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Hydraulic conductivity of bentonite-polymer geosynthetic clay liners to coal combustion product leachates

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ABSTRACT

Hydraulic conductivity of seven geosynthetic clay liners (GCLs) to synthetic coal combustion product (CCP) leachates were evaluated in this study. The leachates are chemically representative of typical and worst scenarios observed in CCP landfills. The ionic strength (I) of the synthetic CCP leachates ranged from 50 mM to 4676 mM (TCCP-50, LRMD-96, TFGDS-473, LR-2577, HI-3179 and HR-4676). One of the GCLs contained conventional sodium bentonite (Na-B) and the other six contained bentonite-polymer (B-P) mixture with polymer loadings ranging from 0.5% to 12.7%. Hydraulic conductivity tests were conducted at an effective confining stress of 20 kPa. The hydraulic conductivity of the Na-B GCLs were $>1 \times 10^{-10}$ m/s when permeated with all six CCP leachates, whereas the B-P GCLs with sufficient polymer loading maintained low hydraulic conductivity to synthetic CCP leachates. All the B-P GCLs showed low hydraulic conductivity ($<1 \times 10^{-10}$ m/s) to low ionic strength leachates (TCCP-50, I = 50 mM and LRMD-96, I = 96 mM). B-P GCLs with P > 5% showed low hydraulic conductivity ($<1 \times 10^{-10}$ m/s) up to HI-3179 leachates. These results suggest that B-P GCLs with sufficient polymer loading can be used to manage aggressive CCP leachates.

1. Introduction

Coal combustion products (CCPs), also known as coal ash/residues, are generated when coal is burnt to generate electricity or steam. CCPs include fly ash, bottom ash, flue gas desulfurization (FGD) materials (FGD solids, FGD Gypsum, Fixated FGD), spray dryer ash, etc. The production of CCPs has grown from 59.5 million Mg in 1974 to 114.7 million Mg in 2013 in the United States. In 2012, approximately 40 percent of the CCPs generated in U.S. were beneficially used, whereas the remaining 60 percent of CCPs were disposed in the surface impoundments and landfills. USEPA (United States Environmental Protection Agency) require the CCP disposal facilities to have a composite liner consisting of a minimum 0.75 mm geomembrane (GM) overlying a 0.6 m thick compacted clay liner (CCLs) to prevent the potential contamination of groundwater. Hydraulic conductivity of CCLs should be $\leq 1 \times 10^{-9}$ m/s [80 Fed. Reg. 21301 (October 14, 2015)]. The regulation allows the use of alternative materials (e.g., geosynthetic clay liners, GCLs) *in lieu* of CCLs to provide an equal or lower liquid flow rate than that of two-feet CCLs.

GCLs are easy to install, thin to save air space (5–10 mm), and have low hydraulic conductivity to water ($<1 \times 10^{-10}$ m/s) (Shackelford et al., 2000; Kolstad et al., 2004; Bradshaw et al., 2013; Scalia et al., 2014; Tian and Benson 2014; Tian et al., 2016, 2019; Setz et al., 2017; Bouazza et al., 2017; McWatters et al., 2019). GCLs have been widely used in landfill liner system, especially areas where clay is not available. Conventional sodium bentonite (Na-B) GCL consists of a thin layer of sodium bentonite sandwiched between two geotextiles (Jo et al., 2001, 2005; Rowe 2020). The sodium bentonite in GCLs is primarily composed of sodium montmorillonite (65–90%) (Shackelford et al., 2000; Lee et al., 2005; Bradshaw and Benson 2014). Sodium montmorillonite has high swelling capability in aqueous solutions (Grim 1968; Mitchell 1993; Shan and Daniel 1991; Rouf et al., 2016b; Acikel et al., 2018). The swelling of Na-B clogs the intergranular void space, resulting in narrower and tortuous flow paths for mobile water/leachate and low hydraulic conductivity ($\leq 1 \times 10^{-10}$ m/s) (Ashmawy et al., 2002; Kolstad et al., 2004; Guyonnet et al., 2005; Scalia et al., 2011; Athanassopoulos et al., 2015; Ören and Akar 2017; Chen et al., 2019). However, several researchers have reported that the aggressive leachates with high ionic

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strength, preponderance of multivalent cations, or/and extreme pH (<3 or >12) can inhibit the swelling of Na-B, resulting in high hydraulic conductivity ($>1 \times 10^{-10}$ m/s) (Katsumi et al., 2008; Razakamanatsoa et al., 2014; Di Emidio et al., 2015; Tian et al., 2016, 2019; Chen et al., 2019).

