

Hydraulic Conductivity of Geosynthetic Clay Liners Exhumed from Landfill Final Covers with Composite Barriers

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Abstract: Geosynthetic clay liners (GCLs) were exhumed from composite barriers, (i.e., geomembrane over GCL) in final covers at four sites after 4.7 to 6.7 years to evaluate the in-service condition. Monovalent bound cations were replaced by divalent cations in all GCLs, with near complete exchange at two-thirds of the sampling locations. Hydraulic conductivity was measured using two dilute solutions commonly used as permeant water: standard water (SW, 0.01M CaCl₂ solution) and type II deionized water (DW). Hydraulic conductivities to SW varied over four orders of magnitude, whereas identical specimens (i.e., from same sample) had hydraulic conductivities to DW consistently $\leq 3 \times 10^{-10}$ m/s. Higher hydraulic conductivities and sensitivity to permeant water did not correspond directly to the amount of cation exchange. Exhumed GCLs with higher gravimetric higher water contents (>50%) exhibited a gel structure indicative of osmotic hydration and had lower hydraulic conductivities to both SW and DW, regardless of the amount of sodium (Na) replaced by divalent cations. These GCLs with higher water contents were placed on subgrade having water content in excess of optimum water content (standard Proctor). Conditions that promote rapid hydration and osmotic swell in a GCL are recommended to ensure that a GCL in a composite barrier maintains low hydraulic conductivity ($\leq 5 \times 10^{-11}$ m/s), even if the native Na is ultimately replaced by divalent cations. Subgrade with water content \geq optimum water content is recommended.

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Introduction

Geosynthetic clay liners (GCLs) are factory-manufactured hydraulic barriers containing sodium (Na) bentonite that are used in waste containment systems to control the migration of liquids and gases. In a final cover, a new GCL typically has a saturated hydraulic conductivity of approximately 10^{-11} m/s (Shan and Daniel 1991; Shackelford et al. 2000; Jo et al. 2001, 2005; Kolstad et al. 2004). Recent studies on GCLs exhumed from final covers have shown, however, that the low hydraulic conductivity of GCLs is not necessarily maintained throughout the service life of a final cover. For example, hydraulic conductivities in the range of 10^{-7} to 10^{-6} m/s have been reported for GCLs exhumed from final covers after 2.0–11.0 years of service (Melchior 2002; Benson et al. 2007; Meer and Benson 2007).

The high hydraulic conductivities observed in exhumed GCLs have been attributed to loss of swelling capacity of the bentonite coupled with formation of cracks and other macroscopic features

during dehydration. During rewetting, swelling of the bentonite is insufficient to seal off these features, which results in high hydraulic conductivity. The loss of swelling capacity is caused by replacement of Na bound to the clay surface by calcium (Ca) and magnesium (Mg), which prevents osmotic swelling in the interlayer of montmorillonite (the primary clay mineral in bentonite). Water entering the GCL from overlying cover soils has been suggested as the primary source of the Ca²⁺ and Mg²⁺ (Melchior 2002; Benson et al. 2007; Meer and Benson 2007; Benson and Meer 2009).

Lin and Benson (2000) hypothesized that GCLs deployed in composite barrier layers, (i.e., GCL overlain by a geomembrane, GM) are unlikely to experience cation exchange and wet-dry cycling, and thus will retain low hydraulic conductivity. However, this hypothesis has remained largely unverified because field data regarding the condition of GCLs in composite barriers are scant and conflicting. Melchior (2002) exhumed a GCL comprised of granular bentonite laminated with a high-density polyethylene (HDPE) geofilm from a final cover test section in Germany 5 years after construction (the GM was oriented upward). The mole fraction of bound Na⁺ decreased from 0.65 to 0.55 while the GCL was in service, whereas near complete replacement of Na⁺ (2 to 4% Na⁺ remaining on the exchange complex) was observed in adjacent GCLs without geofilm or an overlying GM. In contrast, Meer and Benson (2007) exhumed GCL samples from a composite barrier layer (GCL overlain by 1.5 mm textured HDPE GM) in a final cover in Wisconsin that had been in service for 4.1 years. Hydraulic conductivity of the GCL ranged from 5.1×10^{-7} to 1.3×10^{-6} m/s when permeating with 0.01M CaCl₂, more than four orders-of-magnitude higher than the hydraulic conductivity

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of a new GCL. Ca^{2+} and Mg^{2+} replaced at least 55% of the Na^+ originally in the GCL and the swell index (SI) of the bentonite was comparable to Ca-bentonite. Meer and Benson (2007) hypothesized that the cation exchange was due to upward diffusion of divalent cations from the subgrade.

Despite these conflicting reports regarding the in situ GCL condition, composite barriers containing GCLs have performed well as hydraulic barriers in a broad variety of climates. For example, Benson et al. (2007) reported average annual percolation rates between 2.6 and 4.1 mm/year over a 6-year period for a cover containing a GCL laminated with geofilm in a humid continental climate. Similarly, Albright et al. (2004) reported percolation rates ranging from 0 to <0.1 mm/year for two final covers constructed with composite barriers containing a GCL overlain by a HDPE GM that had service lives between 4 and 5 years in arid and semiarid climates (the study described herein includes the GCLs from Albright et al. 2004, and they are shown to be altered). Nevertheless, despite the good performance record, understanding these alterations may be important when making inferences regarding long-term performance.

In this study, GCLs in composite barriers were exhumed from four sites after being in service for 4.7 to 6.7 years. At two of the sites, GCLs were exhumed from test sections simulating covers with composite barriers that were constructed as part of the U.S. EPA Alternative Cover Assessment Program (ACAP) (Albright et al. 2004). GCLs were also exhumed from actual final covers at two municipal solid waste (MSW) landfills that employed composite barrier layers. All GCL samples were tested for saturated hydraulic conductivity, water content, SI, and bound and soluble cations. GCLs were permeated with a 0.01M CaCl_2 solution, which is recommended in ASTM D5084-03 for regions with hard tap water (e.g., Madison, Wisconsin, United States) and is commonly referred to as standard water (SW). Some GCLs were also permeated with DW, including GCLs where high hydraulic conductivity was obtained with SW. Water content and soluble cations in the subgrade were also determined for interpretive purposes.

Background

GCLs containing Na-bentonite typically have hydraulic conductivities ranging from 6×10^{-12} to 2×10^{-11} m/s when permeated with dilute aqueous solutions using conventional test methods and under stresses characteristic of final covers (Petrov and Rowe 1997; Shackelford et al. 2000; Jo et al. 2001; Kolstad et al. 2004; Meer and Benson 2007). Na-bentonite in GCLs has low hydraulic conductivity because much of the water in the bentonite is bound to the clay mineral surface and unavailable for flow (Mesri and Olson 1971; Mitchell 1993; Shang et al. 1994). The association of water molecules with the clay surface during hydration is manifested as swelling, which seals off macroscopic flow paths that can control hydraulic conductivity. Thus, hydraulic conductivity of a GCL and bentonite swell is often related (Shackelford et al. 2000; Jo et al. 2001; Kolstad et al. 2004; Benson and Meer 2009).

Swelling of bentonite occurs in two distinct phases: the crystalline phase and the osmotic phase (Norrish and Quirk 1954). Crystalline swelling occurs first as water molecules move into the interlayer space hydrating the mineral surface and associated cations. Crystalline swelling causes the interlayer to separate by a distance corresponding to several water molecules (McBride 1994). Completion of crystalline swelling corresponds to a gravimetric water content in bentonite of approximately 35% (Mooney

et al. 1952; Norrish and Quirk 1954; Martin 1960; Guyonnet et al. 2005). Osmotic swelling follows crystalline swelling as water molecules flow into the interlayer region in response to the concentration gradient between the interlayer region and the free pore water.

Osmotic swelling can produce far greater swell than crystalline swelling alone (McBride 1994), and is responsible for the high swelling capacity and low hydraulic conductivity of Na-bentonite in DW. The magnitude of osmotic swell is a function of the ionic strength of the pore water, with greater swell occurring when the pore water is more dilute (Norrish and Quirk 1954; McBride 1994; Kolstad et al. 2004; Jo et al. 2005). Bentonites that have undergone osmotic swell generally have water contents exceeding 35%, and in many cases have water contents in excess of 100%. Osmotic swelling only occurs, however, when cations occupying the interlayer space during hydration are predominantly monovalent. When divalent cations are predominant, only crystalline swelling occurs during hydration (Norrish and Quirk 1954; McBride 1994; Guyonnet et al. 2005).

