



Hydraulic Performance of Geosynthetic Clay Liners Comprising Anionic Polymer–Enhanced Bentonites

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Abstract: The hydraulic performance of geosynthetic clay liners (GCLs) comprising anionic polymer–enhanced bentonites (EBs), or EB–GCLs, based on permeation with 500 mM NaCl and 167 mM CaCl₂ at an average effective stress of 27 kPa and hydraulic gradients of ~300 was evaluated. The EBs included powdered sodium bentonite (NaB) enhanced with one of seven anionic polymers, including linear, noncrosslinked poly(acrylic acid) (PA) with low, medium, and high molecular weights, sodium carboxymethylcellulose (CMC) with either low viscosity or high viscosity, a covalently crosslinked sodium polyacrylate, and an in situ polymerized bentonite polymer composite. The NaB was enhanced with polymer at mass loadings of 5%, 8%, or 10% using dry-sprinkling, dry-mixing, and/or wet-mixing methods. Hydraulic conductivity tests at a low hydraulic gradient (~35) also were conducted to determine the effect of seepage forces. Overall, the hydraulic performance of the EB–GCLs to the NaCl solution was better than that to the CaCl₂ solution. Dry sprinkling (DS) with PA generally provided the best hydraulic performance, with hydraulic conductivity values based on permeation with 500 mM NaCl and 167 mM CaCl₂ of 1.3×10^{-11} and 4.0×10^{-11} m/s, respectively, for the high-molecular-weight PA added at 5% via DS and 2.9×10^{-11} and 4.7×10^{-11} m/s, respectively, for the medium-molecular-weight PA added at 8% via DS. The CMC viscosity grade did not affect the hydraulic conductivity of the EB–GCLs. All EB–GCLs eluted polymer during permeation, regardless of mixing method or polymer type. Polymer elution was correlated with preferential interaggregate flow paths in multiple EB–GCL specimens, indicating the importance of polymer retention in maintaining low hydraulic conductivity of EB–GCLs. Because standard termination criteria for the measurement of the hydraulic conductivity of conventional unenhanced GCLs (e.g., as per current ASTM standards) do not address the potential impact of polymer elution from EB–GCLs, caution should be exercised when adapting these criteria to measure the hydraulic conductivity of EB–GCLs such as those evaluated in this study. DOI: [10.1061/\(ASCE\)GT.1943-5606.0002781](https://doi.org/10.1061/(ASCE)GT.1943-5606.0002781). © 2022 American Society of Civil Engineers.

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Introduction

Geosynthetic clay liners (GCLs) comprising natural sodium bentonite (NaB) are used extensively as engineered barriers or components of engineered barrier systems in waste containment applications (e.g., liners for landfills). However, conventional GCLs have proven to be ineffective hydraulic barriers to aggressive inorganic solutions, such as hypersaline solutions (e.g., brines) or extremely acidic or basic leachates, especially under low confining stresses (Shackelford et al. 2000; Benson et al. 2010; Bouazza and Gates 2014; Chen et al. 2019). The hydraulic incompatibility between natural NaB and liquids with aggressive chemistry has motivated the production of a plethora of chemically enhanced bentonites (EBs) for use in GCLs for the purpose of improving hydraulic

performance. The types of chemical enhancements that have been evaluated include organic compounds such as propylene and glycerol carbonate (e.g., Onikata et al. 1999; Katsumi et al. 2008; Mazzieri et al. 2010; Fehervari et al. 2016a, b; Gates et al. 2016), and anionic polymers such as sodium carboxymethylcellulose (CMC) (e.g., Di Emidio 2010; Di Emidio et al. 2015; De Camillis et al. 2014, 2016; Fan et al. 2020) and poly(acrylic acid) (PA) (e.g., Heller and Keren 2002; Scalia et al. 2011; Bohnhoff 2012; Scalia 2012; Bohnhoff and Shackelford 2013; Bohnhoff et al. 2014; Tian et al. 2016a, b, 2017; Chen et al. 2019; Prongmanee et al. 2018a, b; Scalia et al. 2018; Prongmanee and Chai 2019; Tian et al. 2019; Chai and Prongmanee 2020; Li et al. 2021; Tong et al. 2021).

Scalia et al. (2018) compared measured hydraulic conductivity results from multiple sources for conventional GCLs, i.e., GCLs containing only natural (unenhanced) sodium bentonite, as well as enhanced-bentonite GCLs (EB–GCLs) permeated with solutions with a range of ionic strengths, I [$I = (1/2)\sum c_i z_i^2$, where c_i is the molar concentration of ionic species i , and z_i is charge of ionic species i]. They showed that the hydraulic conductivity of conventional GCLs was $\leq 5 \times 10^{-11}$ m/s at $I \leq 20$ mM, but was higher and more variable for solutions with higher I . The hydraulic performance of the EB–GCLs was better than that for conventional GCLs in some cases, but significant variability in hydraulic conductivity ($1.0 \times 10^{-13} \leq k \leq 9.0 \times 10^{-6}$ m/s) was evident for $I > 20$ mM, especially for commercially produced contaminant resistant clays with proprietary amendments. These results illustrate that not all EB–GCLs are equally effective, and that the hydraulic conductivity of EB–GCLs can vary considerably based

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on the properties of the polymer (e.g., molecular weight and degree of polymerization), method of preparing the EB, and the properties of the hydrating and permeant liquids.

Also, the mechanisms underlying the hydraulic compatibility of EBs and EB-GCLs to aggressive inorganic solutions differ from those of natural NaB, thereby rendering traditional indicator parameters, such as swell index, ineffective for EBs (Onikata et al. 1996, 1999; Trauger and Darlington 2000; Katsumi et al. 2001, 2008; Schroeder et al. 2001; Ashmawy et al. 2002; Kolstad et al. 2004; Guyonnet et al. 2009; Di Emidio et al. 2010, 2011; Mazziari et al. 2010; Naismith et al. 2011; Scalia et al. 2011; Bohnhoff et al. 2013; Scalia et al. 2014; Tian et al. 2016a, b; Scalia et al. 2018; Chai and Prongmanee 2020; Tian et al. 2019; Reddy et al. 2020).

The objective of this study was to evaluate the effects of the material properties and method of preparation on the hydraulic conductivity of EB-GCLs comprising anionic-polymer [poly(acrylic acid) or sodium carboxymethylcellulose] EBs based on permeation with aggressive inorganic chemical solutions (500 mM NaCl and 167 mM CaCl₂). The hydraulic conductivity values of the anionic-polymer EB-GCLs were also compared with those of an EB-GCL comprising bentonite polymer composite (BPC), an in situ polymerized EB (e.g., Bohnhoff 2012; Bohnhoff and Shackelford 2013; Bohnhoff et al. 2014; Scalia et al. 2014). Finally, hydraulic conductivity tests at a low hydraulic gradient (i.e., ~35 versus ~300) were also conducted to determine the effect of seepage forces and polymer retention or elution. The results of this study demonstrated that changes in hydraulic behavior of anionic-polymer EBs and EB-GCLs can be related to differences in (1) polymer type and properties (e.g., molecular weight), (2) polymer mass loading, (3) method of specimen preparation, (4) chemical composition of permeant liquid, and (5) hydraulic gradient applied during testing.

Materials

Materials used in this study include NaB, CMC with low viscosity (CMCLV) and high viscosity (CMCHV), noncrosslinked linear PA with low, medium, and high molecular weights (PALW, PAMW, and PAHW, respectively), a crosslinked PA (PAX), and the BPC. The polymer mass loadings representing the mass percentage of the polymer added to the NaB to comprise an EB were 5%, 8%, or 10%, which is indicated by the number appended to the aforementioned designations. These polymer contents are consistent with the range in polymer contents of ~2% to 10% in previous studies evaluating the hydraulic conductivity of EBs and EB-GCLs permeated with aggressive chemical solutions (e.g., Tian and Benson 2019; Di Emidio 2010; Chen et al. 2018; Li et al. 2021; Zainab et al. 2021). For example, PALW5 indicates that 5% of the PALW by mass was added to 95% of the NaB by mass. Finally, the EBs are designated as DS, DM, and WM to indicate the method of preparation as dry sprinkling, dry mixing, and wet mixing, respectively. In addition to these solid materials, different chemical solutions were used as hydrating and permeating liquids. Further details on these materials follow.

Sodium Bentonite

The NaB used in this study was obtained from Colloid Environmental Technologies Company (CETCO) (Hoffman Estates, Illinois) and is the same NaB used in Bentomat GCLs as well as other studies on GCL behavior (Scalia et al. 2011; Bohnhoff 2012; Bohnhoff and Shackelford 2013; Bohnhoff et al. 2014; Scalia et al. 2014). The NaB was evaluated in a powdered form. The particle-size distribution and material properties of the NaB are provided in

Fig. S1 and Table S1. Both dry and wet particle-size distributions were consistent with that of a fine-grained soil.

Sodium Carboxymethylcellulose

The CMCLV used in this study was produced by Calbiochem (Millipore Sigma, Burlington, Massachusetts), and the CMCHV was produced by MP Biomedical (Irvine, California). Carboxymethylcellulose can be differentiated by degree of substitution (DoS), i.e., the number of carboxymethyl groups substituted per anhydroglucose unit, and a weighted average molecular weight, which describes the average chain degree of polymerization (DP) (Buchholz and Graham 1998). The technical specifications for the CMCHV are given in Table 1, whereas those for the CMCLV were not provided by the manufacturer. The particle sizes for the two CMCs were not altered from the manufacturer provided material, and the resulting particle size distributions of both CMCs are provided in Fig. S1(b).

Poly(acrylic Acid)

The PA ($[-CH_2CH(CO_2H)-]_n$) used in this study is a synthetic polymer of acrylic acid. The negatively charged carboxylic moiety (COO⁻) on the polymer repeating unit (*R*) is satisfied by a proton (H⁺). Commercial PA is differentiated primarily by molecular weight and DP (Buchholz and Graham 1998). Poly(acrylic acid) that is covalently crosslinked is known as crosslinked PA.

Technical specifications of the PAs provided by the manufacturer are given in Table 1. The PALW, PAMW, and PAHW were supplied by Polysciences (Warrington, Pennsylvania), and the manufacturer reported molecular weights were approximately 5,000, 50,000, and 345,000 g/mol, respectively. All the PAs were linear, i.e., noncrosslinked.

When used as an enhancement, the PA in solution was air-dried until solidified and then ground and screened. Grinding of the polymer was completed using a rotary blade grinder (KitchenAid BCG211OB, Benton Harbor, Michigan). The PA was ground to achieve a particle-size distribution similar to that for the base NaB to allow for homogeneous mixing (Malusis and Scalia 2007) (Fig. S1).

A nonlinear crosslinked PAX also was used in this study for comparison with the linear noncrosslinked PAs. The PAX was a partial sodium salt-graft-poly(ethylene oxide) from Aldrich Chemistry (St. Louis, Missouri). Properties of the PAX (e.g., molecular weight) were not provided by the manufacturer.