Therefore, the conventional Na-B GCLs have been modified using different organic molecules and polymers to improve their chemical compatibility against aggressive leachates (Onikata et al., 1999; Ashmawy et al., 2002; Katsumi et al., 2008; Özhan et al., 2018; Chai et al., 2020). These polymer modified GCLs, also known as bentonite-polymer (B-P) GCLs, have been found to have low hydraulic conductivity to aggressive leachates (Ashmawy et al., 2002; Katsumi et al., 2008; Razakamanatsoa et al., 2014; Di Emidio et al., 2015; Athanassopoulos et al., 2015; Tian and Benson, 2018; Chen et al., 2019; Zainab and Tian, 2020; Wireko and Abichou, 2021). Scalia et al. (2014) reported that a bentonite polymer composite (BPC) GCL can maintain low hydraulic conductivity ($<1 \times 10^{-11}$ m/s) even when permeated with aggressive inorganic solutions (e.g., 500 mM CaCl₂, 1 M NaOH, and 1 M HNO₃), whereas Na-B showed three orders of magnitude higher hydraulic conductivity than BPC. Tian et al. (2016) reported that B-P GCLs with sufficient polymer loading (e.g., >5%) can maintain low hydraulic conductivity to synthetic low-level radioactive waste (LLW) leachates (e.g., $\sim 1 \times 10^{-11}$ m/s). Tian et al. (2019) proposed the polymer clogging mechanism controls the low hydraulic conductivity of B-P GCL to aggressive leachates other than the swelling of Na-B. The polymer hydrogel absorb water and form three-dimensional structures, resulting in clogging of intergranular pores space. Chen et al. (2019) reported that hydraulic conductivity of conventional Na-B GCLs were $>1 \times 10^{-10}$ m/s with all the CCP leachates (I = 39.5 mM–755 mM), and the B-P GCLs with high polymer loading (>1.9%) can maintain low conductivity ($<1 \times 10^{-10}$ m/s) to CCP leachates with ionic strength ranging from 39.5 mM to 755 mM.

In this study, a new CCP leachate database was compiled through the analysis of leachate data collected from more than 130 coal-fired power plants. Many waste streams generate aggressive leachates with very high ionic strength (e.g., up to 4.6 M), which are much stronger than the leachates used in previous studies (e.g., up to 0.975 M) (Chen et al., 2018, 2019). The increase in aggressiveness of the CCP leachates may be because the gas emissions at coal-fired power plants are regulated more stringently (EPRI 2014). New emission control technologies that evolve to meet the new requirements alter the properties of CCPs, in particular of fly ash and flue gas desulfurization (FGD) gypsum. The leachability of CCPs can significantly increase using dry sorbent injection method to control the gas emission, as reported by Electric Power Research Institute (EPRI 2014). Another reason why CCP leachates are more concentrated may be due to new regulation from Environmental Protection Agency (EPA) on CCP disposal. The regulation requires to close all surface impoundments, by leaving all the CCPs in the surface impoundment and covering it with a cap or by excavating the CCPs from surface impoundments (wet storage) and re-disposing it into the lined CCP landfills. The landfill operation will try to minimize infiltration that pass through the CCP waste, which may potentially reduce the total amount of leachate but generate more aggressive leachates. Therefore, evaluating the hydraulic conductivity of B-P GCLs to current aggressive CCP leachates is necessary for the selection of GCLs as a liner material to construct new CCP disposal facilities. In addition, this study also extended the knowledge of hydraulic performance and chemical compatibility of B-P GCLs with wider ranging of polymer loading (0.5–12.7%) in compared with previous study (e.g., Chen et al., 2019).

