

Hydraulic Conductivity of Geosynthetic Clay Liners to Low-Level Radioactive Waste Leachate – 14082

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ABSTRACT

Experiments were conducted to evaluate how permeation with leachate from low-level radioactive waste (LLW) disposal facilities affects the hydraulic conductivity of geosynthetic clay liners (GCLs) used in composite liner systems. Five commercially available GCLs were evaluated. Two of the GCLs contained conventional sodium (Na) bentonite (C1 and C2) and the others contained polymer-modified bentonite (R1, R2, BPC). The BPC GCL contains a bentonite-polymer composite, whereas the R1 and R2 GCLs contain a dry mixture of polymer and conventional Na bentonite. The GCLs were hydrated and permeated with two synthetic leachates that are chemically identical, except one leachate is prepared without radionuclides (non-radioactive synthetic leachate, or NSL) and the other with radionuclides (radioactive synthetic leachate, or RSL). Control tests were conducted with deionized (DI) water. For the C1, C2, R1, and R2 GCLs, the hydraulic conductivity gradually increased during permeation with synthetic LLW leachate, with final hydraulic conductivities ranging from 1.3×10^{-10} to 5.6×10^{-10} m/s at 20 kPa effective stress. In contrast, the hydraulic conductivity of these GCLs to DI water ranged from 7.6×10^{-12} to 3.1×10^{-11} m/s. The R1 and R2 GCLs, containing a dry mixture of polymer and bentonite, were no more resistant to increases in hydraulic conductivity due to permeation by leachate than conventional Na bentonites, and the largest increases in hydraulic conductivity were obtained with the R1 GCL. In contrast, hydraulic conductivity of the BPC GCL was very low to both LLW leachates ($\approx 6 \times 10^{-12}$ to 8×10^{-12} m/s) and DI water. The BPC GCL had the lowest hydraulic conductivities of all GCLs that were tested for all permeant liquids. Hydraulic conductivities obtained with NSL and RSL were indistinguishable. Permeation with NSL is recommended for design and quality assurance testing.

INTRODUCTION

Geosynthetic clay liners (GCLs) are factory-manufactured hydraulic barriers consisting of a thin layer of sodium (Na) bentonite clay (~ 3 - 5 kg/m²) sandwiched between two geotextiles that are bonded by needle punching or stitching [1]. GCLs are common elements in waste containment facilities due to their low hydraulic conductivity to water (typically $< 10^{-10}$ m/s) and ease of installation [1, 2, 3, 4].

However, the hydraulic conductivity of GCLs can be affected by chemical interactions between the bentonite and liquid to be contained. GCLs permeated with more aggressive leachates having higher ionic strength and/or a predominance of polyvalent cations can be orders of magnitude more permeable than GCLs permeated with deionized (DI) or tap water [2, 3, 5, 6, 7].

GCLs containing polymer-modified bentonites have been proposed for containment of more aggressive liquids [4, 7, 8, 9, 10, 11, 12, 13, 14]. However, not all aggressive leachates are compatible with GCLs containing polymer-modified bentonites. For example, Shackelford et al. [15] conducted hydraulic conductivity tests on GCLs containing a “contaminant-resistant” polymer-modified bentonite using synthetic process water and acidic leachate from a mine waste disposal facility. Control tests were also conducted with site ground water. The GCLs containing “contaminant-resistant” polymer-modified bentonite had hydraulic conductivity to synthetic process water or acidic leachate that was 2300-7600 times higher than the hydraulic conductivity to site groundwater. The conventional GCLs tested with Na bentonite had similar hydraulic conductivity. The high hydraulic conductivities for both types of GCLs were attributed to the high ionic strength and abundance of divalent cations in the process water and acidic leachate.

This example from Shackelford et al. [1] illustrates that leachate-specific testing is needed to confirm that a GCL with conventional Na bentonite or polymer-modified bentonite will have low hydraulic conductivity. This is particularly important for GCLs in low-level radioactive waste (LLW) disposal facilities, which are required to function effectively for at least 1000 years in the United States. This paper describes a study to evaluate the hydraulic conductivity of five commercially available GCLs to synthetic LLW leachate. Two were conventional GCLs containing Na bentonite, two GCLs contained polymer-modified bentonite prepared by dry mixing Na bentonite and proprietary polymers, and one GCL contained the bentonite-polymer composite described in Scalia et al. [7]. Each GCL was permeated with two synthetic leachates representative of LLW leachate encountered in lined on-site disposal facilities operated by the US Department of Energy [16]. The leachates, known as radioactive synthetic leachate (RSL) and non-radioactive synthetic leachate (NSL), are chemically identical except NSL does not contain the radionuclides in RSL.