Chemical interactions that affect swelling concurrently affect hydraulic conductivity. Interactions that prevent osmotic swell (e.g., replacement of monovalent cations by divalent cations prior to osmotic swell) result in high hydraulic conductivity, whereas interactions that promote osmotic swell (e.g., permeation by dilute pore water with monovalent cations) result in low hydraulic conductivity (Jo et al. 2001; Kolstad et al. 2004). Bentonites that have already undergone osmotic swelling can retain relatively low hydraulic conductivity under stresses typical of covers (10–30 kPa), even if the Na^+ is subsequently replaced by divalent cations, provided that the bentonite is not desiccated. For example, Egloffstein (2001, 2002) permeated Na-bentonite GCLs with a 0.3M CaCl_2 solution after 20 days of permeation with DW (which promoted osmotic swelling). After 3 years, at which time complete exchange of Na^+ for Ca^{2+} was assumed, the hydraulic conductivity of the GCL was only 3×10^{-10} m/s. Similarly, Jo et al. (2005) showed that permeation of Na-bentonite GCLs with dilute CaCl_2 solutions (<40 mM) that are known to induce osmotic swelling resulted in hydraulic conductivity less than 6×10^{-10} m/s even though more than 94 pore volumes of flow passed through the bentonite and all of the Na^+ was replaced by Ca^{2+} . Lee et al. (2005) show that replacement of Na^+ by Ca^{2+} in GCLs initially prehydrated with DW (promoting osmotic swell) and then permeated with dilute CaCl_2 solutions results in hydraulic conductivities no greater than 3.5×10^{-10} m/s. Similar results have been reported by Gleason et al. (1997) and Shackelford et al. (2000) for Na-bentonites permeated with dilute solutions of divalent cations mimicking soil eluents.

GCLs manufactured with Na-bentonite that have undergone osmotic swelling can retain low hydraulic conductivity even with a preponderance of bound divalent cations because water molecules associated with osmotic swelling are strongly associated with the clay surface (Jo et al. 2005). Osmotic pressures associated with concentration differences in the interlayer and the bulk pore water during exchange have insufficient energy to remove these tightly bound water molecules (Jo et al. 2005; Benson and Meer 2009). However, if these water molecules are extracted by a source with greater energy, much higher hydraulic conductivities may be realized because montmorillonites containing primarily divalent cations do not undergo osmotic swelling when rehydrated (Meer and Benson 2007). This is the reason why a GCL that has undergone replacement of Na^+ by divalent cations coupled with desiccation can be many orders of magnitude more permeable, (e.g., 10^{-7} m/s) than a new GCL (Melchior 2002; Lin

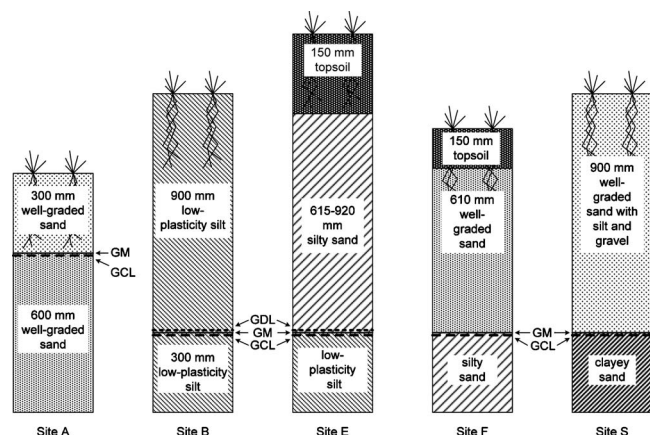


Fig. 1. Profiles of final covers at landfills where GCLs were exhumed. GM=geomembrane, GCL=geosynthetic clay liner, and GDL=geosynthetic drainage layer. Site S originally described in Meer and Benson (2007).

and Benson 2000, Egloffstein 2001; Benson et al. 2007; Benson and Meer 2009). Similarly, rapid exchange relative to the rate of hydration induced by permeation with a concentrated solution of divalent cations (effectively producing Ca-bentonite prior to full osmotic hydration) yields hydraulic conductivities on the order of 10^{-7} m/s (Jo et al. 2005).

Replacement of Na^+ by Ca^{2+} and Mg^{2+} while GCLs are in service is well documented. Downward percolation of pore water containing Ca^{2+} and Mg^{2+} from overlying cover soils is generally cited as the source of divalent cations for exchange (Egloffstein 2001; Melchior 2002; Meer and Benson 2007; Benson et al. 2007). However, Meer and Benson (2007) showed extensive replacement of Na^+ by Ca^{2+} and Mg^{2+} in a GCL exhumed from a composite barrier layer, and attributed the exchange to diffusion of divalent cations into the GCL from the subgrade. Bradshaw (2008) has also shown that divalent cations migrate into GCLs covered by a GM during and after the initial hydration on a subgrade. However, the extent of exchange that commonly occurs in the field in GCLs covered with a GM has not been well documented.

Exhumation of GCLS and Subgrades

GCLs were exhumed from final covers at four sites where they had been used in a composite barrier layer. Two of the sites (A and B) were located in semiarid to arid regions in states on the west coast of the United States; the other two (E and F) were located in the humid continental climate of the midwestern United

States (based on climate definitions in McKnight and Hess 2007). At Sites E and F, samples were exhumed from two adjacent areas that had been constructed at different times. These areas are identified as Site E-01 (Site E, 2001 installation), Site E-02 (Site E, 2002 installation), Site F-03 (Site F, 2003 installation), and Site F-05 (Site F, 2005 installation). All of the GCLs were originally comprised of natural Na-bentonite in granules sandwiched between two geotextiles bonded by needle punching. The cover profile at each landfill is shown in Fig. 1 and the geographic location, service life, and cover thickness are summarized in Table 1. Site S in Fig. 1 and Table 1 is from Meer and Benson (2007). Performance data for Sites A and B are described in Albright et al. (2004).

All GCLs were sampled in accordance with ASTM D6072-08, with a minimum of six square samples (0.3×0.3 m) collected at each site. Soils overlying the composite barrier layer were removed from an area approximately 4×4 m using a tracked excavator until the excavation was within approximately 0.15 m of the uppermost geosynthetic layer. The remaining soil was then removed by hand. Rectangular sections (2×2 m) of geocomposite drainage layer (if present) and GM were removed from the floor of each test pit by cutting the perimeter with a sharp utility knife. No visible defects were observed in any of the overlying GMs. Thus, all of the GCLs were isolated hydraulically from the overlying cover soils.

The perimeter of each GCL sample was cut with a sharp utility knife while the GCL remained on the subgrade [Fig. 2(a)]. GCL surrounding the sampling area was pulled back, and a rigid PVC plate (0.3×0.3 m) was slid under the sample [Fig. 2(b)]. The GCL sample was then wrapped with plastic sheeting to prevent loss of moisture, placed in a plastic tub, and covered with at least 0.1 m of loose soil for protection during transport and storage. A bulk sample of the subgrade immediately beneath the GCL sample (<20 -mm depth) was also collected for determination of water content, particle-size distribution, and chemical analysis of the pore water.

No recommendation is made in ASTM D6072-08 as to how a plated GCL sample should be transported post exhumation, but prior to permeation. One concern is the impact of stress relief prior to testing. To assess this issue, two alternate stress states were tested using split samples from Sites B and E. One state consisted of burying plated GCLs beneath at least 0.5 m of soil in a large plastic tub (henceforth referred to as “with overburden pressure”). The other method consisted of storing the GCLs in a shallow plastic tub beneath a thin (<0.15 -m depth) soil layer (henceforth referred to as “without overburden pressure”). Duplicate samples collected adjacently were used for this evaluation.

Hydraulic conductivity tests were conducted on the duplicate samples using methods described subsequently. A comparison of hydraulic conductivities obtained for both stress states is shown in

Table 1. Description of Covers at Field Sites

	Site						
	West Coast		Midwest			Wisconsin	
Property location	A	B	E	F			S ^a
Installation date	May 2002	November 2000	August 2001	September 2002	August 2003	July 2005	September 1998
Sampling date	March 2007	August 2007	June 2007	June 2007	August 2008	August 2008	October 2002
Service life (year)	4.9	6.7	5.8	4.7	4.9	3.1	4.1
Surface layer thickness (mm)	300	900	915–1,145	915–1,220		760	900

^aMeer and Benson (2007).

