutions.

Hydraulic conductivity tests then were initiated by opening the outflow valve and were continued until at least all of the following criteria were met: (1) steady hydraulic conductivity (i.e., no statistically significant trend in hydraulic conductivity over time and at least four consecutive hydraulic conductivity values within ±25% of the mean per D 5084); (2) at least four consecutive flow ratios within 1.0±0.25; and (3) chemical equilibrium established. Most tests were continued after these termination criteria were met. After termination, the exchange complex of the bentonite was determined using the same method employed when testing the unused air-dry GCL.
Trend in the hydraulic conductivity data was analyzed statistically using the method in Peirce and Witter (1986) at a significance level of 0.05. This method consists of analyzing the slope of a moving series of ten sequential hydraulic conductivity measurements using linear regression. When the slope of the regression is statistically no different from zero [i.e., defined using a t-test as described in Draper and Smith (1998)], the hydraulic conductivity data are deemed to no longer exhibit trend.

Chemical equilibrium was defined based on the EC ratio criterion in ASTM D 6766 (1.0±0.1), equality of the influent and effluent salt concentration, and Na concentrations in the effluent falling below the method detection limit (MDL) for the chemical analysis method that was used. The pH ratio criterion in ASTM D 6766 (1.0±0.1) was not used because the pH of the permeant solutions decreased over time resulting in an increase in pH ratio (Figs. 1 and 7–9). The MDL represents the highest concentration that is statistically different from zero for a given chemical analysis method (Berthouex and Brown 2002). That is, measured concentrations falling below the MDL are statistically no different from zero, even though nonzero concentrations are obtained from the analytical method. The MDL for Na was 0.2 mg/L for the tests conducted at UW and CSU. The FAA spectrometry was used for analysis at UW, whereas inductively coupled plasma–atomic emission spectrometry was used at CSU. All tests conducted with CaCl₂ solutions at UW met the MDL criterion except for one test permeated with 5 mM CaCl₂ solution, in which the Na concentration in the effluent was slightly above the MDL (0.4 mg/L versus MDL=0.2 mg/L). The tests conducted at CSU did not meet the MDL criterion due to clogging problems, which are discussed subsequently. In addition, the EC and MDL criteria were not met for the tests conducted with DI water. After more than 2.5 years, Na⁺ was still eluted from the GCLs permeated with DI water. This slow elution may have been caused in part by slow exchange of H⁺ for Na⁺ on the mottorrillonite surface (Gilbert and Laudelout 1965).

Duplicate hydraulic conductivity tests (labeled A and B) were conducted for each permeant solution, and tests with the CaCl₂ solutions were replicated at UW and CSU. The same methods were used at UW and CSU, with the following exceptions. The average effective stresses used at UW and CSU differed slightly (16.2 kPa at UW and 23.5 kPa at CSU), as did the average hydraulic gradient (130 at UW and 200 at CSU). Although these hydraulic gradients are higher than those specified in ASTM D 5084, they are typical of hydraulic gradients used when testing GCLs (Shackelford et al. 2000). The size of tubes used for the flow lines in the permeameters also differed between UW and CSU. Tubes with a diameter of 3.2 mm were used at CSU,
whereas 6.4 mm diameter tubes were used at UW.

The replicate intralaboratory tests yielded hydraulic conductivities that varied at most by a factor of 1.6 and always were within 30% of the mean, indicating good intralaboratory reproducibility [Fig. 2(a)]. Similar reproducibility is reported by Daniel et al. (1997) and Petrov et al. (1997). For example, the round-robin study conducted by Daniel et al. (1997) included 17 laboratories with each laboratory conducting four replicate hydraulic conductivity tests at the same effective stress and with the same permeant solution. Intralaboratory hydraulic conductivities in their study differed no more than 30% from the mean hydraulic conductivity. Petrov et al. (1997) conducted four replicate hydraulic conductivity tests on GCLs using DI water as the permeant solution, and reported hydraulic conductivities deviating no more than 22% from the mean.