Table 1. Parameters of polymers used in this study

Polymer type	DoS	DP	Molecular weight (g/mol)
CMCHV	0.65–0.85 ^a	3,200 ^a	685,290–736,500 ^b
PALW	N/A	29.9	5,000 ^a
PAMW	N/A	— ^c	50,000 ^a
PAHW	N/A	— ^c	345,000 ^a

Note: DoS = degree of substitution; DP = average chain degree of polymerization; CMCHV = high-viscosity sodium carboxymethylcellulose; PALW, PAMW, and PAHW = low-, medium-, and high-molecular-weight polyacrylate; PAX = covalently crosslinked polyacrylate; and N/A = not applicable.

^aReported by the manufacturer.

^bMolecular weight of CMC calculated as $[C_6H_7O_2(OH)_x(OCH_2COONa)_y]_n$ where *n* is DP, *x* + *y* = 3, and *y* is DoS.

^cNot provided by manufacturer.

Bentonite Polymer Composite

The BPC used in this study was the same BPC as used in previous studies (Scalia et al. 2011, 2014; Bohnhoff et al. 2013). The BPC, which comprised approximately 28.5% of polyacrylate polymer (Scalia et al. 2014), was tested as a dry-mixed enhancement of NaB at 5% polymer by mass (i.e., EB with 17.3% BPC). Properties of the granular NaB used to prepare the BPC have been given by Scalia et al. (2014). For this study, the BPC was ground to a powdered form by mortar and pestle with 51.6% of the particles <0.075 mm. Further details on the BPC material properties are given in Table S1 and Fig. S1(a).

Hydrating and Permeant Liquids

Concentrated inorganic solutions, i.e., 167 mM CaCl₂ and 500 mM NaCl, with $I = 500$ mM, were used as hydrating and permeant liquids to evaluate the hydraulic conductivity of the EB-GCL specimens. The 500-mM NaCl solution was chosen as a permeant liquid based on the results from Petrov and Rowe (1997) for conventional GCLs permeated with NaCl solutions as reported by Shackelford et al. (2000). These results showed a trend of increasing hydraulic conductivity with increasing NaCl concentration of the permeant liquid, with an increase in hydraulic conductivity of approximately two orders of magnitude due to permeation with a 500-mM NaCl solution relative to that based on permeation with water. The 167-mM CaCl₂ solution was chosen to maintain the same ionic strength of 500 mM. Also, the results from Lee and Shackelford (2005) indicated that permeation of a conventional GCL with 167-mM CaCl₂ solution can result in an increase in hydraulic conductivity relative to that based on permeation with water ranging from approximately two to almost five orders of magnitude depending on the quality of the NaB in the GCL.

The electrical conductivity (EC) and pH of the 167-mM CaCl₂ solution were 3,280 mS/m and 5.8, respectively, whereas the respective EC and pH of the 500 mM NaCl were 4,850 mS/m and 6.2. The CaCl₂ solution was prepared by dissolving CaCl₂ dihydrate, CaCl₂ • 2H₂O (Alfa Aesar, Ward Hill, Massachusetts) in de-ionized water (DIW). The NaCl solution was prepared by dissolving anhydrous NaCl (Fisher Chemical, Hampton, New Hampshire) in DIW. The prepared solutions were stored in collapsible carboys with no headspace to limit interaction with the atmosphere.

Methods

Bentonite Modification

The NaB was enhanced with an anionic polymer using three mixing methods, namely dry mixing, dry sprinkling, and wet mixing. Dry-mixed EBs were prepared by adding a percentage (by mass) of air-dried polymer to air-dried NaB. The masses were not corrected for the gravimetric air-dried (hygroscopic) water content for the NaB of 8.0%. The mixture was rotated end-over-end in a sealed container at 30 revolutions per minute (RPM) for 1 min. Dry-sprinkled EBs were prepared by sprinkling a known percentage (by mass) of air-dried polymer evenly as a layer at the inflow side (bottom) of the GCL specimen, i.e., on top of the inflow side geotextile (Fig. S2), before placing the air-dried NaB on top.

Wet-mixed EBs were created by first adding a known, target percentage of 5% of air-dried polymer by mass of air-dried NaB into 350 mL of DIW. For wet-mixed EBs prepared with both CMCHV and CMCLV, the air-dried CMC was added to 350 mL of DIW in a mechanical stirrer (Hamilton Beach Single Spindle Drink

Mixer, Glen Allen, Virginia) set to the highest operating speed (18,000 rpm). The CMCs and DIW were mixed for 5 min to allow for the dispersion and hydration of the dried polymer. After 5 min, 50 g of air-dried NaB was added in the mechanical stirrer cup with additional DIW and mixed at 18,000 rpm for 10 min, resulting in a total of 500 mL of DIW and a total mixing time of 15 min. The total volumes of DIW varied for the EB mixtures and reflected the maximum amount of DIW that could be added to the mixture to fill the cup without losing slurry while mixing. Further details regarding the mixing method for each wet-mixed EB can be found in the Supplemental Materials. The wet-mixed EBs then were oven-dried at 105°C for 24 h and ground using the rotary blade grinder until 100% passed the US No. 40 sieve (0.420 mm). All of the PSDs for the wet-mixed EBs were similar (Fig. S1).

Geosynthetic Clay Liner Preparation

Enhanced bentonites prepared using the dry- and wet-mixing methods were tested for hydraulic conductivity in the form of unreinforced GCL specimens. A more detailed description of the preparation of these specimens as well as schematic representations of the specimens are provided in the Supplemental Materials and Fig. S2.

Hydraulic Conductivity

Hydraulic conductivity tests were performed using flexible-wall permeameters following ASTM D6766-18 (ASTM 2018) and the falling headwater, constant tailwater method except backpressure was not applied (e.g., Kolstad et al. 2004; Lee and Shackelford 2005; Meer and Benson 2007; Bradshaw et al. 2013; Scalia et al. 2014; Tian et al. 2016a). Prior to permeation, each specimen was hydrated with the permeant liquid for 48 h under an average effective confining stress of 27 kPa. A total confining stress of 34.3 kPa was applied during hydration and throughout the hydraulic conductivity test via gravity head. A hydraulic gradient of 200–300 is typical for hydraulic conductivity testing of GCLs due to the typically low k (Shackelford et al. 2000; Scalia et al. 2014). Therefore, for most tests, a target, average hydraulic gradient of 200 was applied via gravity head using glass burettes (falling head). The target hydraulic gradient was based on an assumed typical hydrated GCL thickness of 7.5 mm. However, the final average hydraulic gradients for the permeated specimens ranged from 242 to 327 due to thinner-than-expected final thicknesses (i.e., 4.30 to 6.84 mm) resulting from a significant reduction in swelling of the NaB in the aggressive inorganic solutions as well as material loss via polymer elution.

Finally, to evaluate the effect of the applied hydraulic gradient on the measured hydraulic conductivity of the EB-GCLs, hydraulic conductivity tests were reproduced at a lower hydraulic gradient of ~35 for EB-GCL specimens enhanced by dry sprinkling with 5% PAHW and 5% PALW and permeated with 167 mM CaCl₂. More details about the hydraulic conductivity testing procedures can be found in the Supplemental Materials. Each specimen was permeated from bottom (inflow) to top (outflow) at least until the ASTM D6766 termination criteria were achieved. However, because the potential impact of polymer elution was not considered by the ASTM D6766 termination criteria, permeation was continued beyond the duration required by ASTM D6766 for all but one specimen to evaluate the applicability of the ASTM D6766 termination criteria for EB-GCLs. The potential for preferential flow also was evaluated by adding 5 mg/L Rhodamine WT dye to the influent as described by Scalia and Benson (2011).

Table 2. Properties and hydraulic conductivity test results of a conventional GCL comprising NaB and EB-GCL specimens permeated with 500 mM NaCl

Specimen designation	Specimen properties							Elapsed time, t (days)		PVF		Hydraulic conductivity, k ($\times 10^{-10}$ m/s)			
	Polymer type	Molecular weight or viscosity	P_i (%)	Mixing method	d_f (mm)	L_f (mm)	w_f	t_{6766}	t_f	PVF ₆₇₆₆	PVF _{f}	k_{6766}	k_f	k_{6766}/k_f	k_{EB-GCL}/k_{GCL}
NaB	N/A	N/A	N/A	N/A	152.4	6.10	0.86	1.7	4.8	2.6	5.9	2.8	1.6	1.8	N/A
CMCLV5DM	CMC	LV	5	DM	150.0	5.51	0.91	196	214	14.8	25.8	0.85	1.2	0.71	0.30
CMCLV5WM	CMC	LV	5	WM	151.6	6.10	1.03	120	126	3.1	4.9	0.69	0.67	1.0	0.25
CMCLV5DS	CMC	LV	5	DS	151.8	4.80	0.79	62.5	66.7	9.3	14.6	1.2	1.4	0.86	0.43
CMCHV5DM	CMC	HV	5	DM	145.9	5.75	0.96	199	216	18.7	31.3	1.2	1.4	0.86	0.43
CMCHV5WM	CMC	HV	5	WM	150.1	6.84	1.18	30.5	77.3	3.0	6.9	0.33	0.23	1.4	0.12
PALW5DS	PA	LW	5	DS	149.9	5.30	0.94	2.7	18.0	6.6	19.1	2.4	1.4	1.7	0.86
PALW8DS	PA	LW	8	DS	150.2	4.70	0.85	3.9	4.7	3.7	4.8	3.2	3.0	0.94	0.58
PALW5WM	PA	LW	5	WM	154.1	5.50	0.97	5.9	21.0	2.4	7.3	0.78	0.49	1.6	0.28
PAMW5DS	PA	MW	5	DS	145.7	5.20	0.79	41.4	73.6	5.8	12.5	0.26	0.27	0.96	0.093
PAMW8DS	PA	MW	8	DS	150.0	4.80	0.80	20.9	59.2	6.7	18.1	0.29	0.33	0.88	0.10
PAMW5WM	PA	MW	5	WM	153.7	6.90	1.06	22.4	85.0	4.5	11.5	0.48	0.35	1.4	0.17
PAHW5DS	PA	HW	5	DS	150.2	4.60	0.76	34.6	66.4	3.3	8.1	0.13	0.18	0.72	0.046
PAHW5WM	PA	HW	5	WM	152.1	6.34	1.08	16.4	188	5.3	20.4	0.52	0.15	3.5	0.19
PAX5DS	PAX	Unknown	5	DS	149.7	7.00	0.95	88.9	136	5.4	7.9	0.25	0.25 ^a	1.0	0.089
BPC5DM	PA	Unknown	5	DM	149.1	6.20	0.82	61.5	143	2.9	11.5	0.11	0.19	0.58	0.039

Note: N/A = not applicable; NaB = sodium bentonite; PA = polyacrylate; CMC = carboxymethylcellulose; PAX = polyacrylate; BPC = bentonite polymer composite; HW = high molecular weight; MW = medium molecular weight; LW = low molecular weight; HV = high viscosity; LV = low viscosity; DS = dry sprinkling; WM = wet mixing; DM = dry mixing; P_i = initial polymer content based on mass of polymer added; d_f = final diameter; L_f = final thickness; w_f = final gravimetric water content; PVF = pore volumes of flow; t_{6766} , PVF₆₇₆₆, and k_{6766} = values based on ASTM D6766 termination criteria; t_f , PVF _{f} , and k_f = final values at the end of testing; $k_{GCL} = k_{6766}$ of NaB GCL specimen; and $k_{EB-GCL} = k_{6766}$ of polymer-amended EB-GCL specimen.