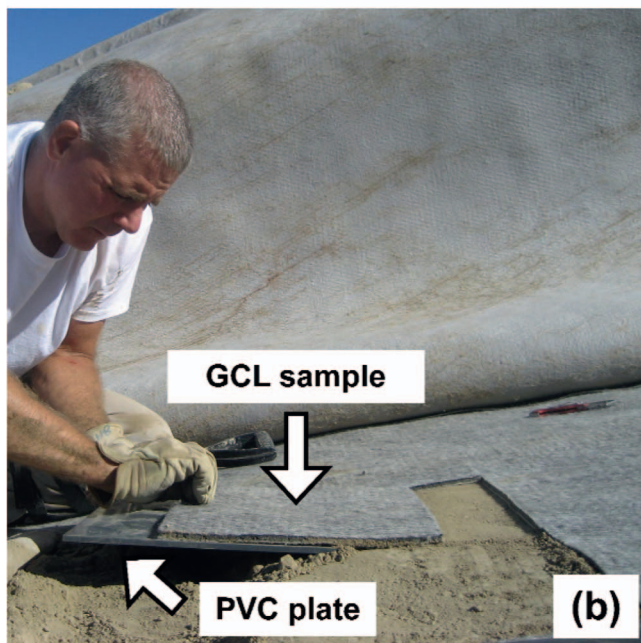
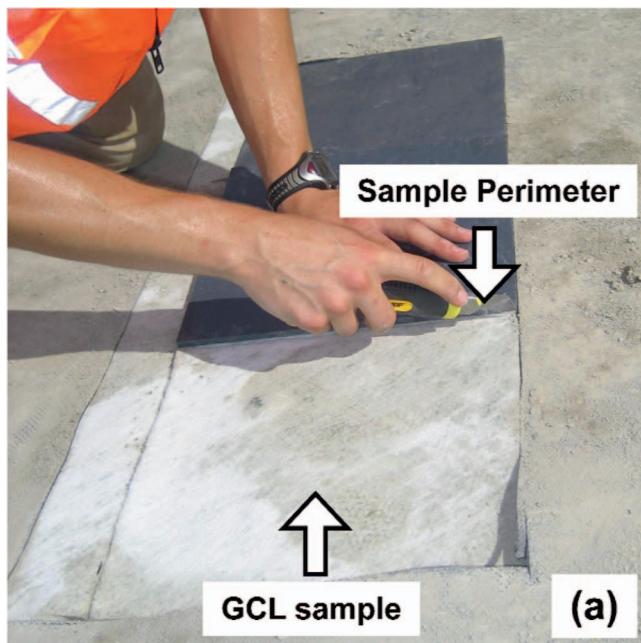


Fig. 2. (Color) Exhumation of GCL samples: (a) cutting around perimeter with razor knife; (b) delicately sliding sample onto rigid plastic plate

Fig. 3. No bias is evident in the hydraulic conductivities, although three of the four specimens from Site B had higher hydraulic conductivity when transported without overburden pressure applied. A *t*-test was performed on the data at a 5% significance level to confirm that the two sets of hydraulic conductivities were statistically similar. The data were transformed logarithmically prior to testing so that the assumption of normality in the *t*-test would be satisfied. A *p*-statistic of 0.17 was calculated, indicating that there was no statistically significant difference between the hydraulic conductivities of both data sets at the 5% level ($0.17 > 0.05$). Overall, hydraulic conductivities from GCLs with overburden pressure were slightly higher (1.1 times, on average) than comparable hydraulic conductivities from GCLs without overbur-

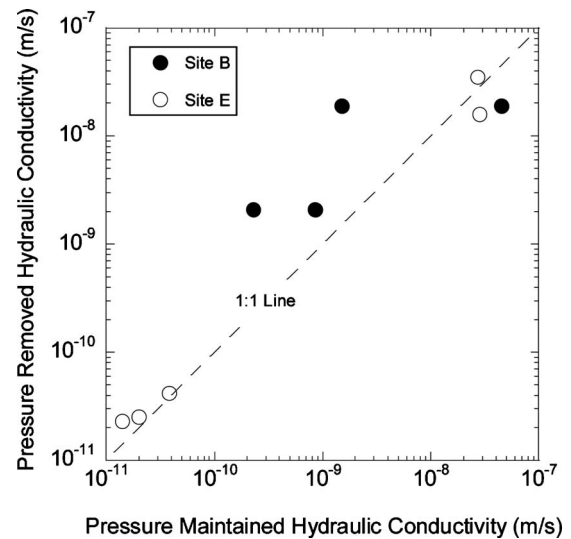


Fig. 3. Hydraulic conductivity of exhumed GCLs with and without overburden pressure prior to permeation

den pressure. However, this difference in hydraulic conductivities is small relative to the overall variation in hydraulic conductivity (three orders of magnitude).

Test Methods

Hydraulic Conductivity

Hydraulic conductivity tests were conducted on GCL specimens in flexible-wall permeameters following the procedures in ASTM D5084-03 and ASTM D6766-06. The hydraulic conductivities are summarized in Table 2. The falling headwater-constant tailwater method was employed. Backpressure was not applied to represent the field condition. The average effective stress was selected to represent the in situ condition, and ranged between 15 and 24 kPa depending on the cover thickness. An average hydraulic gradient of 125 was applied to all specimens. This hydraulic gradient is higher than in the field, but is typical for GCL testing. In addition, Shackelford et al. (2000) showed that hydraulic gradient has negligible impact on the hydraulic conductivity of GCLs when the hydraulic gradient is less than 500.

Specimens having a diameter of 152 mm were cut from the GCL field samples using a razor knife. The GCL sample was retained on the rigid plastic sampling plate during cutting to avoid disturbing any structure within the bentonite. After cutting, geotextile fibers around the perimeter were trimmed back with scissors. Thickness of the GCL specimen was then measured with calipers at six equidistant points around the GCL perimeter and the mass of the specimen was recorded. A frosting of bentonite paste, composed of new Na-bentonite hydrated in the permeant water, was applied to the perimeter of the specimen to prevent sidewall leakage.

Two permeant waters were employed: 0.01M CaCl_2 solution, which is suggested in ASTM D5084-03 for areas with hard tap water, (e.g., Madison, Wis., United States, where the laboratory tests were conducted) and type II DW. All GCL samples were permeated with SW. A portion of the GCLs were tested with DW, particularly those GCLs with high hydraulic conductivity to SW. Solutions similar to SW have been used extensively for permeat-

Table 2. Physical and Chemical Properties of Exhumed GCLs

Site id	Swell index (mL/2 g)	Water content (%)	Hydraulic conductivity (m/s)		Exchange complex (mole fraction) ^a			
			SW ^b	DW ^b	Na ⁺	K ⁺	Ca ²⁺	Mg ²⁺
New ^a	^c 36	—	1.2×10^{-11} ^c	1.1×10^{-11}	^c 0.74	^c 0.02	^c 0.22	^c 0.03
	^c 34	—	1.7×10^{-11} ^c	1.0×10^{-11}	^c 0.65	^c 0.02	^c 0.27	^c 0.03
	20.5	53	1.1×10^{-11}	—	0.32	0.01	0.48	0.18
	18.0	55	1.0×10^{-11}	—	0.33	0.01	0.49	0.17
	22.0	53	9.3×10^{-12}	—	0.39	0.01	0.43	0.17
Site A	19.8	56	1.3×10^{-11}	1.0×10^{-11}	0.35	0.01	0.47	0.16
	13.0	53	1.5×10^{-11}	—	0.25	0.01	0.57	0.17
	20.5	61	1.2×10^{-11}	—	0.30	0.01	0.50	0.19
	20.0	57	1.4×10^{-11}	—	0.29	0.01	0.51	0.19
	16.5	59	1.6×10^{-11}	—	0.34	0.01	0.49	0.16
	12.0	22	1.8×10^{-8}	—	0.37	0.03	0.41	0.20
	14.0	21	2.0×10^{-8}	—	0.45	0.03	0.52	0.00
	20.0	21	4.1×10^{-9}	—	0.52	0.03	0.29	0.16
	16.5	21	1.5×10^{-8}	—	0.59	0.03	0.24	0.14
	16.0	17	2.3×10^{-9}	—	0.46	0.03	0.33	0.18
	14.0	20	8.5×10^{-9}	—	0.46	0.03	0.33	0.18
	17.0	18	2.1×10^{-9}	2.0×10^{-11}	0.43	0.03	0.35	0.19
Site B	13.0	19	4.5×10^{-8}	—	0.41	0.02	0.37	0.20
	15.0	20	1.5×10^{-9}	—	0.54	0.03	0.28	0.15
	18.0	21	1.9×10^{-8}	—	0.52	0.03	0.29	0.16
	8.0	70	4.7×10^{-11}	—	0.06	0.01	0.71	0.21
	8.0	64	4.2×10^{-11}	—	0.06	0.01	0.70	0.22
	10.0	58	4.0×10^{-11}	—	0.05	0.01	0.69	0.25
	10.0	60	2.3×10^{-11}	—	0.05	0.02	0.72	0.22
	8.0	58	1.3×10^{-8} ^d	—	0.05	0.01	0.70	0.25
Site E-01	10.0	56	1.6×10^{-7} ^d	—	0.06	0.02	0.66	0.26
	10.0	56	1.3×10^{-7} ^d	2.5×10^{-10}	0.03	0.01	0.70	0.26
	11.0	63	2.1×10^{-8} ^d	—	0.04	0.00	0.71	0.25
	9.0	60	1.5×10^{-8} ^d	—	0.04	0.01	0.69	0.25
	11.0	68	3.3×10^{-11}	—	0.05	0.01	0.67	0.27
Site E-02	10.0	67	3.2×10^{-11}	—	0.05	0.01	0.69	0.25
	8.0	61	3.7×10^{-11}	—	0.05	0.02	0.72	0.21
	8.0	61	6.5×10^{-9} ^d	8.9×10^{-11}	0.03	0.03	0.95	0.00
	10.0	61	2.6×10^{-9} ^d	9.3×10^{-11}	0.01	0.03	0.96	0.00
Site F-03	10.0	65	3.3×10^{-9} ^d	1.2×10^{-10}	0.01	0.02	0.97	0.00
	13.0	43	3.8×10^{-9}	1.3×10^{-11}	0.14	0.03	0.83	0.00
Site F-05	12.0	46	2.1×10^{-7}	1.4×10^{-11}	0.14	0.04	0.83	0.00
	13.0	45	1.1×10^{-8}	1.3×10^{-11}	0.13	0.03	0.84	0.00