A comparison of the interlaboratory tests is shown in Fig. 2(b). The hydraulic conductivities measured at UW tend to be higher (at most a factor of 2.8) than those measured at CSU [Fig. 2(b)]. The difference in hydraulic conductivities measured at UW and CSU is due in part to the slight difference in average effective stress that was used. For a similar difference in effective stress, Petrov et al. (1997) and Shackelford et al. (2000) show that the hydraulic conductivity of GCLs typically varies by a factor of approximately 2. Clogging also may have contributed to the differences in hydraulic conductivity, as described subsequently.

A comparison of the temporal variation in hydraulic conductivity measured at CSU and UW for the tests conducted with weak (<=20 mM) CaCl₂ solutions is shown in Fig. 3. The hydraulic conductivities measured at UW are higher for nearly all tests, again probably due to the lower effective stress used at UW. When this bias is ignored, the temporal trends obtained at UW and CSU for the tests conducted with the 5 mM CaCl₂ solution are very similar. However, different trends were observed for higher concentrations (i.e., 10 and 20 mM). After the hydraulic conductivity initially climbed, the hydraulic conductivities measured at UW continued to increase slightly, whereas the hydraulic conductivities measured at CSU decreased significantly [Figs. 3(b and c)]. The decrease in hydraulic conductivity at CSU was caused by salt precipitates clogging the flow system. Similar precipitates were present in the outflow tubing at UW. However, the precipitates only affected the tests at CSU because of the smaller tubes (i.e., 3.2 mm in diameter) in the CSU permeameters and when the salt concentration was 10 mM or higher (i.e., when more ions were available to form precipitates). Removing the precipitate at CSU caused the hydraulic conductivity to jump approximately 1 order of magnitude [Figs. 3(b and c)]. However, the clogging gradually reappeared, confounding further interpretation of the tests. Some of the differences between the hydraulic conductivities measured at UW and CSU [Fig. 2(b)] probably are due to this clogging effect. These observations suggest that equipment used for long-term hydraulic conductivity testing must be designed to limit the potential for clogging, and that the results of long-term tests be scrutinized to ensure that clogging has not affected the measurements.

There was also concern that the hydraulic conductivity might be affected by biological activity due to the long duration of the tests (Dennis and Turner 1998; Kamon et al. 2002). To assess if biological activity affected the hydraulic conductivities, a hydraulic conductivity test was conducted on a GCL specimen permeated with a 5 mM CaCl₂ solution spiked with 500 ppm of the biocide DOWICIL QK-20, which has 2,2-dibromo-3-nitropropanamide as the active ingredient. Nelson (2000) found that this biocide eliminates biological activity during hydraulic conductivity testing of paper sludges, which typically have abundant microbial populations (Benson and Wang 2000). Nelson (2000) indicates that DOWICIL concentrations between 500 and 2,000 ppm are effective, and do not induce physicochemical effects that alter hydraulic conductivity.

Hydraulic conductivities of GCLs permeated with 5 mM CaCl₂ solution with and without the biocide are shown in Fig. 4. The data for two of the three tests are essentially identical, regardless of whether biocide was used, and the hydraulic conductivities for all three tests are very similar. Thus, biological activity probably was minimal, and likely had no effect on the hydraulic conductivities that were measured.

Results and Analysis

Results of the hydraulic conductivity tests are summarized in Table 2. Hydraulic conductivities are defined at seven points during a test: (1) when the termination criteria in ASTM D 5084 were satisfied (\(K_{5084}\)); (2) when the termination criteria in ASTM D 6766 were satisfied (\(K_{6766}\)); (3) when the criteria in D 6766 were satisfied and the EC ratio fell within ±0.05, which is a tighter criterion than in D 6766 (\(K_{EC}\)); (4) when the concentration of the permeant salt in the influent and effluent differed by less than 10% (\(K_{p10}\)); (5) when the concentration of the permeant salt in the influent and effluent differed by less than 5% (\(K_{p5}\)); (6) when the Na concentration in the effluent fell below the MDL (\(K_{Na}\)); and (7) when the tests were actually terminated (\(K_t\)). In addition to hydraulic conductivity, PVF and permeation time are reported for each point at which the hydraulic conductivity is defined.