^aHydrogel visible on geotextiles and/or permeameter.

Table 3. Properties and hydraulic conductivity test results of a conventional GCL comprising NaB and EB-GCL specimens permeated with 167 mM CaCl₂

GCL designation	Specimen properties							Elapsed time, <i>t</i> (days)		PVF		Hydraulic conductivity, <i>k</i> (×10 ⁻¹⁰ m/s)			
	Polymer type	Molecular weight or viscosity	<i>P_i</i> (%)	Mixing method	<i>d_f</i> (mm)	<i>L_f</i> (mm)	<i>w_f</i>	<i>t</i> ₆₇₆₆	<i>t_f</i>	PVF ₆₇₆₆	PVF _{<i>f</i>}	<i>k</i> ₆₇₆₆	<i>k_f</i>	<i>k</i> ₆₇₆₆ / <i>k_f</i>	<i>k</i> _{EB-GCL} / <i>k</i> _{GCL}
NaB	N/A	N/A	N/A	N/A	150.3	6.80	0.80	1.6	5.6	3.0	9.2	5.5	4.8	1.1	N/A
CMCLV5DM	CMC	LV	5	DM	150.3	5.10	0.91	3.0	4.4	3.2	16.2	1.4	31 ^a	0.047	0.26
CMCLV5WM	CMC	LV	5	WM	151.9	4.84	0.88	3.2	4.3	6.7	10.4	4.0	4.1	0.98	0.72
CMCHV5DM	CMC	HV	5	DM	149.0	5.60	0.97	65.4	65.5	14.2	21.4	135	181 ^a	0.78	24.5
CMCHV5WM	CMC	HV	5	WM	152.0	6.60	1.06	8.4	8.8	5.7	6.2	3.7	3.8	0.97	0.67
PALW5DS	PA	LW	5	DS	150.7	4.30	0.76	1.4	3.7	8.2	16.1	5.7	3.4 ^b	1.7	1.0
PALW5DS ^c	PA	LW	5	DS	151.6	4.33	0.62	351	494	2.0	3.3	0.12	0.08 ^b	1.5	0.02
PALW10DS	PA	LW	10	DS	150.0	6.20	0.75	3.5	4.3	3.9	5.5	4.1	4.0	1.0	0.75
PALW5WM	PA	LW	5	WM	154.0	5.30	0.85	2.5	3.3	5.0	7.0	3.3	3.8	0.87	0.60
PAMW5DS	PA	MW	5	DS	150.4	4.67	0.75	1.7	2.4	12.4	15.3	4.7	4.1	1.1	0.85
PAMW8DS	PA	MW	8	DS	150.8	4.91	0.75	84.9	117	7.1	13.0	0.11	0.11 ^b	1.0	0.020
PAMW10DS	PA	MW	10	DS	148.2	6.27	0.74	48.7	112	3.8	10.1	0.25	0.27	0.93	0.045
PAMW8DM	PA	MW	8	DM	148.2	5.20	0.75	4.3	5.1	3.8	4.7	2.5	2.5	1.0	0.45
PAMW8WM	PA	MW	8	WM	152.7	5.33	0.82	5.1	6.5	4.4	5.8	0.85	0.82	1.0	0.15
PAHW5DS	PA	HW	5	DS	153.3	4.81	0.79	50.2	50.2	13.3	13.3	0.40	0.40 ^b	1.0	0.073
PAHW5DS ^c	PA	HW	5	DS	145.8	4.37	0.71	515	519	2.6	2.6	0.11	0.11 ^b	1.0	0.020
PAHW8DS	PA	HW	8	DS	148.6	6.18	0.79	24.6	71.3	3.8	10.7	0.46	0.34	1.4	0.084
PAHW5WM	PA	HW	5	WM	151.5	5.25	0.86	1.5	3.3	2.6	5.7	3.1	2.5	1.2	0.56
PAX5DS	PAX	Unknown	5	DS	147.8	4.63	0.85	4.2	5.5	2.9	4.4	1.9	2.1 ^b	0.90	0.35
BPC5DM	BPC	Unknown	5	DM	148.0	5.90	0.69	44.0	77.0	4.1	11.5	0.32	0.55	0.58	0.058

Note: N/A = not applicable; NaB = sodium bentonite; PA = polyacrylate; CMC = carboxymethylcellulose; PAX = polyacrylate; BPC = bentonite polymer composite; HW = high molecular weight; MW = medium molecular weight; LW = low molecular weight; HV = high viscosity; LV = low viscosity; DS = dry sprinkling; WM = wet mixing; DM = dry mixing; *P_i* = initial polymer content based on mass of polymer added; *d_f* = final diameter; *L_f* = final thickness; *w_f* = final gravimetric water content; *t*₆₇₆₆, PVF₆₇₆₆, and *k*₆₇₆₆ = values based on ASTM D6766 termination criteria; *t_f*, PVF_{*f*}, and *k_f* = final values at the end of testing; *k*_{GCL} = *k*₆₇₆₆ of NaB GCL specimen; and *k*_{EB-GCL} = *k*₆₇₆₆ of polymer-amended EB-GCL specimen.

^aPreferential flow.

^bHydrogel visible on geotextiles and/or permeameter.

^cTested at target average gradient of 35.

Results

Hydraulic Conductivity

Summaries of the measured specimen properties and results of the hydraulic conductivity tests are provided in Tables 2 and 3 for specimens permeated with 500-mM NaCl and 167-mM CaCl₂ solutions, respectively. The reported values include the final thickness (L_f), final water content (w_f), and the values for the testing duration (t), pore volumes of flow (PVF), and hydraulic conductivity (k) based on both the ASTM D6766 termination criteria (k_{6766}) and the end-of-the-test conditions (k_f). The ASTM D6766 termination criteria generally captured the hydraulic conductivity at equilibrium for the EB-GCL specimens (Fig. S3). Further discussion of the ASTM D6766 termination criteria and the final hydraulic conductivities can be found in the Supplemental Materials. Unless otherwise noted, all hydraulic conductivity values referred to subsequently are the k_{6766} values.

The results for the conventional GCL specimens comprising the unenhanced NaB are presented in Fig. S4. The hydraulic conductivity to 500 mM NaCl and 167 mM CaCl₂ were 2.8×10^{-10} and 5.5×10^{-10} m/s, respectively. The order-of-magnitude similarity in these values is likely attributable, in part, to the equivalent ionic strength ($I = 500$ mM) of both permeant liquids, and the higher hydraulic conductivity to 167 mM CaCl₂ can be attributed to the greater detrimental impact of the Ca²⁺ versus Na⁺ cation (e.g., Shackelford 1994).

The measured hydraulic conductivity values were lower than those reported in other studies for similar NaB permeated with similarly aggressive solutions. For example, for a GCL comprising granular bentonite with Atterberg limits [liquid limit (LL) = 430 and plasticity index (PI) = 393] that are similar to those for the NaB used in this study (Table S1), Lee and Shackelford (2005) reported a mean hydraulic conductivity for duplicate specimens permeated with 100 mM CaCl₂ of 3.4×10^{-9} m/s, which was 6.1 times higher than the hydraulic conductivity of 5.5×10^{-10} m/s measured in this study to 167 mM CaCl₂. This difference in measured hydraulic conductivity can be attributed, in part, to the initial aggregate size of the NaB, i.e., the NaB in this study was powdered, whereas that in the study by Lee and Shackelford (2005) was granular (e.g., Howell et al. 1997). As noted by Shackelford et al. (2000), the initial aggregate size of NaB can impact the swelling of the NaB and ultimately the measured hydraulic conductivity, with a larger initial aggregate size generally correlating with less swell and higher hydraulic conductivity.

EB-GCLs Enhanced with CMC

The results of the hydraulic conductivity tests for the EB-GCLs enhanced with CMC are shown in Fig. 1 and summarized in Tables 2 and 3. Only a single test was conducted for a specimen prepared using the dry-sprinkling method, i.e., the CMCLV5DS specimen permeated with 500 mM NaCl [Figs. 1(a and b)]. The k_{6766} of this specimen was about two times lower than that for the conventional GCL with unenhanced NaB ($k_{EB-GCL}/k_{GCL} = 0.43$). In contrast, the k_f of 1.4×10^{-10} m/s for the CMCLV5DS specimen was almost the same as that (1.6×10^{-10} m/s) for the conventional GCL specimen (Table 2).

For tests conducted with specimens prepared by dry mixing with CMC [Figs. 1(c and d)], i.e., CMCLV5DM and CMCHV5DM, all of the k_{6766} values based on permeation with either 500 mM NaCl or 167 mM CaCl₂ were lower than the respective values for the conventional GCL (i.e., $0.26 \leq k_{EB-GCL}/k_{GCL} \leq 0.43$), except for the CMCHV5DM specimen permeated with 167 mM CaCl₂, which

resulted in $k_{EB-GCL}/k_{GCL} = 24.5$ (Table 3). In this case, the hydraulic conductivity increased over the course of 10 PVF to a final hydraulic conductivity (k_f) of 1.8×10^{-8} m/s ($k_{6766} = 1.4 \times 10^{-8}$ m/s). As illustrated subsequently, this high hydraulic conductivity was associated with preferential flow in one area of that specimen, which likely occurred due to the elution of polymer (e.g., Rowe and Hamdan 2021).

The same trend and behavior in hydraulic conductivity was observed for the CMCLV5DM specimen permeated with 167 mM CaCl₂, such that the k_f of 3.1×10^{-9} m/s also was greater than that of 4.8×10^{-10} m/s for the conventional GCL, even though the k_{6766} of 1.4×10^{-10} m/s was lower than that of 5.5×10^{-10} m/s for the conventional GCL (i.e., $k_{EB-GCL}/k_{GCL} = 0.26$). These results illustrate that the termination criteria in ASTM D6766 can result in significantly unconservative (low) values of hydraulic conductivity for polymer-amended EB-GCLs when the polymer is eluted from the specimen.