MATERIALS AND METHODS

Geosynthetic Clay Liner (GCL)

Five commercially available GCLs were evaluated in this study. Two of the GCLs contained conventional bentonite (labeled C1 and C2) and the others contained polymer-modified bentonite (labeled R1, R2, and BPC). R1 and R2 employ a dry mixture of bentonite and proprietary polymer granules. BPC employs a bentonite-polymer composite created in a slurry polymerization process as described in [7].

The polymer is analogous to the superabsorbent polymers used for moisture retention in personal care products.

Each GCL consists of granular material (bentonite granules, mixture of bentonite and polymer granules, or granules of bentonite-polymer composite) sandwiched between a non-woven geotextile (top) and a woven geotextile (bottom). Both geotextiles are polypropylene and are bonded by needlepunching. Physical properties of the GCLs are summarized in Table I. The average water content of the GCLs containing polymer-modified bentonite is more than two times higher than the GCLs containing conventional Na bentonite.

Loss on ignition determined via ASTM D7348 was used as an indicator of polymer content in the bentonite. The conventional Na bentonites both lost 1.6% of their weight, which reflects the loss of strongly bound water molecules [17] and decomposition of calcite. The R1 and R2 bentonites have a modest amount of polymer and the BPC has much more polymer.

TABLE I. Physical properties of GCLs used in study.

GC L	Bentonite Material	Initial Thickness (mm)	Initial Water Content (%)	Initial Pore Volume (mL)	Organic Matter (%)	Swell Index (ml/2g)
C1	Na bentonite	5.1	15.9	24.2	1.6	36
C2	Na bentonite	5.1	7.8	28.2	1.6	32
R1	Bentonite-polymer mixture	5.0	22.7	24.5	3.6	28
R2	Bentonite-polymer mixture	5.9	12.6	27.0	6.1	31
BPC	Bentonite-polymer composite	4.8	44.9	29.8	14.6	45

Note: Swell index measured using ASTM D5890 in DI water.

The major mineral components in each bentonite, determined by X-ray diffraction, are summarized in Table II. Montmorillonite is the predominant mineral in each bentonite, but quartz, plagioclase, feldspar, oligoclase, illite, mica, and calcite are present in measureable quantities. Trace amounts of other minerals (orthoclase, siderite, clinoptilolite, and kaolinite) are also present. The mineralogy of the bentonite in the C1, C2, R1, and R2 GCLs is remarkably similar even though these GCLs were obtained from two different manufacturers. The BPC has slightly lower montmorillonite content, and appreciably more illite and mica than the other bentonites.

TABLE II. Mineralogy of bentonite in GCLs (trc = trace amount).

Mineral Constituents	Relative Abundance (%)				
	C1	C2	R1	R2	BPC
Montmorillonite	84	86	86	85	78
Quartz	9	8	8	10	8
Plagioclase, Feldspar, and Oligoclase	3	3	3	2	5
Illite and Mica	1	-	1	1	7
Calcite	1	-	trc	trc	-
Other Minerals	2	3	2	2	2

Mole fractions of the bound cations and the CEC of each bentonite are in Table III. The bentonites in C1, R1, and BPC have a greater mole fraction of sodium (Na) in the exchange complex compared to C2 and R2, with Na comprising nearly the entire exchange complex of BPC. The greater mole fraction of Na in C1 is reflected in the higher swell index (SI) of this bentonite relative to the other conventional bentonite (C2, see Table I). BPC has a very high mole fraction of Na because a sodic polymer is used to manufacture this composite material [7]. The high SI of BPC reflects swelling of the bentonite and polymer components in the composite.

TABLE III Mole fractions of bound cations and cation exchange capacity of bentonite in each GCL determined using ASTM D7503.