The hydraulic conductivity \(K_{5084}\) in Table 2 is regarded as the “short-term hydraulic conductivity” because the termination criteria in D 5084 were typically reached in less than 0.2 year (Table 2, Figs. 5–9). The hydraulic conductivity \(K_t\) is regarded as the “long-term hydraulic conductivity” because this hydraulic conductivity corresponds to the end of testing. The hydraulic conductivities \(K_{6766}\), \(K_{EC}\), \(K_p\), and \(K_{Na}\) were obtained at times between \(K_{5084}\) and \(K_t\).
Table 2. Summary of Data from Hydraulic Conductivity Tests

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<th>Permeant solution</th>
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**Note:** $5$ mM CaCl$_2$ solution spiked with 500 ppm of the biocide DOWICIL QK-20.
Temporal Behavior

Temporal characteristics of the hydraulic conductivity, the pH and EC ratios, and the cation concentrations in the effluent are shown in Figs. 5–9 for GCLs permeated with DI water and the NaCl, KCl, and CaCl₂ solutions. The vertical dashed lines in these graphs correspond to points during each test when the various termination criteria were reached.

Only a small change (~2×) in hydraulic conductivity occurred during the long test duration (>2.3 years) when DI water was used as the permeant solutions (Fig. 5). The hydraulic conductivity was initially 1.7×10⁻⁹ cm/s and increased by at most a factor of 1.8 by the end of testing. However, nearly all of the increase in hydraulic conductivity occurred due to a sudden jump around 0.5 year and 9 PVF (Fig. 5). The reason for this jump is unknown. However, physical mechanisms are more likely the cause (e.g., a subtle and unnoticed change in test setup) rather than chemical mechanisms, because no abrupt changes in pH, EC, or concentration occurred around the same time. Also, no piping of bentonite was observed. Thus, from a practical perspective, no change in hydraulic conductivity occurred during the long-term tests conducted with DI water.

The EC ratio remained high throughout the tests with DI water due to the very low EC of the influent (0.04 mS/m) and the gradual elution of Na (reflected in the elevated Na concentration in the effluent). The slow elution of Na⁺ may have been due to exchange of H⁺ for Na⁺ (Gilbert and Laudelout 1965), which occurs very slowly because the concentration of H⁺ in DI water is very low (~0.0003 mM in this case).

Hydraulic conductivities obtained with 100 mM NaCl and KCl solutions were comparable to those obtained using DI water, and varied by at most a factor of 1.8 throughout the test duration (Fig. 6). For the 100 mM KCl solution, the EC ratio was 1.0±0.1 after 4 PVF and the hydraulic conductivity was stable (i.e., K₅₀₈₄ and K₆₇₆₆ ~4.0×10⁻⁹ cm/s). However, even though the termination criteria in ASTM D 6766 were met, the Na concentration in the effluent remained elevated, and the K concentration remained below that of the influent, indicating that chemical equilibrium had not yet been established and cation exchange likely was still occurring. In fact, 12 PVF (0.3 year) were required before the influent and effluent K concentrations were within 10%, and 140 PVF (2.7 year) were required before the Na concentration reached the MDL and the influent and effluent K concentrations were the same (~1%). At the end of testing (2.7 years), the Na concentrations remained below the MDL and the K concentration was equal to that of the influent.