For tests conducted with specimens prepared by wet mixing with CMC [Figs. 1(e and f)], the k_{6766} values for the CMCLV5WM and CVCHV5WM specimens permeated with 500 mM NaCl and 167 mM CaCl₂ were lower than the respective values for the conventional GCL (i.e., $k_{EB-GCL}/k_{GCL} = 0.25$, $k_{EB-GCL}/k_{GCL} = 0.72$, $k_{EB-GCL}/k_{GCL} = 0.12$, and $k_{EB-GCL}/k_{GCL} = 0.67$, respectively). The k_f values to 500 mM NaCl of 6.7×10^{-11} m/s for the CMCLV5WM specimen and 3.3×10^{-11} m/s for the CMCHV5WM specimen also were lower relative to that of 1.6×10^{-10} m/s for the conventional GCL specimen, whereas the values of 3.8×10^{-10} and 4.0×10^{-10} m/s to 167 mM CaCl₂ for the CMCHV5WM and CMCLV5WM specimens, respectively, were only slightly lower relative to that of 4.8×10^{-10} m/s for the conventional GCL specimen. Thus, the hydraulic conductivity values of the wet-mixed CMC specimens were lower than those for the conventional GCL.

The temporal trend in hydraulic conductivity for the CMCHV5WM specimen was not the same as that for the CMCHV5DM specimen [Figs. 1(c and d)], but the CMCHV5WM specimen was permeated only for 6.2 PVF compared with 21.4 PVF for the CMCHV5DM specimen, even though both specimens were not terminated before the criteria in ASTM D6766 had been achieved. This difference is likely related, in part, to the difference in the two preparation methods, i.e., wet mixing versus dry mixing.

EB-GCLs Enhanced with PA

The results of the hydraulic conductivity tests for EB-GCLs enhanced with PA are shown in Fig. 2 and summarized in Tables 2 and 3. For tests conducted with specimens prepared using the dry-sprinkling method [Figs. 2(a and b)], the PALW5DS, PALW8DS, PAMW5DS, PAMW8DS, and PAHW5DS specimens were permeated with 500 mM NaCl (Table 2), whereas the PALW5DS, PALW10DS, PAMW5DS, PAMW8DS, PAMW10DS, PAHW5DS, and PAHW8DS specimens were permeated with 167 mM CaCl₂ (Table 3). Based on the results for all these specimens, the percentage of PA necessary to reduce the hydraulic conductivity relative to that for the conventional GCL (i.e., $k_{EB-GCL}/k_{GCL} < 1$) varied with the molecular weight of PA and type of permeant liquid (NaCl versus CaCl₂).

Based on the results for all of these specimens permeated with either 500 mM NaCl or 167 mM CaCl₂, only the PALW5DS specimens permeated with 167 mM CaCl₂ did not achieve a lower hydraulic conductivity than that for the conventional GCL, with the k_{6766} values being essentially the same ($k_{EB-GCL}/k_{GCL} = 1.0$). For all the other specimens, the k_{EB-GCL}/k_{GCL} values were lower than unity, and in some cases, significantly lower. For all the specimens

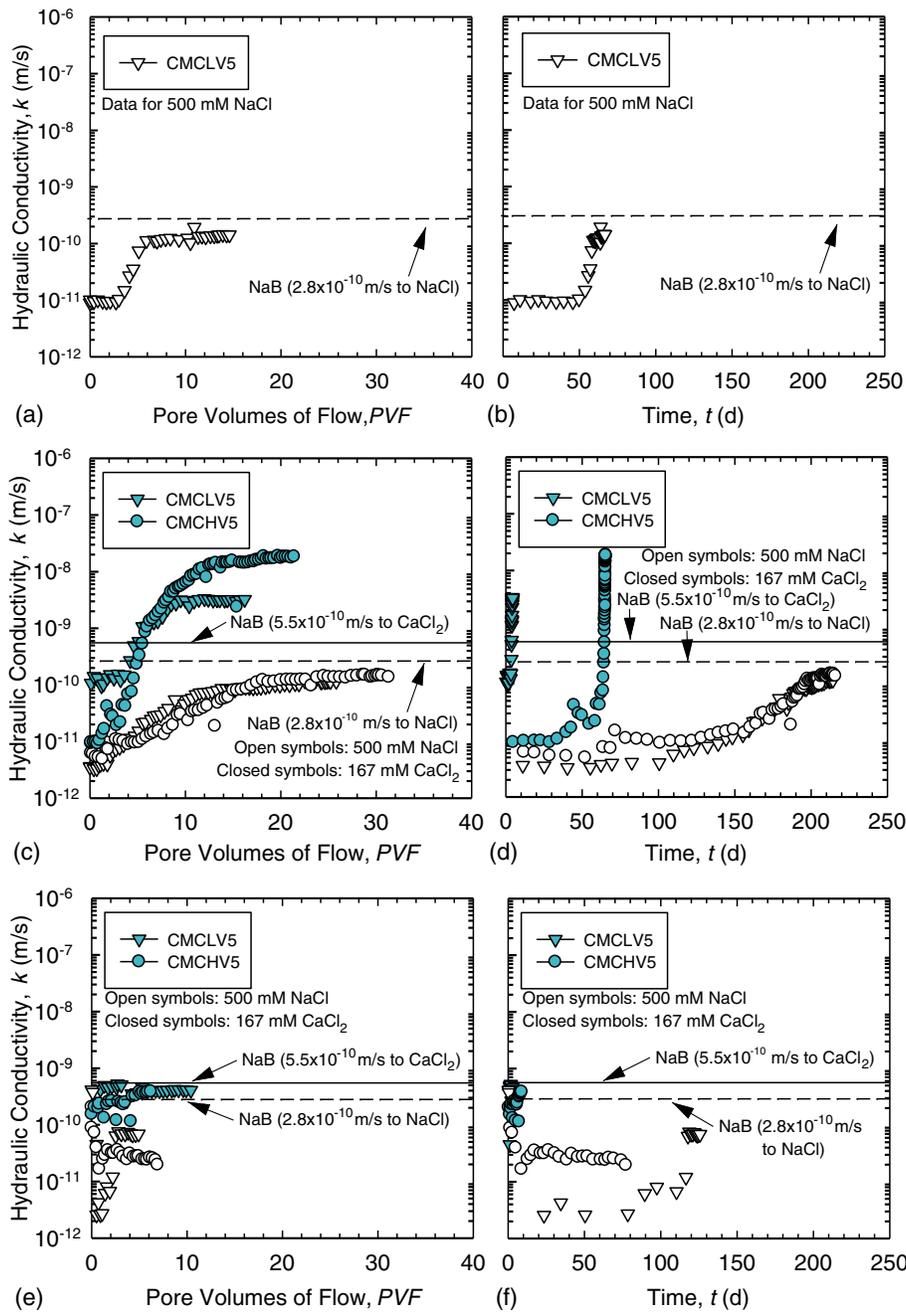


Fig. 1. Hydraulic conductivity test results as a function of pore volumes of flow and elapsed time for EB-GCL specimens comprising sodium carboxymethylcellulose with high viscosity (CMCHV) and low viscosity (CMCLV) at 5% polymer mass loading and permeated with 500-mM NaCl and/or 167-mM CaCl₂ solutions: (a and b) dry-sprinkled specimens; (c and d) dry-mixed specimens; and (e and f) wet-mixed specimens. Standardized hydraulic conductivity (k_{6766}) values for the conventional GCL specimens comprising unenhanced sodium bentonite (NaB) permeated with 500 mM NaCl and 167 mM CaCl₂ also are indicated.

permeated with 500 mM NaCl, $0.046 \leq k_{EB-GCL}/k_{GCL} \leq 0.86$, whereas for all the specimens permeated with 167 mM CaCl₂ (except PALW5DS), $0.020 \leq k_{EB-GCL}/k_{GCL} \leq 0.85$. In general, k_{6766} tended to decrease with increasing molecular weight of PA and/or increasing mass loading of PA.

For permeation with 500 mM NaCl, the greatest decrease in k_{6766} for the EB-GCL specimens relative to that for the conventional GCL specimen occurred with the PAMW5DS, PAMW8DS, and PAHW5DS specimens, i.e., $0.046 \leq k_{EB-GCL}/k_{GCL} \leq 0.10$, versus $k_{EB-GCL}/k_{GCL} = 0.86$ and 0.58 for the PALW5DS and PALW8DS specimens, respectively. Thus, the higher-molecular-weight PA

specimens resulted in significantly lower k_{6766} values. However, polymer mass loading did not have as significant an effect as polymer molecular weight because the k_{EB-GCL}/k_{GCL} for the PAHW5DS specimen was 0.046, which was about half the values of 0.093 and 0.10 for the PAMW5DS and PAMW8DS specimens, respectively, and a factor of 12.6 less than the value of 0.58 for the PALW8DS specimen.

Similar results were obtained for the specimens permeated with 167 mM CaCl₂. For these specimens, the greatest decreases in k_{6766} for the EB-GCL specimens relative to that for the conventional GCL specimen of $0.020 \leq k_{EB-GCL}/k_{GCL} \leq 0.084$ occurred