The objective of this study was to evaluate the hydraulic conductivity of conventional Na-B and B-P GCLs to CCP leachates (I = 50–4676 mM). Tests were conducted on six B-P GCLs with various polymer loading ranging from 0.5 to 12.7% to evaluate the effect of polymer loading on the chemical compatibility of B-P GCLs. Six synthetic CCP leachates were created from data collected from 130 CCP disposal facilities to represent the typical and worst scenarios observed in the field. Control

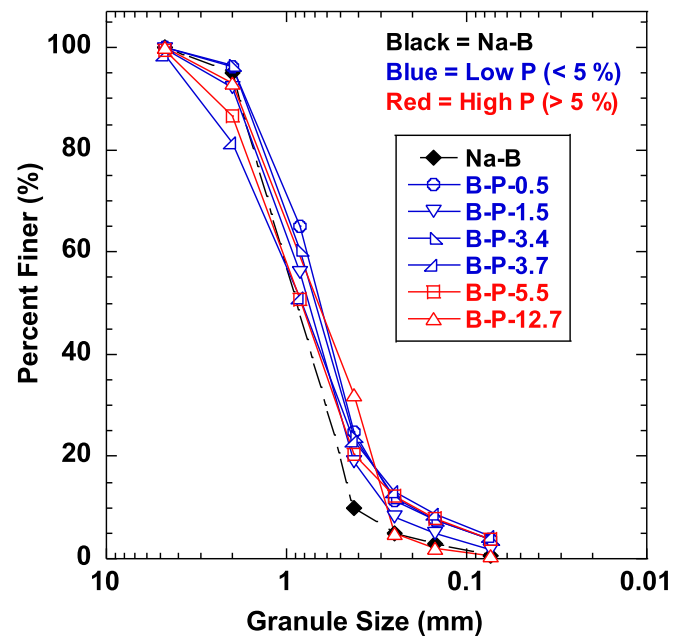


Fig. 1. Grain size distribution of sodium bentonite (Na-B) and bentonite polymer (B-P) GCLs used in this study. (Note: P = polymer loading.)

Table 1

Mass per unit area, initial thickness, and polymer loading of sodium bentonite (Na-B) and bentonite-polymer (B-P) GCLs used in this study.

GCL	Mass per unit area (kg/m ²)	Thickness (mm)	Polymer loading (%)
Na-B	4.0	5.0	–
B-P-0.5	5.7	7.0	0.5
B-P-1.5	6.8	8.9	1.5
B-P-3.4	6.8	7.0	3.4
B-P-3.7	6.1	7.9	3.7
B-P-5.5	6.0	8.1	5.5
B-P-12.7	4.8	7.0	12.7

Note: Polymer loading is calculated based on loss on ignition as per ASTM D7348 and Scalia et al., (2014). Mass per unit area is measured as per ASTM D5993. “–” indicates not applicable. The numeric digits with B-P GCLs represent their polymer loading.

tests were also conducted with deionized water (DI).

2. Materials and methods

2.1. Geosynthetic clay liners

Seven commercially available GCLs were evaluated in this study. One of the GCLs contained conventional Na-B and the other six contained B-P mixtures with varying polymer loading, ranging from 0.5% to 12.7%. The Na-B and B-P GCLs contain the same type of base bentonite. The B-P GCLs were named as B-P-0.5, B-P-1.5, B-P-3.4, B-P-3.7, B-P-5.5 and B-P-12.7 in this study. The numeric digits in the GCL designation indicates the polymer loading of the GCLs. The B-P GCLs contained a dry mixture of Na-B and proprietary polymer, except for B-P-12.7. The B-P-12.7 was created by polymerization of acrylic acid within bentonite slurry, as reported in Scalia et al. (2014).