The exchange complex of the bentonite after permeation with DI water was comparable to that of bentonite in a new GCL (i.e., mole fraction of sodium on the exchange complex Xₐₙₒ =0.7, mole fraction of calcium on the exchange complex Xₐ₅ =0.2). A similar condition was observed for the tests with 100 mM NaCl solution (i.e., Xₐₙₒ =0.8, Xₐ₅ =0.15), even after approximately 3 years permeation. In contrast, for the KCl solution, exchange of K⁺ for Na⁺ was essentially complete (Xₐ₈₀ =0, mole fraction of potassium on the exchange complex, Xₐ₈₀ =1.0) at the end of testing (~2.5 years). Despite these chemical changes, the K₅ obtained with DI water, the KCl solution, and the NaCl solution differed at most by a factor of 1.3 relative to K₅₀₈₄ and K₆₇₆₆ (Table 2), indicating that exchange of Na⁺ by K⁺ had no practical effect on the hydraulic conductivity even though Na and K have different hydrated radii. Petrov and Rowe (1997) and Jo et al. (2001) also report that the hydraulic conductivity to weak solutions containing monovalent cations obtained from short-term tests (<3 months and 10 PVF) is essentially the same as that to DI water, regardless of cation species.

For weak CaCl₂ solutions (i.e., ≤20 mM), the hydraulic conductivity initially stabilized at approximately 2.0×10⁻⁹ cm/s (i.e., K₅₀₈₄ after 3–15 PVF (~0.2 year) had passed through the GCL (Figs. 7–9). This is essentially the same hydraulic conductivity as obtained with DI water (3.0×10⁻⁹ cm/s). At this time, the influent and effluent EC and chemical composition still were appreciably different (Figs. 7–9). After approximately 20–50 PVF, the EC ratio typically stabilized at 1.0±0.1 (i.e., as required in D 6766), but the hydraulic conductivity continued to increase at a slow and statistically significant rate. After 60 PVF, Ca concentrations in the influent and effluent were within 10%, but the Na concentration in the effluent (i.e., 30–500 mg/L) remained above that of the influent (0 mg/L).

The hydraulic conductivity did not become stable (i.e., statistically insignificant trend) until 360 PVF, when the Na concentration dropped below the MDL (Figs. 7–9). Even after the ion concentrations in the effluent and influent were equal, the pH in the effluent still remained above that in the influent, probably due to the gradual change in influent pH over time (Fig. 1). These gradual changes in hydraulic conductivity are similar to those reported by Shackelford et al. (2000) and Egloffstein (2001), as described previously. Exchange complex measurements made after testing showed that exchange of Ca²⁺ for Na⁺ was essentially complete (Xₐ₈₀ =0, Xₐ₅ =1.0) at the end of testing, regardless of concentration of the permeant solution. Thus, by the end of testing, chemical interactions between the permeant liquid and the bentonite were effectively complete.
The hydraulic conductivity of GCLs permeated with solutions having high concentration (\(\geq 50 \text{ mM}\)) was approximately 2 orders of magnitude higher than the hydraulic conductivity of GCLs permeated with DI water (Table 2). Equilibrium was reached quickly (<1 day) for these tests because the high Ca concentration results in rapid exchange for Na, which results in high hydraulic conductivity. A compilation of all of the data from the study can be found in Jo (2003) and Lee (2004).

**Comparison of Hydraulic Conductivities**

Hydraulic conductivity ratios relating \(K_L\) to the hydraulic conductivity defined by each of the termination criteria (\(K_{5084}\), \(K_{6766}\), \(K_{EC}\), \(K_{p10}\), \(K_{p5}\), and \(K_{Na}\)) are summarized in Table 2. Comparison of these ratios indicates that the hydraulic conductivities obtained from the six termination criteria are essentially the same for tests conducted with the solutions containing monovalent cations, and the strong solutions (\(\geq 50 \text{ mM}\)) containing divalent cations. For these solutions, the short-term (\(K_{5084}\)) and long-term (\(K_L\)) hydraulic conductivities differ at most by a factor of 1.8, which is within the reproducibility of the tests.