Chemical Property	Major Cations	C1	C2	R1	R2	BPC
Mole Fraction of Bound Cations	Na	0.45	0.42	0.53	0.40	0.75
	K	0.04	0.03	0.03	0.02	0.01
	Ca	0.29	0.31	0.25	0.37	0.09
	Mg	0.12	0.10	0.11	0.09	0.04
CEC (cmol ⁺ /kg)		73.2	71.4	74.6	91.5	142.8

Permeant Liquids

RSL, NSL, and DI water were used as permeant liquids. Constituents in RSL and NSL are summarized in Table IV. These leachate compositions were defined based on an analysis of leachate data from lined LLW disposal facilities operated by the US Department of Energy [16]. The concentrations in Table IV represent average concentrations for each constituent in the LLW database, except for radionuclides, which are at the upper bound concentrations reported by [16] to represent worst-case conditions. NSL has the same chemical composition as RSL, but does not have radionuclides. Trace elements and radionuclides in RSL and NSL account for less than 1% of the ionic strength. Inclusions of radionuclides altered the ionic strength and relative abundance of monovalent and polyvalent cations (shown as RMD in Table IV) by less than 0.03%.

TABLE IV Characteristics of synthetic LLW leachates used in the study.

Major Cations & Anion Concentrations (mM)		Trace Metal Concentrations (mM)			
Ca	4	As	0.001	Al	0.03
Mg	6	Ba	0.002	Mn	0.01
Na	7	Cu	0.0002	Ni	0.0003
K	0.7	Fe	0.04	Sr	0.02
Sulfate	7.5	Li	0.02	Zn	0.0005
Chloride	8	Bulk Characteristics			
Nitrate	1.5	TOC (mg/L)			8
Alkalinity	3.5	Eh (mV)			120
Radionuclides		pH			7.2
U-238 (µg/L)	1500	^a Ionic Strength (mM)			43.6
H-3 (Bq/L)	4440	^b RMD (M ^{1/2})			0.077
Tc-99 (Bq/L)	29.6				

Notes: ^aI and RMD in Table IV for RSL. ^bRMD = $M_M/M_D 0.5$, where M_M = the total molarity of divalent cations in the liquid (M) and M_D = total molarity of polyvalent cations in the liquid (M).

Hydraulic Conductivity Testing

Hydraulic conductivity tests on GCL specimens were conducted in flexible-wall permeameters using the falling headwater and constant tailwater method described in ASTM D6766. The GCLs were hydrated with permeant liquid in the permeameter for 48 hr at an effective confining stress of 10 kPa. After prehydration, the effective confining stress was increased to 20 kPa, and the hydraulic gradient was set at approximately 150. Influent for the specimens was contained in 50 mL burettes sealed with parafilm to prevent evaporation. Effluent was collected in 60 mL polyethylene bottles. The influent and effluent solutions were analyzed periodically for elemental concentrations by inductively coupled plasma-optical emission spectroscopy (ICP-OES). A typical data record from a hydraulic conductivity test is shown in Figure 1.