The granule size distribution of the GCLs was determined by mechanical sieve analysis following ASTM D6913. All the GCLs had similar grain size distribution ($D_{50-avg.} = 0.78$ mm, medium to fine sand) as shown in Fig. 1. Mass per unit area, thickness and polymer loading of the GCLs are given in Table 1.

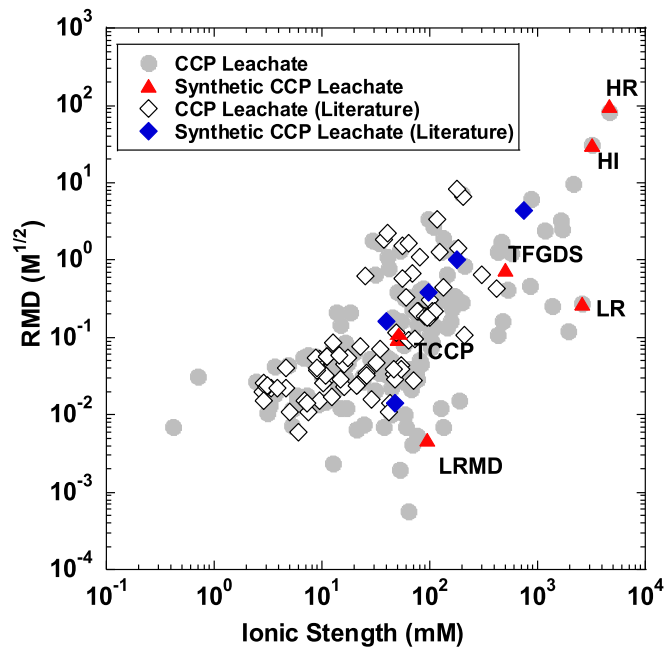


Fig. 2. RMD versus ionic strength of CCP leachates database collected in this study, along with the CCP leachate in literature. Synthetic CCP leachates used in this study are shown in red triangle, while the synthetic CCP leachates in literature are shown in blue diamond. Note: The maximum RMD observed in case of CCP leachate was ∞ (i.e. only monovalent cations present) and draw at RMD = 100 has been used. Data from literature was obtained from Chen et al. (2018, 2019).

2.2. Permeant solutions

Ionic strength (I), relative abundance of monovalent and polyvalent cations (RMD) and pH are the master variables known to affect the hydraulic properties of GCLs (Jo et al., 2001, 2005; Kolstad et al., 2004). Ionic strength is the measure of concentration of ions, defined by

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

where c_i = molar concentration of i th ion in solution; and z_i = valence of the i th ion. RMD is defined as follows (Kolstad et al., 2004):

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

where M_M = total molarity of monovalent cations; and M_D = total molarity of divalent cations.

Leachates with high ionic strength, preponderance of polyvalent cations (low RMD) and extreme pH (<3 or >12) can inhibit the swelling

of bentonite resulting in high hydraulic conductivity of the GCLs (Jo et al., 2001; Kolstad et al., 2004; Lee et al., 2005; Katsumi et al., 2008; Scalia et al., 2011; Tian et al., 2016; Chen et al., 2019). Tian et al. (2017), also related the sensitivity of the B-P GCL to anion ratio ($R = Cl^-/SO_4^{2-}$) and attributed it to the collapse of the polymer. When the Cl^- concentration increases, it opens the intergranular pores and results in higher hydraulic conductivity.