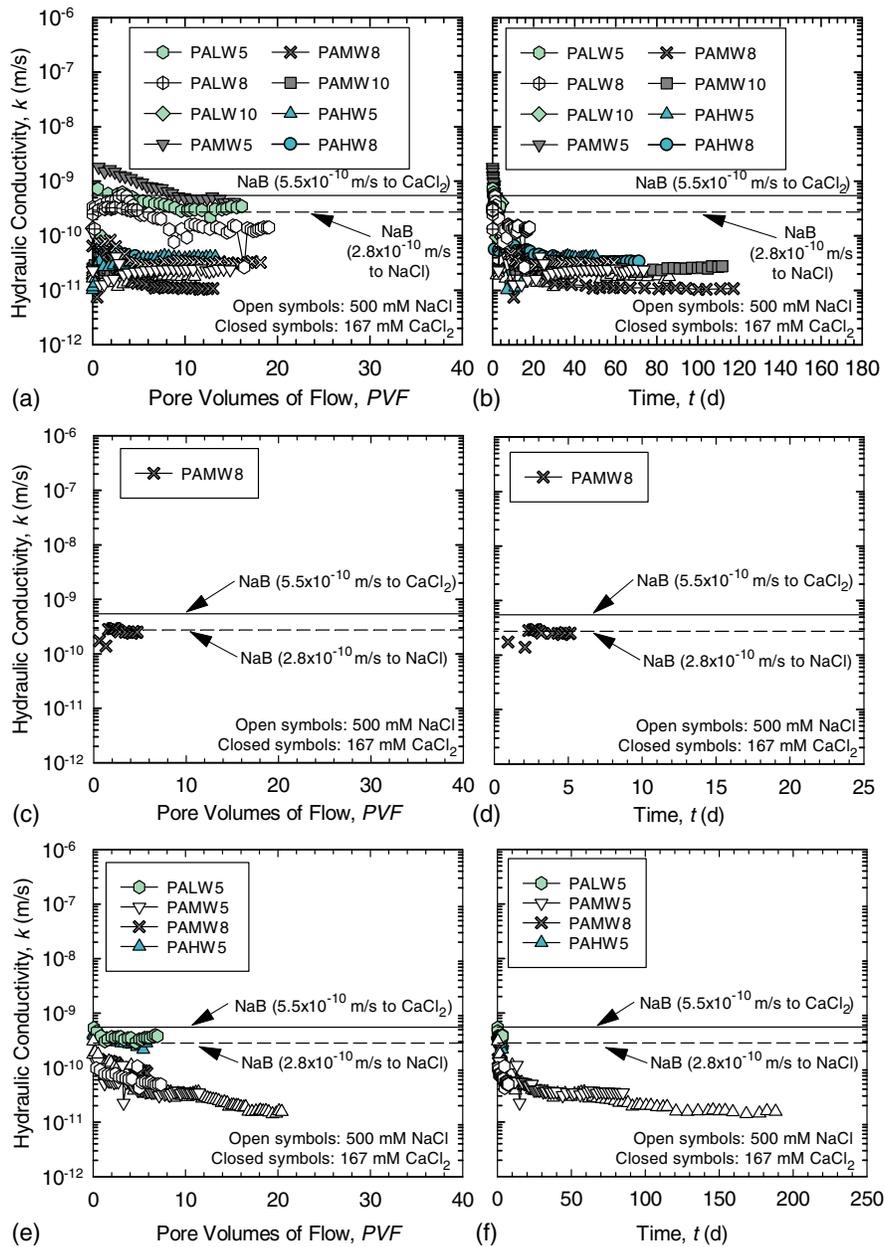


Fig. 2. Hydraulic conductivity test results as a function of pore volumes of flow and elapsed time for EB-GCL specimens comprising sodium poly (acrylic acid) with low molecular weight (PALW), medium molecular weight (PAMW), and high molecular weight (PAHW) at different polymer mass loadings (5%, 8%, or 10%) and permeated with 500-mM NaCl and 167-mM CaCl_2 solutions: (a and b) dry-sprinkled specimens; (c and d) dry-mixed specimens; and (e and f) wet-mixed specimens. Standardized hydraulic conductivity (k_{6766}) values for the conventional GCL specimens comprising unenhanced NaB permeated with 500 mM NaCl and 167 mM CaCl_2 also are indicated.

for the PAMW8DS, PAMW10DS, PAHW5DS, and PAHW8DS specimens versus $0.75 \leq k_{\text{EB-GCL}}/k_{\text{GCL}} \leq 1.0$ for the PALW5DS, PALW10DS, and PAMW5DS specimens. However, in contrast to the results based on permeation with 500 mM NaCl, polymer mass loading apparently had a somewhat greater effect on the results. For example, $k_{\text{EB-GCL}}/k_{\text{GCL}} = 0.85$ for the PAMW5DS specimen permeated with 167 mM CaCl_2 , which was 9.1 times greater than that of 0.093 based on permeation with 500 mM NaCl, whereas $k_{\text{EB-GCL}}/k_{\text{GCL}} = 0.020$ for the PAMW8DS specimen permeated with 167 mM CaCl_2 .

Thus, increasing the polymer mass loading of the dry-sprinkled PAMW specimens from 5% to 8% significantly improved the hydraulic performance of these specimens permeated with 167 mM CaCl_2 , but had minimal effect on the hydraulic performance of the

same specimens permeated with 500 mM NaCl. This requirement for more polymer to achieve a similar hydraulic conductivity may be due to the lower molecular weight of the PAMW (~50,000 g/mol) relative to that of the PAHW (~345,000 g/mol), which equates to a reduction in polymer chain length. For example, increasing the mass loading of PAHW for the dry-sprinkled EB-GCL specimens from 5% to 8% did not significantly impact the hydraulic performance of these specimens permeated with 167 mM CaCl_2 , with a k_{6766} of 4.0×10^{-11} m/s at 5% versus 4.6×10^{-11} m/s at 8%.

An additional test was performed on a specimen prepared by dry mixing with 8% of PAMW, i.e., PAMW8DM, and permeated with 167 mM CaCl_2 [Figs. 2(c and d)]. The behavior was similar to that for the PAMW8DS specimen. However, unlike the results for

the PAMW8DS specimen, the hydraulic conductivity of the PAMW8DM specimen increased after approximately 2.4 PVF and plateaued at approximately 2.5×10^{-10} m/s, which was 25 times greater than the hydraulic conductivity of 1.1×10^{-11} m/s for the PAMW8DS specimen to the same solution (Table 3). This difference in behavior likely reflects the difference in specimen preparation method (dry mixing versus dry sprinkling).

For tests conducted with specimens prepared using the wet-mixing method [Figs. 2(e and f)], the PALW5WM, PAMW5WM, and PAHW5WM specimens were permeated with 500 mM NaCl (Table 2), whereas the PALW5WM, PAMW8WM, and PAHW5WM specimens were permeated with 167 mM CaCl₂ (Table 3). Comparisons of the results for the two specimens with 5% polymer mass loading permeated with 500 mM NaCl and the two specimens with 5% polymer mass loading and permeated with 167 mM CaCl₂ (PALW5WM and PAHW5WM) provide a direct assessment of the effect of the polymer molecular weight on the hydraulic conductivity with respect to each permeant liquid, a comparison of the results for the PALW5WM specimen and the PAHW5WM specimen based on permeation with 500 mM NaCl versus those based on permeation with 167 mM CaCl₂ provided a direct assessment of the effect of type of permeant liquid.

All of the wet-mixed EB-GCL specimens permeated with 500 mM NaCl resulted in similar k_{6766} values that were significantly lower than that for the conventional GCL, i.e., $0.17 \leq k_{EB-GCL}/k_{GCL} \leq 0.28$, whereas the k_{6766} values for the two specimens with 5% polymer mass loading (PALW5WM, PAMW8WM, and PAHW5WM) permeated with 167 mM CaCl₂ also were similar and lower than that for the conventional GCL, i.e., $0.15 \leq k_{EB-GCL}/k_{GCL} \leq 0.60$. Thus, for the wet-mixed EB-GCL specimens, the molecular weight of the polymer had little effect on the improvement in hydraulic performance of the EB-GCLs relative to that for the conventional GCL, although the improvement was less significant in the case of permeation with 167 mM CaCl₂ versus 500 mM NaCl. Moreover, for the PALW5WM specimen, permeation with 500 mM NaCl resulted in $k_{EB-GCL}/k_{GCL} = 0.28$ whereas permeation with 167 mM CaCl₂ resulted in $k_{EB-GCL}/k_{GCL} = 0.60$.

Similarly, for the PAHW5WM specimen, permeation with 500 mM NaCl resulted in $k_{EB-GCL}/k_{GCL} = 0.19$ whereas permeation with 167 mM CaCl₂ resulted in $k_{EB-GCL}/k_{GCL} = 0.56$. Thus, the wet-mixed EB-GCL specimens were less effective based on permeation with 167 mM CaCl₂ relative to 500 mM NaCl. Nonetheless, all the wet-mixed EB-GCL specimens resulted in improved hydraulic performance relative to that for the conventional GCL. Also, an initial decreasing temporal trend in hydraulic conductivity occurred for several of the wet-mixed EB-GCL specimens [e.g., Figs. 1(e and f) and 2(e and f)]. These decreasing trends in hydraulic conductivity were similar to those exhibited by EB-GCLs comprising HYPER clay, which is a CMC-enhanced bentonite prepared via a more advanced wet-mixing method (e.g., Di Emidio et al. 2015).

Tests conducted with the wet-mixed EB-GCLs enhanced with PA [Figs. 2(e and f)] behaved differently from those conducted with the dry-sprinkled EB-GCLs enhanced with PA. For example, for permeation with 167 mM CaCl₂, the hydraulic conductivity of 3.1×10^{-10} m/s for the PAHW5WM specimen was approximately 7.8 times higher than that of 4.0×10^{-11} m/s for the PAHW5DS specimen, and the hydraulic conductivity of 8.5×10^{-11} m/s for the PAMW8WM specimen was approximately 7.7 times higher than that of 1.1×10^{-11} m/s for the PAMW8DS specimen. Also, for permeation with 500 mM NaCl, the hydraulic conductivity of 4.8×10^{-11} m/s for the PAMW5WM specimen was 1.8 times higher than that of 2.6×10^{-11} m/s for the PAMW5DS specimen, whereas the hydraulic conductivity of 5.2×10^{-11} m/s for the

PAHW5WM specimen was 4.0 times higher than that of 1.3×10^{-11} m/s for the PAHW5DS specimen.

In contrast, the hydraulic conductivity to 500 mM NaCl of 7.8×10^{-11} m/s for the PALW5WM specimen was 3.1 times lower than that of 2.4×10^{-10} m/s for the PALW5DS specimen. Thus, in general, the hydraulic conductivity of the wet-mixed specimens was higher than that of the dry-sprinkled specimens, with the lone exception occurring for the lowest molecular weight PA at the lowest polymer mass loading (i.e., PALW5). These results indicate that the method of specimen preparation, i.e., dry versus wet, can affect the hydraulic conductivity of PA-based EB-GCLs.

EB-GCLs Enhanced with PAX

The results of the hydraulic conductivity tests for EB-GCLs enhanced with PAX are shown in Figs. 3(a and b) and summarized in Tables 2 and 3. These specimens, designated as PAX5DS, were prepared by dry-sprinkling the NaB with 5% PAX by mass. The hydraulic conductivity of the PAX5DS specimen permeated with 500 mM NaCl was 2.5×10^{-11} m/s (Table 2), which is about an order of magnitude lower than the value of 2.8×10^{-10} m/s for the conventional GCL specimen (i.e., $k_{EB-GCL}/k_{GCL} = 0.089$). The hydraulic conductivity of the PAX5DS specimen permeated with 167 mM CaCl₂ was approximately 2.1×10^{-10} m/s (Table 3), which is about 2.4 times lower than the value of 5.5×10^{-10} m/s for the conventional GCL specimen (i.e., $k_{EB-GCL}/k_{GCL} = 0.35$). In comparison, the values of k_{EB-GCL}/k_{GCL} for the PAMW5DS and PAHW5DS specimens based on permeation with 500 mM NaCl were 0.093 and 0.046, respectively, whereas those for the PALW5DS, PAMW5DS, and PAHW5DS specimens based on permeation with 167 mM CaCl₂ were 1.0, 0.85, and 0.073, respectively. Thus, the use of the covalently crosslinked PA does not appear to have been as effective as the linear PA with respect to the 500-mM NaCl solution. However, with respect to the 167-mM CaCl₂ solution, the PAX appears to be more effective than the PALW or PAMW and similar in effectiveness to PAHW.