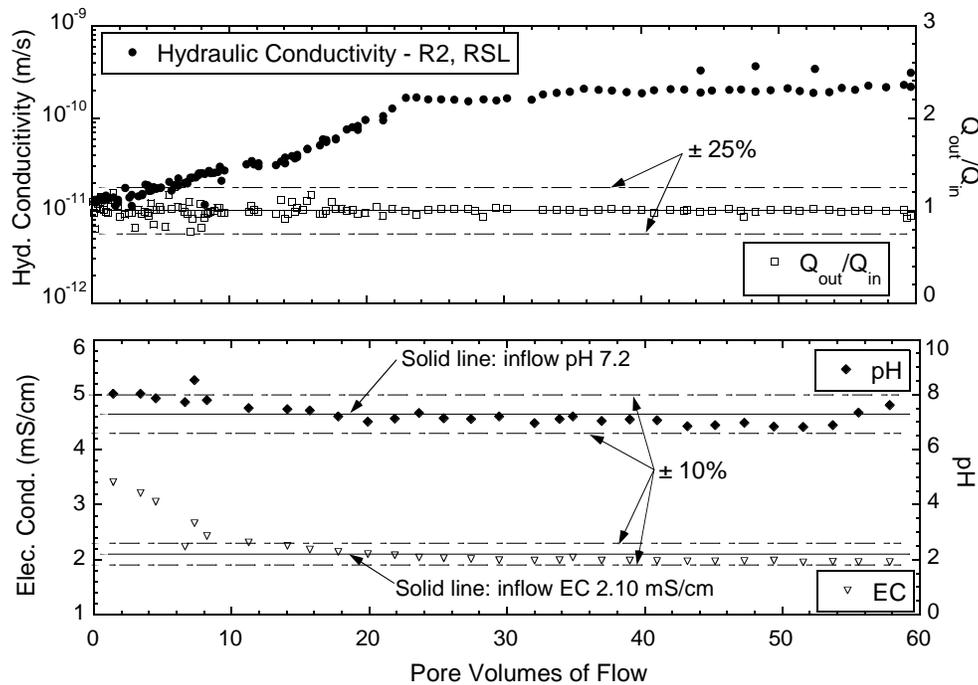


Fig. 1. Data from hydraulic conductivity test on GCL R2 using RSL as the permeant liquid.

Equilibrium was defined using the hydraulic and chemical equilibrium criteria in ASTM D6766 along with an additional criterion for influent and effluent concentrations. The criteria in D6766 require no temporal trend in the hydraulic conductivity measurements, hydraulic conductivity falling within 25% of the mean for three consecutive measurements, incremental effluent volume (Q_{out}) within

25% of the incremental influent volume (Q_{in}) for at least 3 measurements, and the ratio Q_{out}/Q_{in} exhibiting no temporal trend. Chemical equilibrium is defined as the electrical conductivity and pH of the effluent (EC_{out} and pH_{out}) showing no temporal trend and falling within 10% of the electrical conductivity and pH of the influent (EC_{in} and pH_{in}). The additional criterion used in this study required that major cation concentrations in the effluent fall within 10% of major cation concentrations in the influent.

The hydraulic conductivity of the GCL portrayed in Figure 1 increased appreciably during the first 22 PVF due to chemical exchange processes. At approximately 22 PVF, the EC and pH of the effluent leveled off and fell within the range associated with the EC and pH termination criteria. However, the hydraulic conductivity continued to increase slowly after these criteria were met, which reflects slow rate-limited cation exchange processes as described in [6, 18]. The hydraulic equilibrium criterion was achieved at approximately 56 PVF. The criterion for equivalence of cation concentrations in the influent and effluent was also achieved at approximately 56 PVF (not shown).

Effluent concentrations for the C1 and R1 GCLs permeated with RSL and NSL are shown in Figure 2. The solid horizontal lines correspond to the influent concentrations. Na concentrations in the effluent are substantially higher than in the influent during the early portions of the test, as soluble Na is eluted and bound Na is entrained in the effluent due to exchange reactions with Ca and Mg in the influent. The effluent concentration of Na becomes comparable to the influent concentration at around 50 PVF, which is similar to the PVF required to reach hydraulic equilibrium (56 PVF) for GCL R2 permeated with RSL in Figure 1. In contrast, the Ca and Mg concentrations in the effluent gradually increase as the exchange reactions with Na are exhausted. The Ca concentration in the effluent ultimately becomes higher than the influent concentration, most likely due to dissolution of calcite as described in [19].