^aOnly major cations satisfying the CEC are presented.^bSW=standard water (0.01M CaCl₂); DW=deionized water.^cTests conducted by Meer and Benson (2007).^dPreferential flow observed.

ing GCLs exhumed from final covers (Egloffstein 2001, 2002; Lin and Benson 2000; Benson et al. 2007; Meer and Benson 2007). Both solutions generally are considered to be nonreactive permeant waters that result in the same hydraulic conductivity for GCLs (Shackelford et al. 2000; Jo et al. 2001; Kolstad et al. 2004). However, DW was used to ensure that the permeant water would not be a source of cations for exchange reactions.

Hydraulic conductivity tests were conducted until the termination criteria stipulated in ASTM D5084-03 were met. For specimens that exhibited high hydraulic conductivity ($>10^{-9}$ m/s), rhodamine WT dye (5 mg/L) was added to the influent liquid at

the conclusion of testing to determine if sidewall leakage was occurring. No indication of sidewall leakage was found in any test. The effluent lines and effluent were also monitored throughout testing for bentonite particle migration. No particles were observed in the effluent lines and effluent.

SI

SI of bentonite from the GCLs was measured using 2 g of oven-dry bentonite removed from each GCL sample. Methods described in ASTM D5890-04 were followed. All tests were

Table 3. Arithmetic Mean Water Content, TCM, and MDR of Exhumed GCLs

Site	Water content (%)		TCM (cmol ⁺ /kg) ^a		MDR ^b	
	Mean	Std. dev.	Mean	Std. dev.	Mean	Std. dev.
A	52	3	5.5	0.9	0.96	0.02
B	20	2	9.2	0.6	0.99	0.01
E-01	62	5	2.6	1.0	0.85	0.10
E-02	59	5	2.8	1.5	0.77	0.19
F-03	63	2	2.5	0.5	0.70	0.02
F-05	45	2	8.4	0.4	0.95	0.00

^aTCM=total charge of soluble cations per mass of soil solid.

^bMDR=ratio of the total charge of monovalent soluble cations relative to the total charge of divalent soluble cations.

conducted with DW as the hydrating solution. Duplicate tests were performed for each sample, but the SI were identical. The SI of each sample is summarized in Table 2.

Soluble Cations, Bound Cations, and Cation Exchange Capacity

Soluble cations (SC), bound cations (BC), and cation exchange capacity (CEC) were determined following the procedures in ASTM D7503-10. Chemical analysis of extracts from the SC and BC tests was conducted using inductively coupled plasma-optimal emission spectroscopy (ICP-OES) following USEPA Method 6010 B (U.S. EPA 2007). BC mole fractions of the major exchangeable cations (Na⁺, K⁺, Ca²⁺, and Mg²⁺) for each GCL sample are presented in Table 2. BC mole fractions were calculated as the ratio of total charge per unit mass of bentonite associated with a particular cation to the CEC.

Strength and relative abundance of SC (cations that can be released by rinsing with water) were quantified by the total soluble cation charge per mass (TCM) and the ratio of monovalent-to-divalent cations (MDR). TCM is defined as the total charge of monovalent and divalent SC per mass of soil solid. MDR is the ratio of the total charge of monovalent SC relative to the total charge of divalent SC. The average TCM and MDR of bentonite from each site is presented in Table 3. These charge and mass-based metrics associated with the bentonite solid are analogous to the ionic strength (I) and ratio of monovalent-to-divalent cations (RMD) used to describe the characteristics of permeant water (Kolstad et al. 2004). MDR is also analogous to the sodium adsorption ratio (SAR), which describes the relative abundance of Na⁺, Ca²⁺, and Mg²⁺ cation in pore water in equilibrium with the

soil solid (McBride 1994). Because TCM and MDR are mass-based metrics, their use precludes the need for a dilution correction to account for differences in liquid-to-solid ratio between the field and in laboratory extracts.

Subgrade Soils

Water contents (ASTM D422-07) and Unified Soil Classification System (USCS) classifications (ASTM D2487-06) of the subgrade soils are summarized in Table 4. Water content of the subgrade varied between sites from 2.3% (Site B) to 15.9% (Site F-03). Sites located in arid climates (Sites A and B) had lower subgrade water contents (2.3–9.8%) than the sites in continental climates (Sites E and F) (8.5–15.9%). The subgrade soils range from well-graded sand (Site A) to low plasticity silt (Site B).

Pore water in the subgrade was characterized using a batch test method similar to the procedure described in ASTM D6141-04. Meer and Benson (2007) indicated that this method “provides a relatively simple and expedient method to generate a test liquid representative of flow-through conditions.” DW was used as the eluent with a liquid-to-solid ratio of 1.3. The soil-water mixture was placed in a sealed 250-mL bottle and rotated for 24 h. The solution was then separated by centrifugation and vacuum filtered through 0.45-μm filter paper. Concentrations of major cations (Ca²⁺, Mg²⁺, Na⁺, and K⁺) in the eluent were measured by ICP-OES following U.S. EPA Method 6010 B (U.S. EPA 2007) and used to compute the TCM and MDR for each subgrade soil (Table 4).

Table 4. USCS Classification (ASTM D2487-06) and Arithmetic Mean Water Content (ASTM D422-07), TCM, and MDR of Subgrade Soils

Site	Soil classification	Water content (%)		TCM ^a (cmol ⁺ /kg)		MDR ^b	
		Mean	Std. dev.	Mean	Std. dev.	Mean	Std. dev.
A	SW	9.8	0.0	0.73	0.09	0.74	0.08
B	ML	2.3	0.2	0.97	0.12	0.76	0.03
E-01	ML-CL	14.2	1.6	0.63	0.08	0.62	0.08
E-02		14.9	1.2	0.58	0.15	0.64	0.07
F-03	SM	15.9	0.0	0.46	0.09	0.52	0.02
F-05		8.5	0.0	1.20	0.15	0.76	0.04

^aTCM=total charge of soluble cations per mass of soil solid.

^bMDR=ratio of the total charge of monovalent soluble cations relative to the total charge of divalent soluble cations.

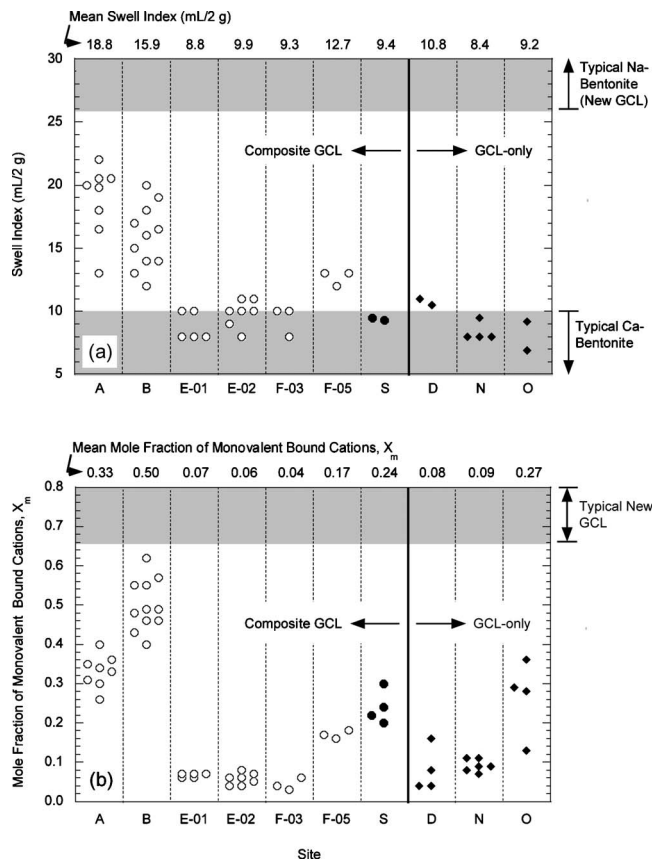


Fig. 4. (a) Swell index in deionized water; (b) mole fraction monovalent bound cations for GCL-only and composite GCL covers. Data are from this study (open symbols) and Meer and Benson (2007) (closed symbols).