CCP leachate data collected from more than 130 CCP disposal units (landfills and impoundments) located within United States was chemically characterized based on dominant CCP type i.e., bottom ash, fly ash, flue gas desulfurization (FGD) materials (e.g., FGD gypsum, FGD solids, fixated FGD), mixed coal ash. The key variables that are known to affect the hydraulic conductivity of GCL include major cations (Na^+ , K^+ , Mg^{2+} , Ca^{2+}), major anions (Cl^- , SO_4^{2-}), ionic strength (I), pH, relative abundance of monovalent and divalent cations (RMD) and anion ratio (Cl^-/SO_4^{2-}) (Jo et al., 2001; Kolstad et al., 2004; Tian et al., 2017). Most of the CCP leachates were basic in nature with mean pH ranging from 7.4 to 9.3. Out of total samples collected, approximately 50% comprised of mixed coal ash. The CCP leachate data obtained from disposal sites primarily contained four major cations i.e., Na^+ (0.07 mM–3448.7 mM), K^+ (0.02 mM–1395.1 mM), Mg^{2+} (0.001 mM–507.5 mM) and Ca^{2+} (0.10 mM–538.1 mM) and two major anions Cl^- (0.05 mM–4366 mM) and SO_4^{2-} (0.01 mM–696.4 mM). The concentration of Al^{3+} ranged from 0.00026 to 4.36 mM. The CCP leachates produced at the disposal sites also contained high concentrations of heavy metals. The predominant heavy metals in the CCP leachate were As (0.67–3132 $\mu g/L$), Pb (0.07–490 $\mu g/L$), Hg (0–31 $\mu g/L$), Cr (0.16–4885 $\mu g/L$), Be (0.04–5 $\mu g/L$), Cd (0.10–237 $\mu g/L$) and Tl (0.05–36 $\mu g/L$). RMD ranges from a minimum value of 0.002 $M^{1/2}$ to a maximum of infinity (one leachate sample without divalent cations). Anion ratio (Cl^-/SO_4^{2-}) ranges from 0.01 to 122.30. And ionic strength ranged from 0.42 mM to 4685 mM. The comparison between the CCP leachate used by previous researchers (Chen et al., 2018, 2019; i.e., based on EPRI database) and the leachate data recently collected from more than 130 CCP disposal facilities located within United States is shown in Fig. 2.

Six synthetic CCP leachates were created from data collected from 130 CCP disposal facilities to represent the typical and worse scenarios observed in the field. The concentration of major cations, anions and the bulk properties (I, RMD, R and pH) of the six synthetic CCP leachates selected for this study are given in Table 2. Na^+ , K^+ and Mg^{2+} , Cl^- and SO_4^{2-} are the dominant ions present in the synthetic CCP leachates. I range from 50 mM to 4676 mM, RMD from 0.005 to infinite (∞ , no divalent cations), R from 0.01 (sulphate rich) to 122.29 (chloride rich). The synthetic CCP leachates used in this study were limited to the neutral conditions, ranging from 5.5 to 8.4. (1) Typical CCP (TCCP) leachate (I = 50 mM, RMD = 0.1 $M^{1/2}$, R = 0.25) represents the typical CCP leachate found in the field, obtained by taking geometric mean of all the CCP leachates. (2) Low RMD (LRMD) leachate (I = 96 mM, RMD = 0.005 $M^{1/2}$, R = 0.55) represents the leachate with highest concentration of divalent cations in comparison to monovalent cations and is

Table 2
Properties of synthetic CCP leachates.

Leachate	Major cations			Major anions		Bulk properties			
	Na^+	K^+	Mg^{2+}	Cl^-	SO_4^{2-}	I	RMD	R = Cl^-/SO_4^{2-}	pH
	(M)	(M)	(M)	(M)	(M)				
TCCP	0.01	0.00	0.01	0.003	0.01	50	0.10	0.25	6.8
LRMD	0.001	0.00	0.03	0.01	0.02	96	0.005	0.55	6.6
TFGDS	0.17	0.00	0.06	0.06	0.11	473	0.65	0.56	6.5
LR	0.19	0.01	0.57	0.01	0.67	2577	0.27	0.01	5.5
HI	2.23	0.03	0.005	0.47	0.90	3179	32.02	0.53	7.4
HR	3.30	1.34	0.00	4.56	0.04	4676	∞^a	122.29	8.4

Note: R = Anion ratio, I = Ionic strength, RMD = Relative abundance of monovalent and divalent cations, HR = High anion ratio, HI = High ionic strength, LR = Low anion ratio, TFGDS = Typical FGD solids, LRMD = Low RMD, and TCCP = Typical CCP leachate.