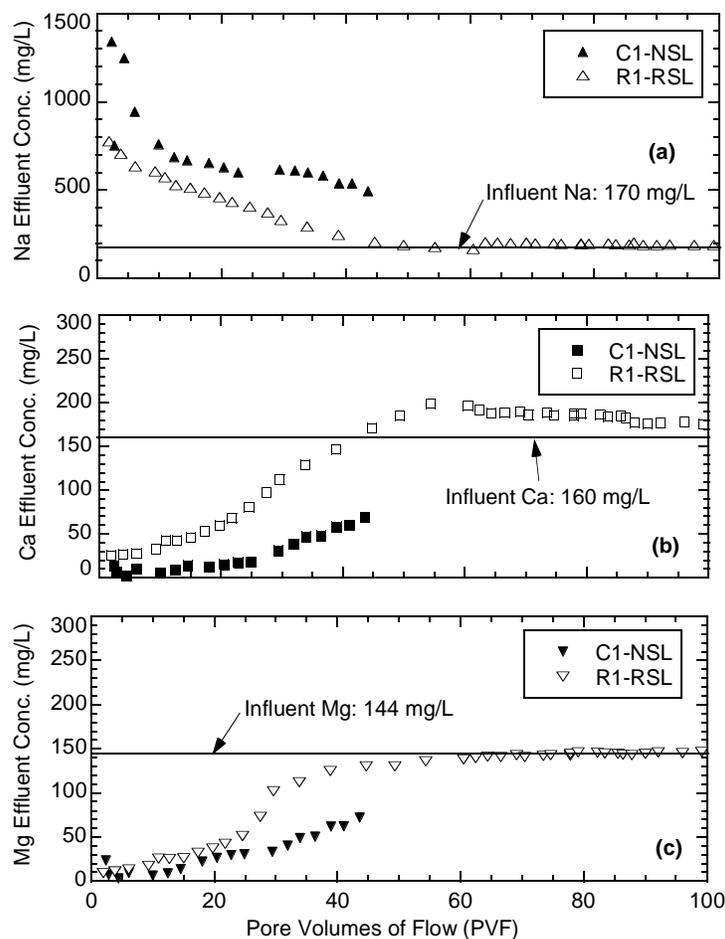


Fig. 2. Concentration of (a) Na, (b) Ca, and (c) Mg in effluent of conventional GCL (C1) and polymer modified GCL (R1) exposed to NSL or RSL.

RESULTS AND DISCUSSION

One specimen of each GCL was permeated with RSL, NSL, and DI water. Summary information from the tests is reported in Table V. The PVF reported in Table V are based on the initial pore volume. At the time this paper was prepared, the tests had been conducted for 0.1-1.3 yr. Of the 10 tests conducted with RSL and NSL, five had met the termination criteria in ASTM D6766 but only two had met the supplemental concentration criterion.

The tests conducted in this study are still ongoing, and the hydraulic conductivities that are reported represent a snapshot in time. Tests reported as in equilibrium met the aforementioned hydraulic conductivity, flow, EC, and pH criteria that are stipulated in D6766, but may not have met the supplemental concentration criterion. Jo et al. [6] indicate that hydraulic conductivities at concentration equilibrium are no more than 3 times higher than hydraulic conductivities obtained using the equilibrium criteria in D6766.

TABLE V. Hydraulic conductivity of GCLs permeated with RSL, NSL, and DI water.

GCL	Test Time (yr)	Permeant Liquid	PVF	D6766 Termination Criteria		Hydraulic Conductivity (m/s)
				Hydraulic Equilibrium	EC and pH Equilibrium	
C1	1.0	RSL	62.6	Yes	No	1.6×10^{-10}
	1.3	NSL	90.5	Yes	Yes	2.3×10^{-10}
	0.1	DI	3.8	Yes	No	2.5×10^{-11}
C2	0.5	RSL	18.0	Yes	No	6.7×10^{-11}
	0.5	NSL	20.7	Yes	No	5.9×10^{-11}
	0.1	DI	3.5	Yes	No	3.1×10^{-11}
R1	1.0	RSL	109.5	Yes	Yes	1.8×10^{-10}
	1.3	NSL	88.3	Yes	Yes	1.3×10^{-10}
	0.1	DI	4.1	Yes	No	2.1×10^{-11}
R2	1.0	RSL	92.1	Yes	Yes	4.3×10^{-10}
	1.3	NSL	62.6	Yes	Yes	5.6×10^{-10}
	0.1	DI	1.6	Yes	No	7.6×10^{-12}
BPC	0.7	RSL	3.9	No	No	6.0×10^{-12}
	0.6	NSL	3.1	No	No	7.6×10^{-12}
	0.2	DI	1.9	Yes	No	6.7×10^{-12}

Hydraulic conductivity records from tests conducted on the C1, R1, and BPC GCLs with RSL are shown in Figure 3. These records are typical of trends for GCLs permeated with RSL or NSL. The hydraulic conductivity of GCLs C1 and R1 decreased slightly for the first 5-10 PVF, increased monotonically from approximately 10 to 60 PVF, and then leveled off (R1) or continued to increase slowly (C1). Jo et al. [6] report similar long-term trends for Na bentonite GCLs exposed to dilute solutions with divalent cations (e.g. $\text{CaCl}_2 \leq 20 \text{ mM}$) similar to RSL and NSL. The hydraulic conductivity in the experiments by Jo et al. [6] ultimately increased by a factor of 3-15 while exchange reactions occurred (Na^+ in the montmorillonite interlayer slowly replaced by Ca^{2+} [6]).