EB-GCLs Enhanced with BPC

The results of the hydraulic conductivity tests for EB-GCLs enhanced with BPC are shown in Figs. 3(c and d) and summarized in Tables 2 and 3. The specimens, designated as BPC5DM, were prepared by dry mixing the BPC with the NaB to achieve a polymer mass loading of 5%. The k_{6766} values of the specimens to both 500 mM NaCl and 167 mM CaCl₂ were 1.1×10^{-11} and 3.2×10^{-11} m/s, respectively, which reflected a significantly better hydraulic performance for the EB-GCL relative to that of the conventional GCL (i.e., $k_{EB-GCL}/k_{GCL} = 0.039$ and $k_{EB-GCL}/k_{GCL} = 0.058$, respectively). By comparison, the k_{EB-GCL}/k_{GCL} values for the best-performing EB-GCLs comprising 5% of PA were 0.093 and 0.046 for the PAMW5DS and PAHW5DS specimens, respectively, based on permeation with 500 mM NaCl and 0.073 for the PAHW5DS specimen based on permeation with 167 mM CaCl₂. Thus, the hydraulic performance of the specimen based on in situ polymerization to produce the BPC was no better than that of the best performing EB-GCLs comprising 5% of PA.

Polymer Retention and Elution

All EB-GCLs permeated in this study eluted a fraction of polymer during permeation, indicating that complete polymer retention was not achieved regardless of the preparation method, polymer type, or polymer mass loading. The PA and CMC polymers are water soluble and can increase solution viscosity with increasing polymer concentration. However, as shown in Fig. 4, the crosslinking and

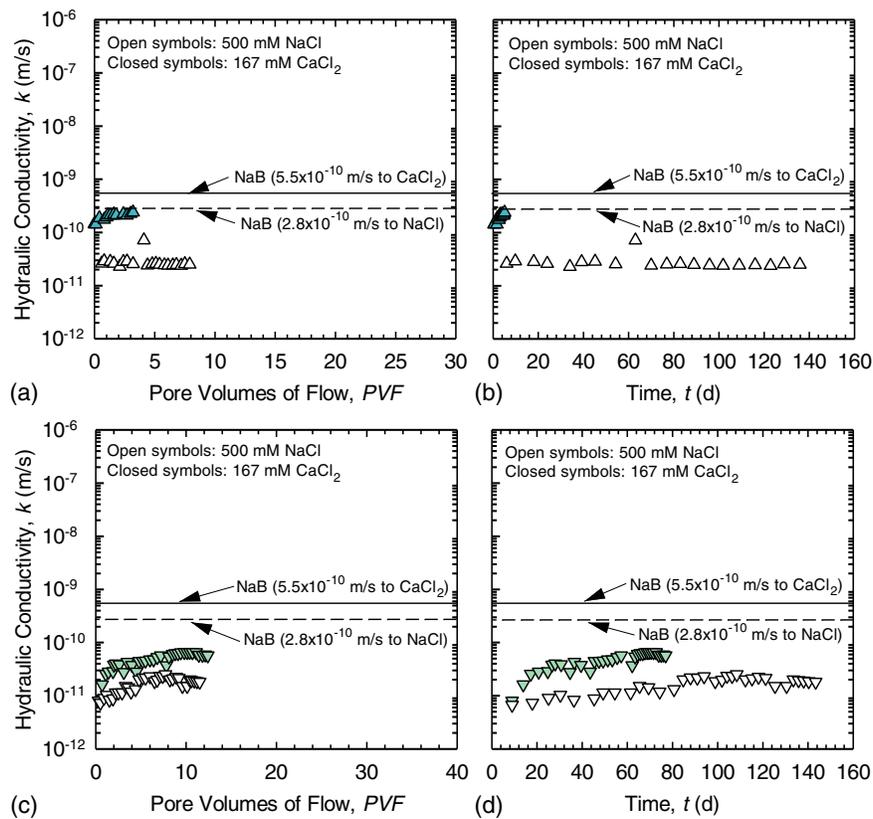


Fig. 3. Hydraulic conductivity test results as a function of pore volumes of flow and elapsed time for EB-GCL specimens comprising of dry-sprinkled (a and b) covalently crosslinked sodium polyacrylate (PAX); and (c and d) bentonite polymer composite (BPC) at 5% polymer mass loading and permeated with 500-mM NaCl and 167-mM CaCl₂ solutions. Standardized hydraulic conductivity (k_{6766}) values for the conventional GCL specimens comprising unenhanced NaB permeated with 500 mM NaCl and 167 mM CaCl₂ also are indicated.

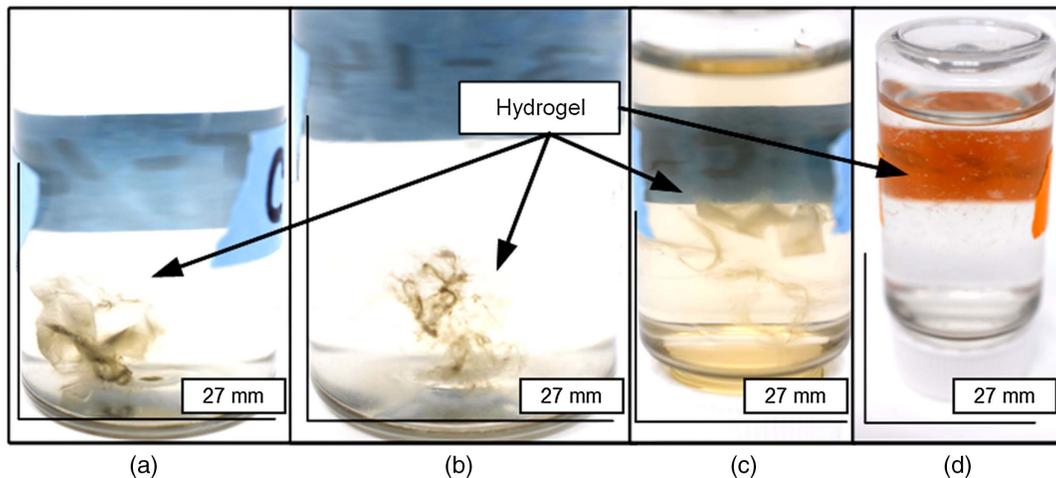


Fig. 4. Hydrogel strands within effluent sample bottles from the PALW5DS specimens permeated with (a and b) 167 mM CaCl₂; (c) 500 mM NaCl; and (d) the CMCHV5DM specimen permeated with 500 mM NaCl.

formation of visible polymer strands, or hydrogel, was evident in the effluent samples of the EB-GCLs prepared with PA and CMC. Hydrogel has been hypothesized to form primarily due to divalent cations (predominantly Ca²⁺) available in the permeant liquid that ionically bond and crosslink multiple anionic polymer chains (Scalia et al. 2014, 2018; Tian et al. 2019). The soluble Ca²⁺ and, to a lesser extent, Mg²⁺ within the base NaB as well as the Ca²⁺ in the CaCl₂ permeant liquid [Figs. 4(a and b)] provided cations for ionic

crosslinking. Due to the existence of multivalent cations in the pore water and on the exchange complex of the NaB, principally Ca²⁺ (Table S1), crosslinking also can occur in the EB-GCLs permeated with 500 mM NaCl [Figs. 4(c and d)]. However, crosslinking via cations creates a transient network that can be altered due to changes in solution pH, cation concentrations, or application of shear forces (Buchholz and Graham 1998), such as by varying the hydraulic gradient applied during hydraulic conductivity testing.

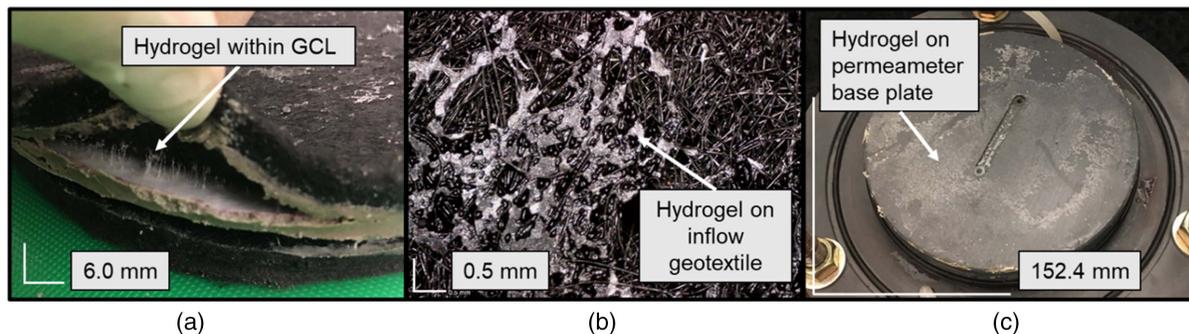


Fig. 5. Visible crosslinked polymer in hydraulic conductivity testing of EB-GCL specimens: (a and b) PAHW5DS permeated with 167 mM CaCl_2 ; and (c) PAMW8DS permeated with 167 mM CaCl_2 .

The existence of hydrogel in the effluent does not necessarily confirm that the same hydrogel existed within the interparticle, intra-aggregate, and/or interaggregate pores of the EB-GCL specimens during permeation. However, as shown in Fig. 5, hydrogel was present on the inflow sides of the nonwoven geotextile and base plate of the permeameter. Polymer migration toward the inflow side likely began during hydration as a result of diffusion resulting from a dissolved polymer concentration gradient [similar to the polymer elution reported by Rowe and Hamdan (2021)], especially in the case of EB-GCLs prepared via dry-sprinkling, where the polymer is placed on the inflow side of the GCL. Specimens that exhibited hydrogel formation on the nonwoven geotextiles or within the permeameter are indicated with respect to the k_f values in Tables 2 and 3. The presence of hydrogel within the permeameter (e.g., on the base plate and geotextiles) as well as in the effluent implies that hydrogel also was present within the interparticle, intra-aggregate, and/or interaggregate pores of the EB-GCL specimens. These observations support the hypothesis previously postulated by Scalia et al. (2014, 2018) and Tian et al. (2016a, b, 2017, 2019) that clogging of pores with polymer hydrogel is the primary mechanism reducing the hydraulic conductivity of EB-GCLs relative to the conventional GCL comprising unenhanced NaB.

Preferential Flow

Tests where preferential flow was indicated by the presence of concentrated rhodamine dye on the outflow side of the specimen are indicated with respect to the k_f values in Tables 2 and 3. Preferential flow was evident under two conditions. First, when the hydraulic conductivity of the EB-GCL was unexpectedly higher than that for the conventional GCL with the same permeant liquid, the tests were dyed as previously described, and the specimen was inspected for evidence of preferential flow paths upon termination of the test. If preferential flow was observed to be the result of setup error, such as being caused by geotextiles pinching together during setup due to uneven bentonite loading, then the test was considered unacceptable and repeated. Data from tests that were deemed unacceptable are not reported herein.