^a ∞ = No divalent cations present.

Table 3
Hydraulic conductivity of GCLs permeated with CCP leachates and DI water.

GCL	Permeant liquid	PVF	Total test time (days)	Termination criteria satisfied		Hydraulic conductivity (m/s)
				Hydraulic	Chemical	
Na-B	DI	12.8	110	Yes	No	2.2×10^{-11}
	TCCP-50	30.6	78	Yes	Yes	2.3×10^{-9}
	LRMD-96	7.3	12	Yes	Yes	4.9×10^{-9}
	TFGDS-473	5.7	5	Yes	Yes	1.7×10^{-7}
	LR-2577	4.3	5	Yes	Yes	3.1×10^{-7}
	HI-3179	11.6	17	Yes	Yes	7.9×10^{-7}
B-P-0.5	HR-4676	7.5	7	Yes	Yes	7.6×10^{-7}
	DI	5.6	184	Yes	No	2.2×10^{-11}
	TCCP-50	22.7	233	Yes	No	2.6×10^{-11}
	LRMD-96	89.9	512	Yes	Yes	2.7×10^{-11}
	TFGDS-473	11.8	33	Yes	Yes	1.4×10^{-7}
	LR-2577	16.3	89	Yes	Yes	4.4×10^{-8}
B-P-1.5	HI-3179	17.5	48	Yes	Yes	2.5×10^{-7}
	HR-4676	6.0	15	Yes	Yes	6.0×10^{-7}
	DI	10.8	395	Yes	No	7.8×10^{-12}
	TCCP-50	11.8	501	Yes	No	5.3×10^{-12}
	LRMD-96	47.7	550	Yes	Yes	1.2×10^{-11}
	TFGDS-473	25.2	142	Yes	Yes	6.0×10^{-8}
B-P-3.4	LR-2577	6.8	16	Yes	Yes	8.5×10^{-9}
	HI-3179	2.3	15	Yes	Yes	4.0×10^{-8}
	HR-4676	4.8	9	Yes	Yes	6.4×10^{-7}
	DI	6.5	155	Yes	No	5.1×10^{-12}
	TCCP-50	10.6	182	Yes	No	5.4×10^{-12}
	LRMD-96	7.7	183	Yes	No	5.0×10^{-12}
B-P-3.7	TFGDS-473	12.8	71	Yes	Yes	6.3×10^{-9}
	LR-2577	9.2	33	Yes	Yes	1.6×10^{-8}
	HI-3179	18.0	62	Yes	Yes	3.2×10^{-7}
	HR-4676	16.2	106	Yes	Yes	2.4×10^{-8}
	DI	3.0	192	Yes	No	2.4×10^{-12}
	TCCP-50	4.0	194	Yes	No	3.0×10^{-12}
B-P-5.5	LRMD-96	6.2	224	Yes	No	1.5×10^{-12}
	TFGDS-473	5.8	42	Yes	Yes	2.7×10^{-9}
	LR-2577	33.9	256	Yes	Yes	7.9×10^{-10}
	HI-3179	7.1	56	Yes	Yes	3.0×10^{-8}
	HR-4676	13.7	71	Yes	Yes	9.5×10^{-8}
	DI	15.8	453	Yes	No	9.4×10^{-12}
B-P-12.7	TCCP-50	13.9	500	Yes	No	6.3×10^{-12}
	LRMD-96	9.4	490	Yes	No	4.5×10^{-12}
	TFGDS-473	27.4	431	Yes	Yes	1.4×10^{-11}
	LR-2577	34.3	380	Yes	Yes	3.7×10^{-11}
	HI-3179	3.8	5	Yes	Yes	4.7×10^{-8}
	HR-4676	2.2	4	Yes	Yes	7.6×10^{-8}
B-P-12.7	DI	1.9	37	Yes	No	6.7×10^{-12}
	TCCP-50	3.6	146	Yes	No	2.4×10^{-12}
	LRMD-96	11.8	457	Yes	No	5.0×10^{-12}
	TFGDS-473	7.6	174	Yes	Yes	3.2×10^{-12}
	LR-2577	6.2	100	Yes	Yes	2.9×10^{-11}
	HI-3179	37.8	388	Yes	Yes	8.6×10^{-12}
	HR-4676	10.1	10	Yes	Yes	5.0×10^{-7}