Results

GCL Water Content and Cation Exchange

SI and the mole fraction of monovalent cations (X_m) of the exhumed GCLs are shown in Fig. 4 along with the data from Meer and Benson (2007) for a GCL in a composite barrier (Site S) and GCLs that were the sole barrier layer in a final cover (Sites D, N, and O). Swell indices typical of Ca-bentonite (5–10 mL/2 g) and Na-bentonite (26–36 mL/2 g) are denoted in Fig. 4(a), and the X_m typical of a new GCL (>0.65) is marked in Fig. 4(b). Cation exchange and loss of swell are common and extensive in GCLs deployed in composite barriers. At four of the six sampling locations, the GCLs exhumed from composite barriers had SI near those typical of Ca-bentonite (7–11 mL/2 g) and concurrently low X_m (i.e., bound cations predominantly divalent). These SI and X_m are congruent with those for the GCLs exhumed by Meer and Benson (2007). Two of the six sampling locations (locations A and B) in this study had SI and X_m falling between the SI and X_m for Na- and Ca-bentonite, even though all of the GCLs in this study were covered with a GM.

GCLs installed in composite barrier layers are protected from downward percolation of overlying soil eluents. However, cation exchange and loss of swell from multivalent cations is not prevented by the overlying GM, and is limited in only some cases. Multivalent cations may still enter the GCL by advective transport from underlying soil pore water during hydration, diffusion from underlying soil pore water, or a combination of both mecha-

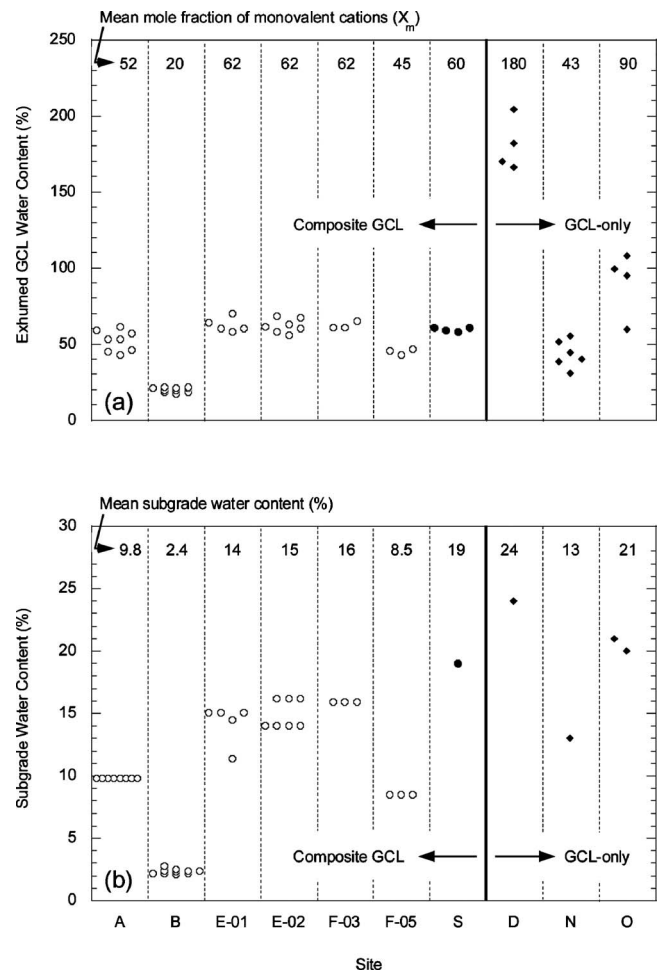


Fig. 5. (a) Gravimetric water content of exhumed GCLs; (b) subgrades for covers where GCLs were in composite barriers or the sole barrier layer. Data are from this study (open symbols) and Meer and Benson (2007) (closed symbols).

nisms (Meer and Benson 2007; Bradshaw 2008). Given the similarity of the SI and X_m of the GCLs exhumed in this study and those in Meer and Benson (2007), transport of cations from the subgrade appears to be as equally important as downward percolation of soil eluent. Eventually, GCLs in most composite barriers probably will undergo complete cation exchange and have swell indices typical of Ca-bentonite once sufficient divalent cations migrate into the GCL.

Water content of exhumed GCLs is shown in Fig. 5(a) along with the data from Meer and Benson (2007). GCLs exhumed from composite barriers exhibit less overall variation in water content (20–63%) than the GCLs not covered by a GM (43–180%), that were reported in Meer and Benson (2007). At a given site, the water content varies by at most 12 percentage points for GCLs in composite barriers, compared to 49 percentage points for GCLs used as the sole barrier layer in Meer and Benson (2007). Moreover, the average water content varies between 45 and 62% for six of the seven sampling locations, where the GCL was part of a composite barrier, but between 43 and 180% for the GCLs used without a GM described in Meer and Benson (2007). More consistent in situ water content in composite barrier GCLs was likely a result of the overlying GM eliminating wet-dry cycling of the GCL.

Water content of the exhumed GCLs varies systematically

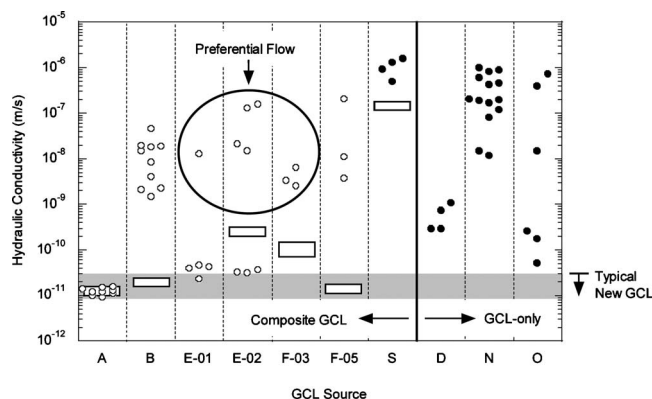


Fig. 6. Hydraulic conductivity of exhumed GCLs from covers where GCLs were in composite barriers or the sole barrier layer. Data are from this study (open symbols) and Meer and Benson (2007) (closed symbols). Circles are hydraulic conductivity to SW; open boxes are hydraulic conductivity to DW.

with the water content of the subgrade, as shown in Fig. 5(b). For example, the driest exhumed GCLs (Site B) were installed on the driest subgrade (water content=2.4%) and the wettest exhumed GCLs (Site D) were installed on the wettest subgrade (water content=24%). Subgrades underlying composite barriers in this study tended to have lower water contents than subgrades underlying GCLs installed as the sole barrier, (i.e., from Meer and Benson 2007). Lower subgrade water content is likely a result of the GM preventing downward percolation.

Hydraulic Conductivity

Hydraulic conductivities of the GCLs exhumed in this study are shown in Fig. 6 along with hydraulic conductivities of GCLs exhumed by Meer and Benson (2007). Hydraulic conductivities of the GCLs exhumed in this study range nearly five orders of magnitude (9.3×10^{-11} to 1.3×10^{-6} m/s). For GCLs permeated with SW, high hydraulic conductivities ($>10^{-9}$ m/s) were obtained at all but one site. Additionally, when permeated with SW, many of the GCLs from composite barriers and those GCLs serving as the sole barrier layer have comparable hydraulic conductivity, except a greater number of GCLs exhumed in this study had hydraulic conductivities to SW close to 10^{-11} m/s. In contrast, the GCLs exhumed in this study had lower hydraulic conductivity to DW than those exhumed by Meer and Benson (2007).

Lower hydraulic conductivities were obtained in most cases when GCLs were permeated with DW rather than SW (Fig. 6). For example, the hydraulic conductivity to SW at Site B ranged between 2×10^{-9} and 5×10^{-8} m/s, whereas the hydraulic conductivity to DW ranged from 2×10^{-11} to 3×10^{-11} m/s. However, at Site A, essentially the same hydraulic conductivity was obtained with SW and DW ($\approx 1 \times 10^{-11}$ m/s). Meer and Benson (2007) (e.g., Site S in Fig. 6) and Benson et al. (2007) did not find similar sensitivity to water type. For the GCLs that they exhumed, GCLs having high hydraulic conductivity to SW also had high hydraulic conductivity to DW.