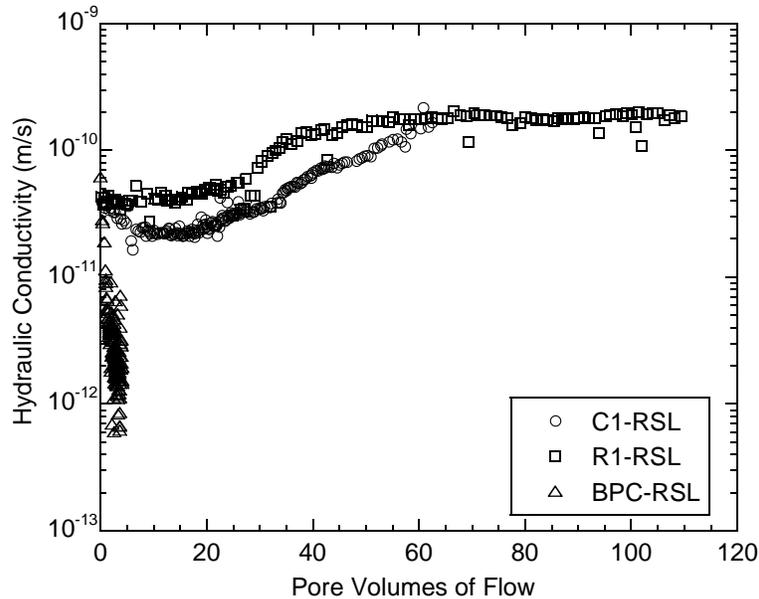


Fig. 3. Hydraulic conductivity vs. pore volumes of flow for GCL C1, R1, and BPC exposed to radioactive synthetic leachate (initial pore volume flow were used because no permeameters were terminated)

In contrast to the C1 and R1 GCLs, hydraulic conductivity of the BPC GCL decreased substantially during the test period. The BPC GCL had fewer PVF during testing than the C1 and R1 GCLs, in part due to testing over a shorter duration (approximately 50% shorter test time) but largely due to the low flow rate in the BPC GCL due to very low hydraulic conductivity. The low hydraulic conductivity was due to clogging of pores in the BPC by eluted polymer in a manner analogous to the clogging reported in [7]. Clogging is evident in the scatter in the hydraulic conductivity and incremental flow ratio (Q_{out}/Q_{in}) records shown in Figure 4; similar scatter is reported in [7]. Because so few PVF had passed through the BPC GCL, chemical equilibrium was not established. Longer-term testing of the BPC GCL is underway to ensure that the hydraulic conductivity that is reported represents hydraulic and chemical equilibrium.

Hydraulic conductivities of the GCLs to RSL are compared to the hydraulic conductivities to NSL in Figure 5. Essentially the same hydraulic conductivities were obtained using either leachate, as all of the data fall within the band corresponding to a factor of 2 (reproducibility of hydraulic conductivities of GCLs [6, 20]). The similarity in the hydraulic conductivities to RSL and NSL reflects the very small impact of the radionuclides on total ionic strength and RMD of the leachate (both < 0.03%). The SI data are consistent with this finding (Figure 6). Similar SI were obtained for NSL and RSL for each bentonite. All of the bentonites had lower SI to RSL and NSL than to DI water, which is also consistent with the higher hydraulic conductivities to RSL and NSL compared to the hydraulic conductivity to DI water for all of the GCLs except the BPC GCL (Figure 6). Scalia

et al. [7] also show that the hydraulic conductivity of the BPC GCL is not related systematically to SI.