For weak solutions (\(\leq 20 \text{ mM}\)) containing divalent cations, \(K_L\), \(K_{5084}\), \(K_{6766}\), \(K_{p10}\), and \(K_{p5}\) can be appreciably different. The \(K_L\) for the weak divalent solutions ranges from 2 to 13 times \(K_{5084}\), with \(K_L\) being on average 7.3 times higher than \(K_{5084}\). The \(K_L\) and \(K_{6766}\) are closer, differing on average by a factor of 5.3, but can differ by as much as a factor of 13 (Table 2). In contrast, \(K_{EC}\) and \(K_{Na}\) are very close to \(K_L\) (at most a factor of 2.2 for \(K_{p10}\), 1.5 for \(K_{p5}\), and 1.0 for \(K_{Na}\)).

The ratios \(K_L/K_{5084}\) and \(K_L/K_{6766}\) are shown in Fig. 10 as a function of concentration. The ratio \(K_L/K_{5084}\) is nearly insensitive to concentrations below 10 mM (\(K_L/K_{5084} = 2.0–13.0\)) and above 50 mM (\(K_L/K_{5084} = 1.0–1.4\)), although appreciable scatter exists for concentrations below 20 mM. For concentrations between 10 and 50 mM, \(K_L/K_{5084}\) decreases as the concentration increases. Similar sensitivity to concentration exists for \(K_L/K_{6766}\) although the ratio is much smaller than \(K_L/K_{5084}\). The ion exchange that occurs between the points when the termination criteria in \(D 5084\) and \(D 6766\) are met results in conditions closer to equilibrium, and less difference between \(K_L\) and \(K_{6766}\) than exists between \(K_L\) and \(K_{5084}\).

The ratios \(K_L/K_{5084}\) and \(K_L/K_{6766}\) are insensitive to concentration when the concentration is low (\(\leq 20 \text{ mM}\)) because weak divalent solutions yield short-term hydraulic conductivities (i.e., \(K_{5084} \sim 2.0 \times 10^{-9} \text{ cm/s}\)) that vary by at most a factor of 2, intermediate-term hydraulic conductivities (i.e., \(K_{6766} \sim 8.0 \times 10^{-9} \text{ cm/s}\)) that vary by at most a factor of 3, and long-term
hydraulic conductivities ($K_f \sim 2.0 \times 10^{-8}$ cm/s) that vary by at most a factor of 2. That is, although the hydraulic conductivity varies with time due to ion exchange, the magnitude of the hydraulic conductivity determined using each of the termination criteria is insensitive to concentration when the concentration is less than 20 mM. Jo et al. (2001) report similar findings for weak solutions containing divalent cations. In their short-term tests (i.e., less than 6 months), they found that the hydraulic conductivity to solutions containing divalent cations varied by at most a factor of 2 when the concentration was less than 10 mM or greater than 100 mM. Similarly, $K_f/K_{5084}$ and $K_f/K_{6766}$ are also insensitive to concentration for strong solutions ($\geq 50$ mM). However, in contrast to the tests conducted with weak solutions, strong solutions result in a rapid increase in hydraulic conductivity and chemical equilibration by the time the short-term criteria are met. Consequently, the short-term ($K_{5084}$), intermediate-term ($K_{6766}$), and long-term hydraulic conductivities ($K_f$) are comparable (i.e., $\sim 1.0 \times 10^{-6}$ cm/s), differing by at most a factor of 2 (Table 2 and Fig. 10).

**Assessment of Termination Criteria**

The data in Fig. 10 and Table 2 illustrate that chemical interactions are not necessarily complete when the termination criteria in

**ASTM D 5084 and D 6766** are met, particularly when solutions containing divalent cations with low concentrations ($\leq 20$ mM) are used as the permeant solution. Thus, what criteria should be applied if the long-term equilibrium hydraulic conductivity is to be measured? The data in Table 2 suggest that suitable criteria should include: (1) no statistically significant trend in hydraulic conductivity over time; (2) equality of the permeant salt concentration in the influent and effluent (e.g., within 5%); and (3) effluent that is free of ions initially in the exchange complex that are not in the permeant solution (e.g., Na concentration below the method detection limit for the tests using Ca solutions in this study, which is referred to henceforth as “no Na elution”). Hydraulic conductivities measured after meeting these criteria are shown in Fig. 11 along with $K_f$. All of the hydraulic conductivities meeting these criteria fall close to the 1:1 line, indicating that they are representative of $K_f$.