Because actual pore waters contain a mixture of cations (Meer and Benson 2007; Benson and Meer 2009; Scalia and Benson 2010a), the actual hydraulic conductivity of the GCLs exhumed in this study probably exists between the hydraulic conductivities to SW and DW. However, a definitive inference regarding the actual in-service hydraulic conductivity is not possible. Neverthe-

less, the sensitivity to water type and the high hydraulic conductivity to SW, both of which are atypical of a new GCL, indicate that covering a GCL with a GM does not preclude alteration of the GCL while the GCL is in service. If alteration to the GCLs had not occurred in situ, permeation with SW and DW would have yielded essentially the same hydraulic conductivity, as observed with new GCLs.

After terminating the hydraulic conductivity tests, GCLs with high hydraulic conductivity were permeated with dye to detect if preferential flow was occurring. Preferential flow was observed in the GCLs from Site F-03 and Site E that had high hydraulic conductivities (see call out in Fig. 6). Preferential flow in GCLs from Site E occurred along nearly all of the bundles of needle-punched fibers and was concomitant with dark mineral precipitates (see Scalia and Benson 2010b). For Site F-03, preferential flow in the GCLs also occurred along bundles of needle-punched fibers. However, only 5% of the bundles of needle-punched fibers transmitted preferential flow at Site F-03. GCLs exhibiting preferential flow behaved differently than the other GCLs exhumed in this study and are separated out of the remaining discussion in this paper. They are described in greater detail in Scalia and Benson (2010b).

Hydraulic conductivities of the GCLs are shown in Fig. 7 as a function of SI [Fig. 7(a)] and X_m [Fig. 7(b)]. New GCLs permeated with SW and GCLs from Site S permeated with SW and DW by Meer and Benson (2007) are also shown in Fig. 7. Data from GCLs that did not exhibit preferential flow fall into two bands corresponding to higher hydraulic conductivity and lower hydraulic conductivity. The band with higher hydraulic conductivity shows strong sensitivity to SI and X_m , whereas the band with lower hydraulic conductivity has much less sensitivity to SI and X_m . Higher hydraulic conductivities correspond almost exclusively to GCLs permeated with SW [one data point for DW from Meer and Benson (2007) is in this region], whereas lower hydraulic conductivities correspond to data from GCLs permeated with either SW or DW.

Effect of Subgrade Condition

The relationship between hydraulic conductivity to SW and exhumed water content is shown in Fig. 8(a). When the GCLs with preferential flow are excluded, low hydraulic conductivity is obtained consistently when the water content of the GCL exceeds 50%. For lower water contents, the hydraulic conductivity consistently is higher than 10^{-9} m/s. Meer and Benson (2007) report a similar step relationship, except the transition occurred at a water content of 85%. The data from Site S from Meer and Benson (2007) and the GCLs with preferential flow in this study do not follow this trend.

The relationship between water content of the exhumed GCL and the subgrade water content is shown in Fig. 8(b). Water content of GCLs increases as the water content of the subgrade increases, as has also been shown in laboratory studies (Daniel 1993; U.S. EPA 1996; Thiel and Criley 2005). Subgrade water content also influenced the soluble and bound cations in the bentonite, as shown in Fig. 9. GCLs that had lower TCM [Fig. 9(a)] and lower X_m [Fig. 9(b)] were from subgrades having higher water content. Most importantly, when the GCLs with preferential flow are excluded, low hydraulic conductivity ($<5 \times 10^{-11}$ m/s) to SW was achieved consistently when the subgrade water content was at least 10%.

Water content of the exhumed GCLs is shown as a function of water content of the subgrade relative to optimum water content

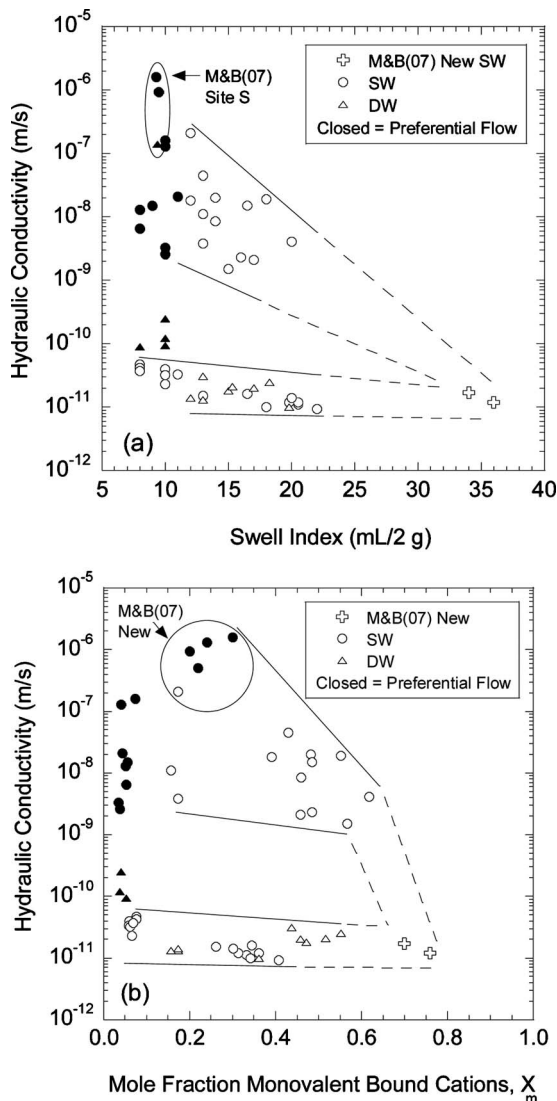


Fig. 7. Hydraulic conductivity to standard water (SW) versus: (a) swell index in deionized water (DW); (b) mole fraction bound sodium for exhumed GCLs from composite barriers. Data for new GCL are from Meer and Benson (2007).

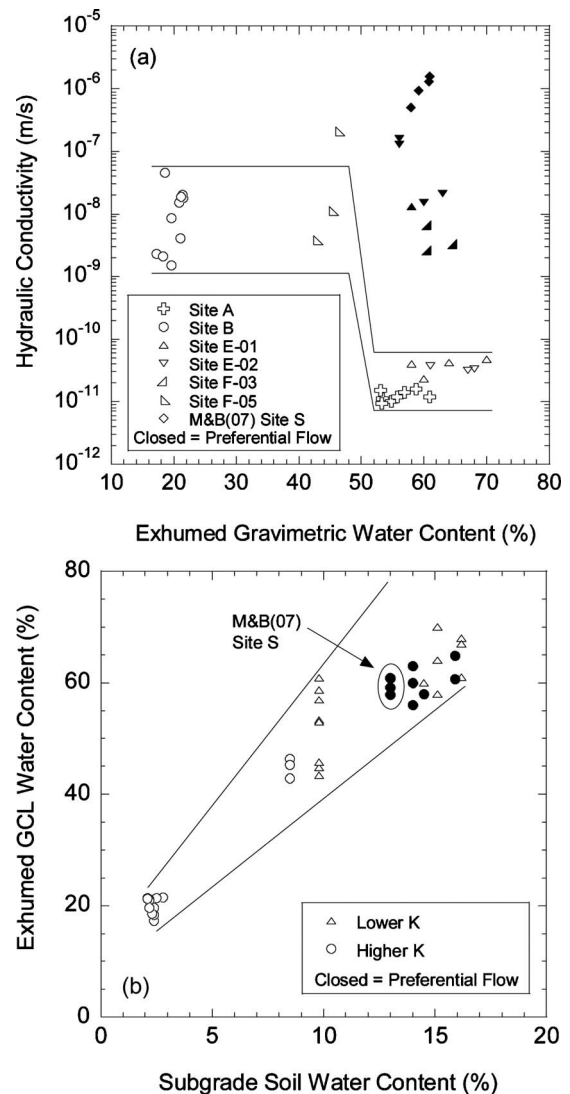


Fig. 8. (a) Hydraulic conductivity (K) of exhumed GCLs to standard water versus water content; (b) water content of exhumed GCLs versus corresponding water content of subgrade. GCLs with lower K had $K < 5 \times 10^{-11}$ m/s, whereas GCLs with higher K had $K > 1 \times 10^{-9}$ m/s.

(OWC, standard Proctor) in Fig. 10. Data from by U.S. EPA (1996) and Bradshaw (2008) are also included in Fig. 10. Both U.S. EPA (1996) and Bradshaw (2008) examined GCL hydration from an underlying compacted subgrade. The duration of these laboratory hydration studies was limited to 75 days (U.S. EPA 1996) and 90 days (Bradshaw 2008). While this duration is much less than the hydration period for the field samples in this study, the duration was sufficient to achieve essentially complete GCL hydration (Daniel 1993; U.S. EPA 1996; Thiel and Criley 2005; Bradshaw 2008). As shown in Fig. 10, water content of the GCLs increases as the water content of the subgrade relative to OWC increases. When the water content of the subgrade soil exceeds OWC, the hydrated GCLs consistently have water contents $> 50\%$, which corresponds to the range associated with osmotic swell and low hydraulic conductivity.