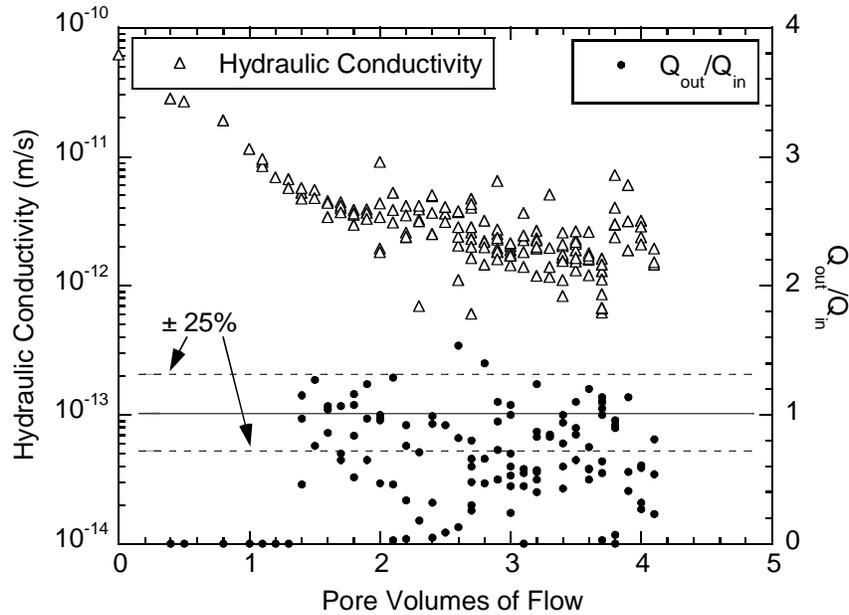


Fig. 4. Hydraulic conductivity (open symbols) and incremental flow ratio (Q_{out}/Q_{in} , closed symbols) of BPC GCL as a function of pore volumes of flow when permeated with RSL leachate.

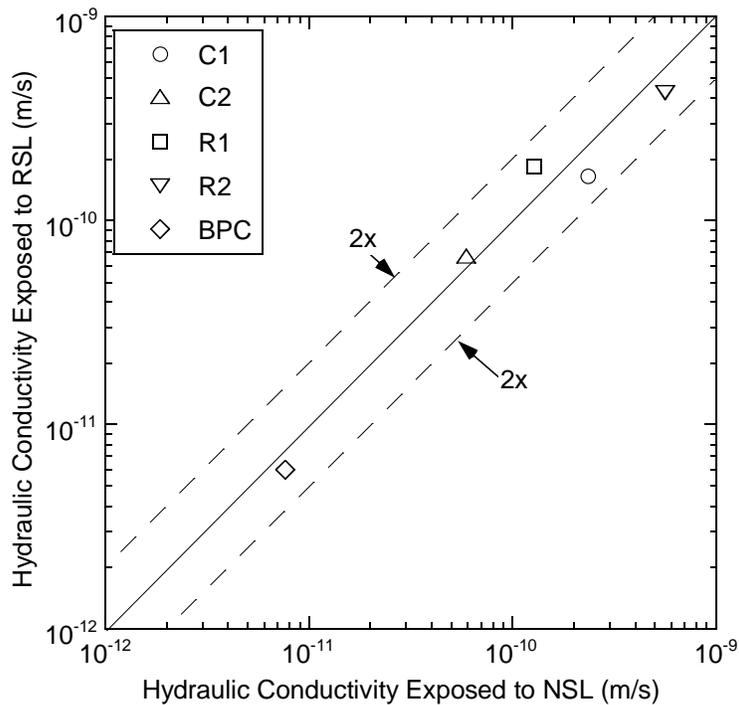


Fig. 5. Hydraulic conductivity of GCLS exposed to radioactive synthetic leachate (RSL) compared to hydraulic conductivity to non-radioactive synthetic leachate (NSL). Data represent test conditions at time paper was prepared.

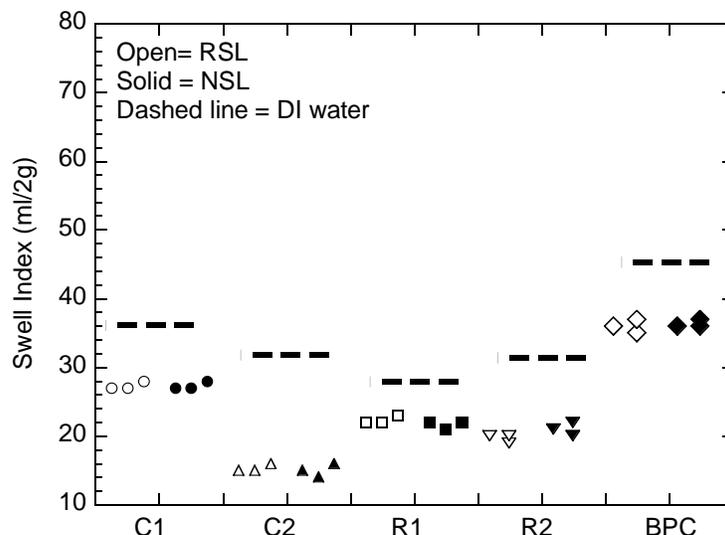


Fig. 6. Swell index (SI) of bentonite from each GCL in RSL, NSL, and DI water.