Second, preferential flow was also manifested by a steadily increasing temporal trend in hydraulic conductivity, resulting in a final hydraulic conductivity higher than the baseline hydraulic conductivity of the conventional GCL. This preferential flow behavior was evident for the CMCLV5DM and CMCHV5DM specimens permeated with 167 mM CaCl_2 , where the dye revealed a preferential flow path in the center of these specimens. Both EB-GCLs were prepared by the dry-mixing method, resulting in a mixture of polymer granules within the base NaB (Fig. S2). The increasing trend in hydraulic conductivity observed for these specimens

[Figs. 1(c and d)] is believed to be due to polymer elution. In the dyed portion of the specimen, the polymer network likely created an interconnected path from the inflow to the outflow side of the specimens. As the polymer was eluted, the flow likely occurred through the pathway opened by the loss of polymer. Although the initial results at ~ 2 PVF for the PAMW8DM specimen permeated with 167 mM CaCl_2 exhibited an increasing trend in hydraulic conductivity, the hydraulic conductivity eventually plateaued at 2.5×10^{-10} m/s. Additional hydraulic conductivity testing of EB-GCLs prepared by dry mixing with CMC and PA at higher polymer contents is required to verify the role of the dry-mixing method or polymer type on preferential flow.

Effect of Hydraulic Gradient on Hydraulic Conductivity

Results of the tests performed at a lower hydraulic gradient of ~ 35 based on permeation with 167 mM CaCl_2 are summarized in Table 3. The results of the lower gradient tests can be found in Fig. S5. Two distinct behaviors are reflected in the trends in hydraulic conductivity. First, the hydraulic conductivity of the PAHW5DS specimen was about 3.6 times lower at the lower hydraulic gradient, i.e., 1.1×10^{-11} m/s versus 4.0×10^{-11} m/s. In contrast, the hydraulic conductivity of the PALW5DS specimen at the hydraulic gradient of ~ 35 of 1.3×10^{-11} m/s was approximately 1.5 orders of magnitude lower than that of 5.7×10^{-10} m/s at the hydraulic gradient of ~ 300 . The decrease in hydraulic conductivity with decrease in hydraulic gradient likely indicates that the polymer was less mobile at a lower hydraulic gradient (i.e., lower seepage force).

Polymer hydrogel was visible on both specimens postpermeation. Nonetheless, polymer hydrogel clogged the outflow end of the PALW5DS specimen tested at a lower hydraulic gradient of ~ 35 after approximately 135 days of permeation [Fig. S5(b)], illustrating that a low hydraulic gradient did not prevent mobilization of the polymer. This clogging occurred after the test was paused (closed) during lab closure due to restrictions caused by the COVID-19 pandemic. The polymer obstruction was removed from the outflow tubing using pipe cleaners, and the test was resumed, resulting in a re-establishing of the previously measured, higher hydraulic conductivity.

Discussion

Effects of Polymer Properties and Specimen Preparation Methods

The hydraulic conductivity (k_{6766}) values for all the EB-GCL specimens are shown in Fig. 6 as a function of polymer type and mixing

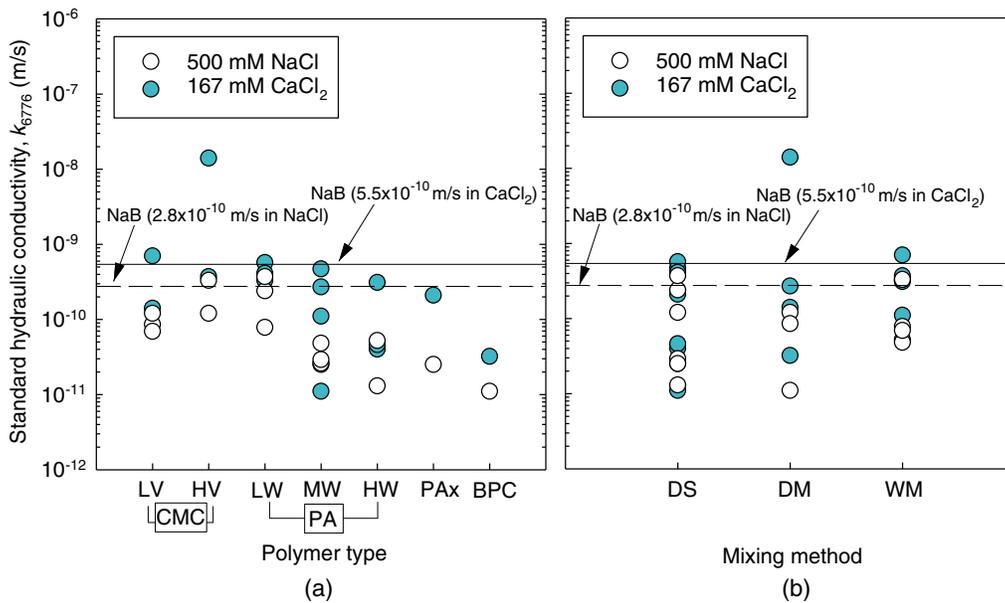


Fig. 6. Standard hydraulic conductivity (k_{6776}) as a function of (a) polymer type [i.e., CMC with low viscosity (LV) or high viscosity (HV) and PA with low, medium, or high molecular weight (LW, MW, HW, respectively), covalently crosslinked sodium polyacrylate and BPC]; and (b) method of specimen preparation [dry sprinkle (DS), dry mixing (DM), and wet mixing (WM)]. Standardized hydraulic conductivity (k_{6766}) values for the conventional GCL comprising unenhanced NaB permeated with 500 mM NaCl and 167 mM CaCl_2 also are indicated.

method. Overall, the hydraulic performance of the EB-GCLs to the NaCl solution was better than that to the CaCl_2 solution [Fig. 6(a)]. Unexpectedly, increasing PA molecular weight did not correlate necessarily with decreasing hydraulic conductivity because the lowest hydraulic conductivity of the EB-GCLs with PA occurred for the PAMW8DS specimen. In contrast, the PALW was not effective in reducing hydraulic conductivity relative to that of unenhanced NaB, even at the highest polymer mass loading of 10%. As previously mentioned, considerations need to be given to the final polymer content of the specimen after permeation as well as the effective initial distribution of polymer across specimen during preparation. Polymer elution, a thin layered section, or gap in polymer placement in an EB-GCL, or all, may result in a higher hydraulic conductivity.

As shown in Fig. 6(a), the viscosity of the CMC did not affect the hydraulic conductivity. The decrease in hydraulic conductivity of the CMC-enhanced GCLs relative to that for the conventional GCL, which was approximately one order of magnitude or less, was not as significant as that for the other EB-GCLs, which was greater than one order of magnitude.

The structural differences between PA and CMC may be a controlling factor in the differences in behavior. The crosslinking potential for the CMC may be lower than that for the PAs due to a lower number of carboxyl groups, which depends on the DoS. In this case, CMC bridging to bound cations within the interparticle, intra-aggregate, and/or interaggregate pore matrix of the EB-GCLs would be lessened, resulting in less retention of the CMC and a higher hydraulic conductivity.

As shown in Fig. 6(b), the dry-mixed EB-GCL comprising 5% of BPC (BPC5DM) resulted in the lowest hydraulic conductivity to 500 mM NaCl of 1.1×10^{-11} m/s, with the dry-sprinkled EB-GCL comprising 5% of covalently crosslinked PAx (PAx5DS) resulting in a hydraulic conductivity only 2.3 times greater (2.5×10^{-11} m/s). In contrast, the hydraulic conductivity for the PAx5DS specimen permeated with 167 mM CaCl_2 of 1.9×10^{-10} m/s was higher than that achieved for the PAMW8DS (1.1×10^{-11} m/s) and PAHW5DS (4.0×10^{-11} m/s) specimens.

Based on the results of this study, the PAx may result in a suitably low hydraulic conductivity when permeated with a concentrated NaCl solution but did not appear to offer any advantage relative to PAHW or PAMW (at PAMW concentrations higher than 5%) when permeated with a concentrated CaCl_2 solution.

As shown in Fig. 6(b), all the hydraulic conductivity values for the dry-sprinkled EB-GCLs except for one were lower than those for the conventional GCL, with the lone exception being the PALW5DS specimen, with a hydraulic conductivity to 167 mM CaCl_2 that was essentially the same as that for the conventional GCL specimen (Table 3). Due to the location of the polymer at the inflow side of the dry-sprinkled specimens (Fig. S2), if the polymer failed to mobilize and clog the pores of the adjacent NaB, then the hydraulic conductivity was governed solely by the NaB. In contrast, dry or wet mixing the polymer with the NaB led to the potential for incomplete distribution of the polymer throughout the specimen during preparation, especially with a lower polymer mass loading. In this case, elution of the polymer may lead to a preferential flow path resulting in a hydraulic conductivity that is higher than that of the conventional GCL due to flow occurring through larger interaggregate pores created by the polymer elution.

Effect of Polymer Mass Loading

The effect of polymer mass loading on the hydraulic conductivity (k_{6766}) of all the EB-GCL specimens is illustrated in Fig. 7 based on permeation with 500 mM NaCl [Figs. 7(a, c, and e)] and 167 mM CaCl_2 [Figs. 7(b, d, and f)]. For the dry-sprinkled prepared specimens [Figs. 7(a and b)], the lowest hydraulic conductivity to both permeant liquids for specimens at 5% polymer mass loading occurred for the EB-GCL specimens with PAHW, whereas the lowest hydraulic conductivity for either 8% or 10% polymer mass loadings to both permeant liquids occurred for the EB-GCL specimens with PAMW. The overall lowest hydraulic conductivity based on permeation with 500 mM NaCl of 1.3×10^{-11} m/s occurred for the specimen with 5% of PAHW [Fig. 7(a)], whereas the overall lowest hydraulic conductivity based on permeation with 167 mM

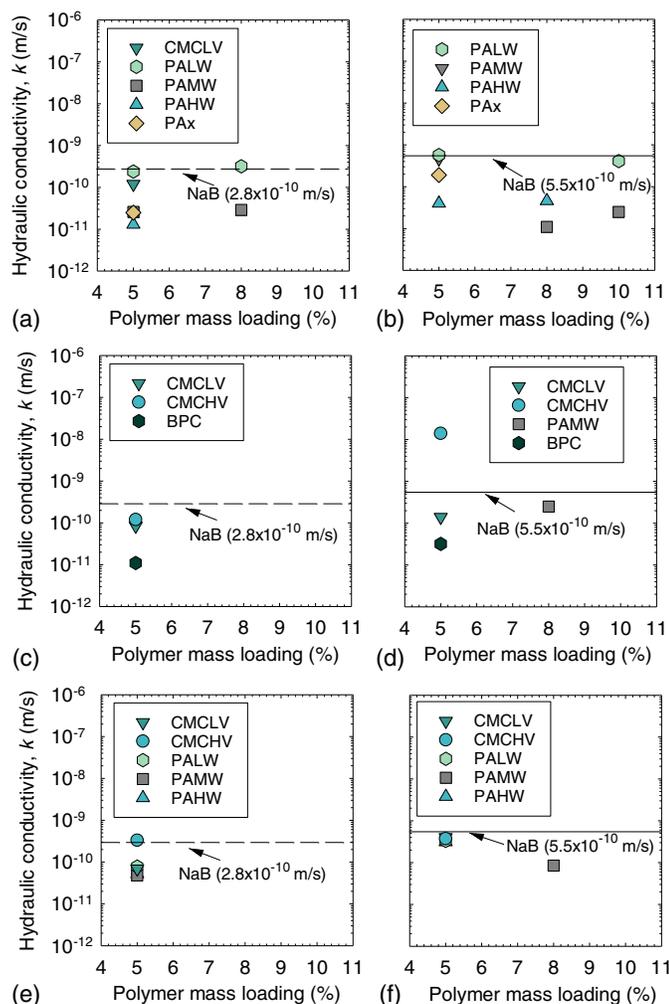


Fig. 7. Effect of polymer mass loading on hydraulic conductivity (k_{6766}) of EB-GCL specimens prepared by (a and b) dry sprinkling; (c and d) dry mixing; and (e and f) wet mixing of CMCHV and CMCLV, PALW, PAMW, and PAHW, covalently crosslinked sodium PAX, and BPC and permeated with (a, c, and e) 500 mM NaCl; and (b, d, and f) 167 mM CaCl_2 . Standardized hydraulic conductivity (k_{6766}) values for the conventional GCL comprising unenhanced NaB permeated with 500 mM NaCl and 167 mM CaCl_2 also are indicated.