The subgrade also influences soluble cations in the bentonite (Fig. 11). TCM of the GCL increases as TCM of the subgrade increases [Fig. 11(a)], but the relative abundance of monovalent and divalent cations in the GCL (as indicated by MDR) is nearly

independent of the relative abundance in the subgrade [Fig. 11(b)]. The GCLs with preferential flow are an exception; MDRs for these GCLs are comparable to MDRs of the subgrade, which may indicate that these GCLs are closer to equilibrium than those without preferential flow (perhaps due to preferential flow). Most importantly, when GCLs with preferential flow are excluded, low hydraulic conductivity to SW is consistently obtained when the GCL TCM is ≤ 7 cmol⁺/kg and the subgrade TCM is ≤ 0.8 cmol⁺/kg. More dilute pore water in the GCL (lower TCM) promotes osmotic swelling of the bentonite, and therefore lower hydraulic conductivity to SW even if the Na⁺ on the bentonite has been replaced by divalent cations.

Discussion

The aforementioned results have shown that GCLs in composite barriers are altered by their environment even though they are covered by a GM. When GCLs that exhibit preferential flow paths

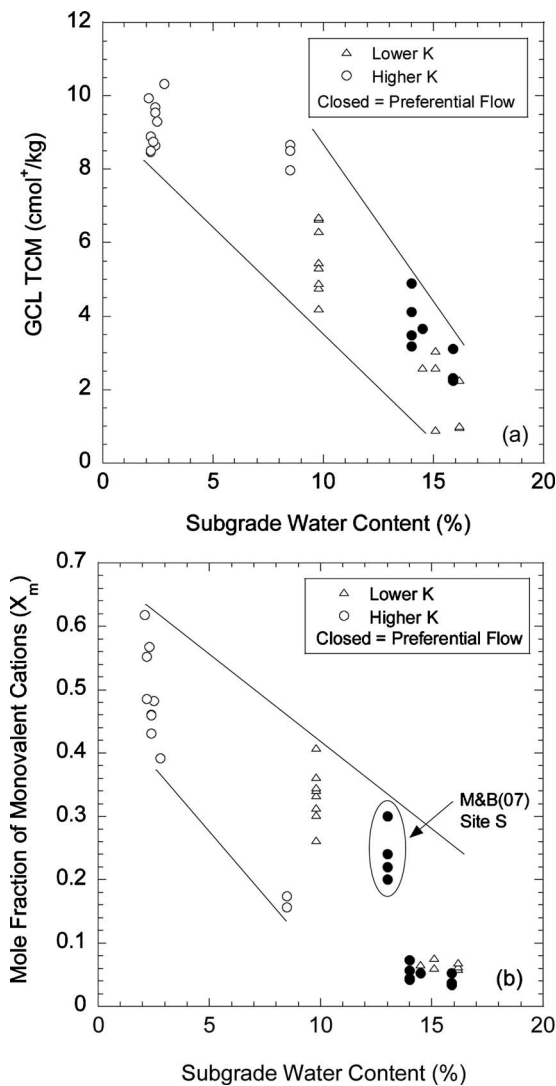


Fig. 9. (a) Total soluble cation charge per mass (TCM); (b) mole fraction of monovalent cations of GCL versus water content of subgrade. GCLs with lower hydraulic conductivity (K) had $K < 5 \times 10^{-11}$ m/s, whereas GCLs with higher K had $K > 1 \times 10^{-9}$ m/s.

are excluded, GCLs in composite barriers that are hydrated sufficiently on a moist subgrade with modest TCM have low hydraulic conductivity ($< 5 \times 10^{-11}$ m/s) and are insensitive to water type. In contrast, GCLs that do not hydrate sufficiently and/or hydrate on a subgrade with lower water content and higher TCM can have high hydraulic conductivity to SW and can be very sensitive to water type (Figs. 8 and 11).

This behavior is in marked contrast to new GCLs, which have essentially identical hydraulic conductivities to SW and DW when permeated for durations similar to the tests conducted in this study (< 30 days) (Jo et al. 2001, 2005; Kolstad et al. 2004). Moreover, the GCLs exhumed in this study have hydraulic conductivities to SW as much as four orders of magnitude higher than the hydraulic conductivity reported for new GCLs after very long-term permeation with solutions similar to SW ($\sim 2 \times 10^{-10}$ m/s) that result in complete replacement of Na⁺ by Ca²⁺ and/or Mg²⁺ (Egloffstein 2001; Jo et al. 2005; Benson et al. 2007). These findings indicate that alterations that occur within composite barriers can introduce unique sensitivity to GCLs, and suggest that the sensitivity is affected by the hydration state.

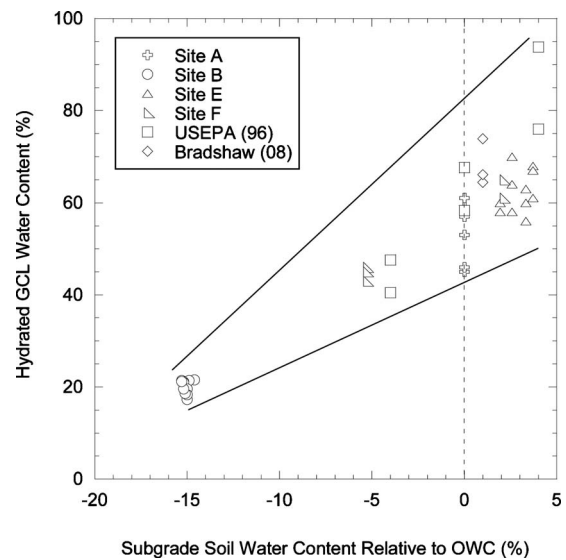


Fig. 10. Water content of GCLs at exhumation as a function of subgrade water content relative to optimum water content (OWC, standard Proctor)

In a composite barrier, hydration of the GCL occurs gradually over a period of approximately 30 days as water migrates upward from the subgrade in the liquid and vapor phases (Daniel 1993; U.S. EPA 1996; Bradshaw 2008). The amount of hydration depends on the water content of the subgrade (Daniel 1993; U.S. EPA 1996; Bradshaw 2008). If the subgrade is sufficiently moist to induce osmotic swelling of the bentonite, (i.e., water content at least 35%) before divalent cations from the subgrade replace the Na⁺, the swollen structure of the bentonite will be retained and permeation with SW or DW will yield low hydraulic conductivity (even if divalent cations in the permeant water replace Na⁺ in the bentonite). For example, at Sites A and E, the GCL had a water content $> 53\%$ and hydraulic conductivities to DW and SW in the range of 9×10^{-12} to 5×10^{-11} m/s, even though divalent cations replaced 48% of the Na⁺ (on average) at Site A and 90% at Site E (GCLs with preferential flow at Site E excluded). Moreover, when exhumed, the GCLs at Sites A and E exhibited the gel-like consistency of bentonite that had undergone osmotic swell [Fig. 12(a)], as described in Guyonnet et al. (2005). Pore water in the subgrade at Sites A and E (Table 4) was also more dilute (TCM < 0.8 cmol⁺/kg), which promotes osmotic swell.

In contrast, if the subgrade has insufficient moisture to promote or complete osmotic swell, and divalent cations replace a substantial portion of the Na⁺ in the bentonite, then the hydraulic conductivity of the GCL to SW can be orders of magnitude higher because osmotic swell is precluded as Ca²⁺ in SW replaces the remaining Na⁺ in the bentonite. For example, the GCLs from Site B and Site F-05 had water content $< 46\%$, substantial replacement of Na⁺ by divalent cations, high hydraulic conductivity to SW, and a granular structure characteristic of a GCL that had not undergone osmotic swell [Fig. 12(b)]. Pore water in these subgrades was also more concentrated (TCM > 0.8 cmol⁺/kg, Table 4). As a result, pore water in the GCL will be more concentrated [Fig. 11(b)], which will suppress osmotic swell.