Polymer-modification with a dry blend of bentonite and polymer (R1 and R2 GCLs) did not consistently reduce the impact of leachate on the hydraulic conductivity of the GCLs, as shown in Figure 7. The C2 GCL with conventional bentonite had lower hydraulic conductivity to leachate than the C1 GCL with conventional bentonite and both GCLs with polymer-modified bentonites. The C1 (conventional) and R1 (polymer modified) GCLs had similar and modestly higher hydraulic conductivity to leachate (6.2 to 9.2 times higher than the hydraulic conductivity to DI water), and the R2 (polymer-modified) GCLs were the most permeable (56-74 times higher than the hydraulic conductivity to DI water). In contrast, the BPC GCL had the lowest hydraulic conductivity of all GCLs evaluated.

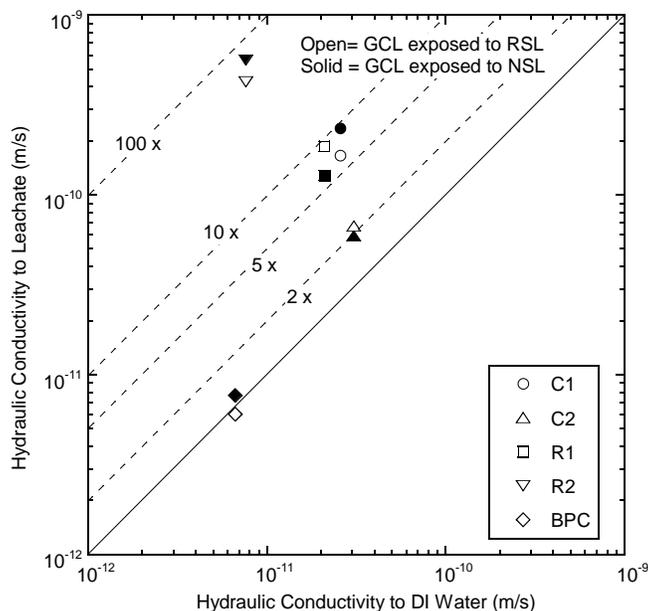


Fig. 7. Hydraulic conductivity of GCLs to leachate compared to hydraulic conductivity to DI water

SUMMARY AND CONCLUSIONS

This study evaluated how the hydraulic conductivity of GCLs is affected by permeation with liquids characteristic of leachate from LLW disposal facilities. Five commercially available GCLs were evaluated. Two of the GCLs contained conventional Na bentonite and the others contained polymer-modified bentonite. One of the GCLs with polymer-modified bentonite was a bentonite-polymer composite, whereas the others were a dry mixture of bentonite and polymer granules. The GCLs were hydrated and permeated with two synthetic leachates that are chemically identical, except one leachate is prepared without radionuclides (NSL) and the other with radionuclides (RSL). Control tests were conducted with DI water.

The following conclusions are drawn based on the findings of this study:

- The hydraulic conductivity of GCLs containing conventional Na bentonite and a dry mixture of polymer and bentonite gradually increases during permeation with synthetic LLW leachate. The final hydraulic conductivities range from 1.9 to 74 times the hydraulic conductivity to water.
- GCLs containing bentonite-polymer composite have very low hydraulic conductivity to leachate and DI water. These GCLs have lower hydraulic conductivity to DI water and both synthetic LLW leachates than all other GCLs evaluated.

- Polymer-modified bentonites consisting of a dry mixture of polymer and bentonite may be no more resistant to increases in hydraulic conductivity due to permeation by leachate than conventional Na bentonites. Leachate specific testing is needed to confirm that a polymer-modified bentonite will be more resistant to chemical interactions with LLW leachate than conventional Na bentonite.
- Hydraulic conductivities of GCLs to non-radioactive and radioactive synthetic LLW leachate are indistinguishable. Testing with non-radioactive leachate is recommended to simplify health and safety concerns when testing GCLs for LLW facilities.

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