Measuring the chemical composition of the influent and effluent precisely may be impractical, and meeting a no Na-elution criterion can require extremely long testing durations. For example, at Ca concentrations $\leq 20$ mM, meeting the no Na-elution criterion may require more than 1.6 years and 300 PVF (Table 2). An alternative is to reduce the tolerance in EC ratio to more precisely reflect differences between the composition of the influ-
Summary and Conclusions

Hydraulic conductivity tests were conducted in duplicate on GCLs for more than 2.5 years at two institutions using salt solutions in a climate-controlled laboratory. Statistical analysis of the EC data collected in this study suggests that the lower limit on the tolerance is ±0.05 using conventional EC probes calibrated with standard solutions. However, there is a practical lower limit on EC due to measurement scatter. A comparison of $K_L$ and $K_{EC}$ corresponding to an EC ratio of 1.0±0.05 is shown in Fig. 12(a). Even with the lower tolerance, $K_L$ generally is higher than $K_{EC}$. However, the ratio $K_L/K_{EC}$ is low (≤3.0) and the times and PVF required to meet the tighter EC ratio criterion are appreciably lower (although the testing times are still long) than those required to ensure complete chemical equilibrium (Table 2). For example, the hydraulic conductivity of the “UW-a” specimen permeated with the 10 mM CaCl$_2$ solution increased by only a factor of 1.3 between 40 PVF or 0.3 year (i.e., EC ratio=1.0±0.05) and 373 PVF or 1.8 years (i.e., Na concentration < MDL) (Fig. 8 and Table 2). Thus, a more practical approach may be to terminate tests once the EC ratio is within 1.0±0.05, and acknowledge that the long-term hydraulic conductivity may be as much as three times higher than the reported hydraulic conductivity. Similarly, if tests are terminated when the EC ratio is within 1.0±0.1 (as in $D_{6766}$), then the hydraulic conductivity could be acknowledged to be as much as 10 times higher than the reported hydraulic conductivity [Fig. 12(b)].

Fig. 10. Effect of influent Ca concentration on $K_L/K_{5084}$ (a) and $K_L/K_{6766}$ and (b) ($K_L$ = hydraulic conductivity at end of testing, $K_{5084}$ = hydraulic conductivity at which ASTM D 5084 termination criteria are met, and $K_{6766}$ = hydraulic conductivity at which ASTM D 6766 termination criteria are met). All data are for tests conducted at Univ. of Wisconsin due to ambiguities in Colorado State Univ. data caused by clogging of tubes. Outliers marked with “?”

Fig. 11. Relationship between hydraulic conductivities determined at end of testing ($K_L$) and hydraulic conductivities determined when no trend in hydraulic conductivity exists, permeant salt concentration in influent and effluent are equal, and concentrations of ions in exchange complex (and not in permeant salt) are no longer measurable in effluent.
Fig. 12. Relationship between hydraulic conductivities at end of testing and hydraulic conductivity at (a) electrical conductivity ratio \(= 1.0 \pm 0.05\) (b) and electrical conductivity ratio \(= 1.0 \pm 0.10\).

Ultimately a factor of 3, but still may require a considerable testing time (\(=0.3–1.3\) years).

Because a limited number of solutions were used in this study, these findings apply strictly to the solutions that were used. However, the results presented in Jo et al. (2001) and Kolstad et al. (2004) suggest that the findings from this study are likely to apply generally to solutions of monovalent and divalent cations.

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