The GCLs from Site S in Meer and Benson (2007) and the GCLs from Sites E and F with preferential flow are exceptions. These data cluster in Figs. 8(a) and 11(b) and are inconsistent with data for the other GCLs. The GCL from Site S also did not have the gel-like consistency associated with bentonite that had

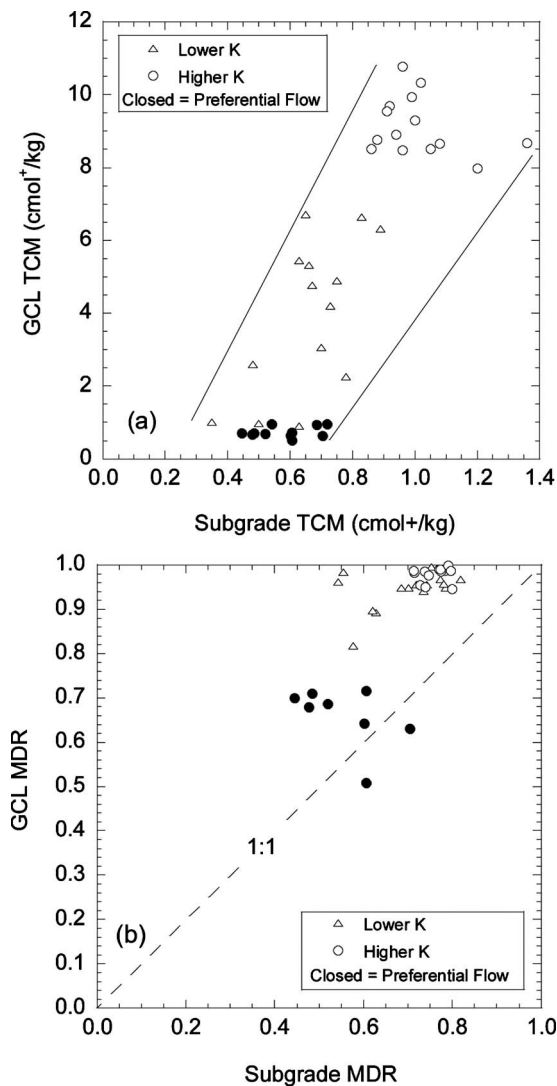


Fig. 11. (a) Total soluble cation charge per mass (TCM) of GCL versus TCM of subgrade; (b) monovalent-to-divalent cation ratio (MDR) of GCL versus MDR of subgrade. GCLs with lower hydraulic conductivity (K) had $K < 5 \times 10^{-11}$ m/s, whereas GCLs with higher K had $K > 1 \times 10^{-9}$ m/s.

undergone osmotic swell and contained cracks typically associated with wet-dry cycling [Fig. 13(a)]. This GCL may have undergone hydration, cation exchange, and then dehydration, even though the GCL was overlain by a GM (e.g., if cover soil was not placed on the GM promptly). However, information regarding the installation and service life of the GCL at Site S is insufficient to confirm whether this sequence of processes could have occurred. Because the GCL from Site S had relatively low X_m and SI (Fig. 4), cracks in the bentonite probably did not swell shut during permeation and acted as preferential flow paths. Consequently, the GCL had similar hydraulic conductivity to SW and DW (Fig. 6).

The GCLs with preferential flow paths from Sites E and F are highly unusual and are different from those at Site S. These GCLs had distinct preferential flow occur along bundles of needle-punched fibers [Fig. 13(b)]. Mechanisms causing these flow paths were beyond the scope of this study, but cation exchange as water from the subgrade is wicked upward through bundles of needle-punched fibers is a likely cause (Scalia and Benson 2010b). Con-

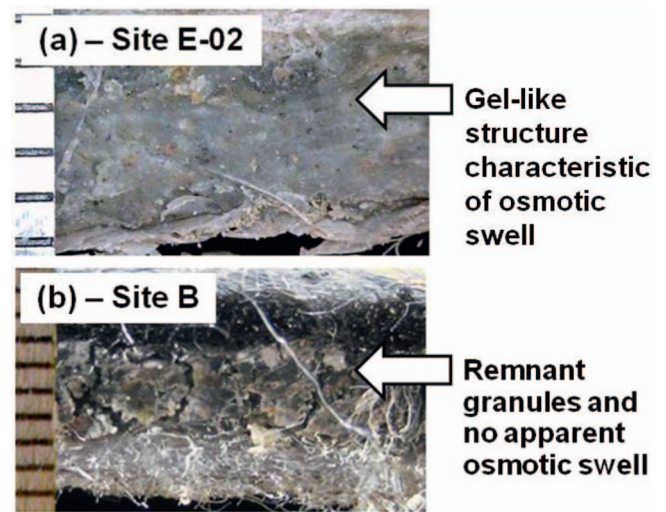


Fig. 12. (Color) (a) Cross sections of exhumed GCLs from Site E-02; (b) Site B. Vertical scale in millimeters.

ditions causing this phenomenon have not yet been identified. However, the bentonite adjacent to the needle-punching fibers did not exhibit the gel-like structure associated with osmotic swell. Remnant granules and inter-granule pores were visible [Fig. 13(b)].

Conclusions and Recommendations

GCLs were exhumed from final covers with composite barriers at four sites after 4.7–6.7 years of service. Hydraulic conductivity of the GCLs was measured using standard water (SW, 10 mM CaCl_2 solution) and type II DW to represent a typical permeant water and nonreactive permeant water. GCLs were also tested for SI, w , BC, SC, and CEC. Subgrade soils were exhumed and evaluated for pore-water composition, w , and particle size distribution.

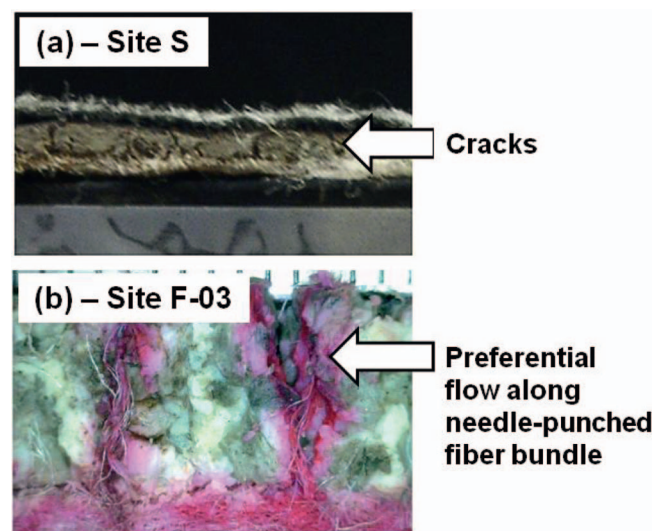


Fig. 13. (Color) (a) Cross sections of exhumed GCLs from Site S; (b) Site F-03. Photo of GCL from Site S is from Meer (2004). Photo of GCL from Site F-03 is after permeation with rhodamine WT dye. Horizontal scale in millimeters.

Based on the findings from these analyzes, the following conclusions and recommendations are made regarding GCLs deployed in final covers with composite barriers:

1. In most environments, divalent cations likely will replace the native Na^+ in GCLs deployed in composite barriers. Exchange appears to occur more rapidly and completely when the GCL is installed on a subgrade with higher water content.
2. The hydraulic conductivity of GCLs exhumed from composite barriers can be sensitive to the type of permeant water. GCLs hydrated to water content in excess of 50% tended to have low hydraulic conductivity regardless of the amount of Na^+ replaced by divalent cations or the type of permeant water. Therefore, conditions that promote rapid hydration to a water content $>50\%$ are recommended to ensure that a GCL has low hydraulic conductivity regardless of the type of pore water migrating through the GCL.
3. The water content of GCLs exhumed from composite barriers was directly related to the water content of the subgrade underlying the GCL. Subgrades with water contents above 10% or in excess of optimum water content were associated with GCLs that had higher water contents ($>50\%$) at exhumation and low hydraulic conductivity regardless of type of permeant water.
4. GCLs with more dilute pore water (lower GCL TCM) were associated with subgrades with more dilute pore water (lower subgrade TCM). GCLs without preferential flow that had low hydraulic conductivity regardless of water type were exhumed from subgrades having $\text{TCM} < 0.8 \text{ cmol}^+/\text{kg}$.
5. Preferential flow was observed in some GCLs along bundles of needle-punched fibers. These GCLs had higher hydraulic conductivity to SW and DW and essentially complete replacement of Na^+ by divalent cations. The mechanisms underlying preferential flow in these GCLs have not yet been identified. However, bentonite surrounding the needle-punched fibers in these GCLs did not exhibit the gel-like appearance of bentonite that had undergone osmotic swell.
6. Subgrade placed in excess of OWC is recommended to ensure rapid hydration and osmotic swell in GCLs used in composite barriers. Provided that desiccation is prevented, GCLs placed under these conditions are likely to maintain low hydraulic conductivity ($\leq 5 \times 10^{-11} \text{ m/s}$) to dilute permeant waters even after complete exchange of divalent for monovalent cations has occurred.

While these findings illustrate that GCLs in composite barriers are altered by their environment, field data indicate that final covers containing composite barriers with a GCL function very well. Percolation rates less than approximately 4 mm/year have been reported in continental climates, and near zero percolation has been reported in semi-arid and arid climates.

Acknowledgments

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