CaCl_2 of 1.1×10^{-11} m/s occurred for the specimen with 8% of PAMW [Fig. 7(b)], which was also the overall lowest hydraulic conductivity to either permeant liquid.

The dry-sprinkling method used in this study was implemented by hand such that the results may highlight variations in the distribution of polymer for a given specimen. Replicate tests are required to confirm whether these results reflect a limitation to the reduction of hydraulic conductivity with increasing polymer loading or are an artifact of the variation of polymer distribution resulting from the dry-sprinkling method.

As shown in Figs. 7(c and e), no conclusions can be drawn with respect to the effect of polymer mass loading on the hydraulic conductivity of the dry-mixed or wet-mixed EB-GCL specimens permeated with 500 mM NaCl because only the 5% polymer mass loading was evaluated for these specimens. The data also are limited with respect to the dry-mixed and wet-mixed EB-GCL specimens permeated with 167 mM CaCl_2 [Figs. 7(d and f)], with all but one of the specimens comprising 5% polymer mass loading.

In terms of the results for the dry-mixed specimens [Figs. 7(c and d)], the EB-GCL enhanced with 5% BPC resulted in the overall lowest hydraulic conductivity to either permeant liquid of 1.1×10^{-11} m/s based on permeation with 500 mM NaCl (Table 2). An interesting observation is that this hydraulic conductivity value was essentially the same as the aforementioned lowest values to either permeant liquid for specimens dry-sprinkled with 5% of PAHW (500 mM NaCl) or 8% of PAMW (167 mM CaCl_2).

For the wet-mixed specimens enhanced with 5% polymer mass loading and permeated with 500 mM NaCl [Fig. 7(e)], all the measured hydraulic conductivity values were similar (4.8×10^{-11} m/s $\leq k_{6766} \leq 7.8 \times 10^{-11}$ m/s) and were similarly lower than that of 2.8×10^{-10} m/s for conventional GCL, except for the specimen enhanced with CMCHV with a hydraulic conductivity of 3.3×10^{-10} m/s (Table 2). In contrast, none of the wet-mixed specimens enhanced with 5% polymer and permeated with 167 mM CaCl_2 resulted in hydraulic conductivity values that were appreciably different from that of 5.5×10^{-10} m/s for the conventional GCL, where the specimen enhanced with 8% PAMW produced a somewhat lower hydraulic conductivity ($k_{\text{EB-GCL}}/k_{\text{GCL}} = 0.15$). Overall, simply increasing the polymer mass loading does not necessarily result in lower hydraulic conductivity because other factors such as type of polymer and method of specimen preparation may control the hydraulic behavior.

Effect of Hydraulic Gradient on Hydraulic Conductivity

Hydraulic conductivity testing of conventional GCLs at elevated hydraulic gradients (>200) is common to reduce the testing duration (e.g., Shackelford et al. 2000). Although an increase in hydraulic gradient typically corresponds to an increase in effective stress in the specimen and a concomitant lower hydraulic conductivity, multiple studies have shown that the magnitude of hydraulic gradient up to values as high as 550 resulted in a relatively minor effect on the measured hydraulic conductivity of conventional GCLs (Rad et al. 1994; Petrov et al. 1997; Shackelford et al. 2000). However, as previously mentioned, a decrease in the applied hydraulic gradient from ~ 300 to ~ 35 for the PALW5DS and PAHW5DS specimens resulted in a decrease in hydraulic conductivity of 1.5 orders of magnitude and approximately 3.6 times, respectively. This behavior suggests that the PALW was less mobile at a lower hydraulic gradient and more effective in terms of achieving a low hydraulic conductivity due to a reduction in polymer elution. The lower hydraulic conductivity at the lower hydraulic gradient suggests that, unlike conventional GCLs, the magnitude of the hydraulic gradient may affect the measured hydraulic conductivity of EB-GCLs, and the effect may be opposite to that expected based on effective stress considerations. This sensitivity to hydraulic gradient was consistent with the recognized shear thinning of polymers under higher shear stresses, such as under higher hydraulic gradients (e.g., Markovitz and Kimball 1950; Van Krevelen 1990). Nonetheless, because these results are limited, additional study is needed to understand the influence of hydraulic gradient on the measured hydraulic conductivity of EB-GCLs.

Conclusions

Hydraulic conductivity tests were conducted in accordance with ASTM D6766 on specimens of a conventional GCL comprising a natural (unenhanced) powdered NaB and EB-GCLs comprising the NaB enhanced with one of several anionic polymers at mass loadings of 5%, 8%, or 10%. Three methods were used to prepare

the EB-GCL specimens, namely, dry sprinkling, dry mixing, and wet mixing. The polymers included CMC with low and high viscosity, linear PA with low, medium, and high molecular weights of approximately 5,000, 50,000, and 345,000 g/mol, respectively, and a covalently crosslinked PA. An EB previously produced by in situ polymerization to include 28.5% by mass of polyacrylate, known as BPC, was also used to prepare dry-mixed EB-GCL specimens at a polymer mass loading of 5% (i.e., an EB with 17.3% of BPC). The hydraulic conductivity tests were conducted at low effective stress (27 kPa) using concentrated electrolyte solutions (500 mM NaCl and 167 mM CaCl₂) with high ionic strength ($I = 500$ mM) to represent aggressive chemical conditions. Qualitative polymer elution analysis was performed based on observations of hydrogel formation and linear polymer crosslinking.

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

- EB-GCLs comprising anionic polymers at low polymer mass loadings ($\leq 10\%$) and prepared using different mixing methods have the potential to improve chemical incompatibility relative to conventional GCLs with natural (unenhanced) NaB based on permeation with high-ionic-strength (500 mM) electrolyte solutions.
- The measured hydraulic conductivity of the EB-GCLs prepared in this study with anionic polymers were affected by the type and properties (molecular weight and viscosity) of the polymer, the polymer mass loading (i.e., 5%, 8%, and 10%), and the method of specimen preparation, i.e., dry sprinkling, dry mixing, or wet mixing. However, the benefits of an increase in molecular weight or percentage enhancement in resisting hydraulic incompatibility and resulting in a lower hydraulic conductivity relative to that for the conventional GCL comprising the unenhanced NaB were limited. Thus, limitlessly increasing either or both of these parameters will not necessarily continuously decrease the hydraulic conductivity.
- The overall best hydraulic performance for the EB-GCLs prepared in this study with linear anionic polymers occurred for those prepared by dry sprinkling with 5% of PAHW and 8% of PAMW, with a hydraulic conductivity of 1.3×10^{-11} m/s to 500 mM NaCl and 4.0×10^{-11} m/s to 167 mM CaCl₂ for the specimen with PAHW, and 2.9×10^{-11} m/s to 500 mM NaCl and 1.1×10^{-11} m/s to 167 mM CaCl₂ for the specimen with PAHW. The EB-GCL enhanced with 5% of BPC by dry mixing resulted in similarly good performance with a hydraulic conductivity of 1.1×10^{-11} m/s to 500 mM NaCl and 3.2×10^{-11} m/s to 167 mM CaCl₂. Thus, low hydraulic conductivity ($\leq 4.0 \times 10^{-11}$ m/s) of EB-GCLs to aggressive inorganic chemical solutions can be achieved with low mass percentage polymer enhancement.
- The dry-sprinkling method of specimen preparation provided the best hydraulic performance for the EB-GCLs with the least amount of required effort of the mixing methods tested. The dry- and wet-mixing methods result in the polymer being dispersed within the matrix of particles and aggregates of particles such that, if the polymer was eluted upon permeation, a preferential flow path through the EB-GCL could be created resulting in an increase in hydraulic conductivity.
- All EB-GCLs tested in this study eluted a fraction of polymer during permeation. Observations of hydrogel in multiple effluent samples and on postpermeated EB-GCLs specimens supported pore clogging by the hydrogel within the pore network of EB-GCLs as the primary mechanism for low hydraulic conductivity to the aggressive permeant liquids used in this study. However, preferential interaggregate flow paths that correlated with polymer elution and higher hydraulic conductivity

also were identified in multiple EB-GCL specimens, emphasizing the importance of polymer retention for maintaining the low hydraulic conductivity of the EB-GCLs.

- Tests conducted at a lower hydraulic gradient of ~ 35 with EB-GCL specimens prepared by dry sprinkling with 5% of PALW and 5% of PAHW and permeated with 167 mM CaCl₂ resulted in a lower hydraulic conductivity. For the EB-GCL enhanced with PALW, the hydraulic conductivity at the lower hydraulic gradient was 1.5 orders of magnitude lower than that the higher hydraulic gradient of ~ 300 , whereas for the EB-GCL enhanced with PAHW, the hydraulic conductivity at lower hydraulic gradient was approximately 3.6 times lower than that at the higher hydraulic gradient. These results suggest that the magnitude of the hydraulic gradient may affect the measured hydraulic conductivity of EB-GCLs and, unlike conventional GCLs, the effect may be opposite to that expected based on effective stress considerations. This sensitivity to hydraulic gradient was consistent with the recognized shear thinning of polymer hydrogels under higher shear stresses. Thus, the implications of using a hydraulic gradient in laboratory hydraulic conductivity testing that is different from that applied in the field should be considered, especially because hydraulic gradients in field applications often are lower than those used in laboratory hydraulic conductivity testing.
- The ASTM D6766 termination criteria are sufficient for generally, but insufficient for universally, measuring the hydraulic conductivity of the permeated EB-GCLs with lower percentages (5%–10%) of anionic polymer enhancements. These criteria do not consider the long-term impact of polymer elution from EB-GCLs comprising polymers. As a consequence, caution should be exercised when adapting these criteria to measure the hydraulic conductivity of EB-GCLs such as those evaluated in this study.

Data Availability Statement

All data, models, and code generated or used during the study appear in the published article.

Supplemental Materials

Figs. S1–S5 and Tables S1 and S2 are available online in the ASCE Library (www.ascelibrary.org).

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