Note: PVF = Pore volumes of flow. The numeric digits with CCP leachates represent their ionic strength in mM.

obtained by taking geometric mean of all the leachates collected from FGD gypsum disposal facilities. (3) Typical FGD solids (TFGDS) leachate ($I = 473$ mM, $RMD = 0.65 M^{1/2}$, $R = 0.56$) represents the most aggressive CCP type leachate obtained by taking geometric mean of all the leachates collected from FGD solids disposal facilities. (4) Low anion ratio (LR) leachate ($I = 2577$ mM, $RMD = 0.27 M^{1/2}$, $R = 0.01$) represents the leachate with minimum anion ratio i.e., with the highest concentration of sulphate ions. (5) High ionic strength (HI) leachate ($I = 3179$ mM, $RMD = 32.0 M^{1/2}$, $R = 0.53$) represents the leachate with high ionic strength but relatively less aggressive RMD and R. (6) High anion ratio (HR) leachate ($I = 4676$ mM, $RMD = \infty$, $R = 122.3$) represents the most aggressive leachate with highest ionic strength and highest concentration of chloride ions. Fig. 2 shows that the synthetic CCP leachates selected for this study are much more aggressive than the synthetic leachate used by previous researchers both in terms of high ionic strength (e.g., $I = 4.6$ M versus 0.975 M) and low RMD (e.g., 0.005 versus $0.014 M^{1/2}$).

All the synthetic CCP leachates were prepared by dissolving reagent-

grade NaCl, KCl, Na_2SO_4 , $MgCl_2$, $MgSO_4$ and K_2SO_4 in Type II DI water as per ASTM D1193. The synthetic leachates were stored in sealed carboys to prevent interaction with atmosphere (CO_2). The electrical conductivity (EC) and pH of the synthetic leachates were checked periodically to assure no changes in the leachate chemistry.

2.3. Hydraulic conductivity test

Hydraulic conductivity tests were conducted on 152.4 mm (6-inch) circular GCL specimens in flexible wall permeameters using the falling headwater and constant tailwater method in accordance with ASTM D6766 (2012). All the GCL specimens were first hydrated in the permeameter with the specific leachate at an effective confining stress of 20 kPa for 48 h with hydraulic head of 1.30 m. After hydration, tests were continued at an effective stress of 20 kPa and at an average hydraulic gradient of approximately 150. No backpressure was applied to mimic field condition. 50 mL graduated glass burettes were used as influent reservoir. Effluent was collected in 70 mL polyethylene bottles and its

pH and EC were periodically measured to check chemical equilibrium. The tests were continued until the hydraulic and chemical equilibrium was achieved in accordance with ASTM D6766 (2012). Tests were conducted on seven GCLs (one Na-B GCL and six B-P GCLs) using six synthetic CCP leachates. Controls tests were conducted using DI water.

All hydraulic conductivity tests were conducted until reaching hydraulic equilibrium and chemical equilibrium. Hydraulic equilibrium criteria require that three consecutive hydraulic conductivity values are within 25% of the mean and without temporal trend, and the ratio of inflow over outflow (Q_{out}/Q_{in}) is within 0.75 and 1.25. The chemical equilibrium criteria require that the electric conductivity (EC) of the effluent (EC_{out}) is falling within 10% of influent value (EC_{in}), and the ratio of the pH of effluent (pH_{out}) and influent (pH_{in}) are within 10% (Tian et al., 2016; Chen et al., 2019).

2.4. Swell index

Swell index of B-P GCLs and Na-B GCL was measured using DI water and the six synthetic CCP leachates following the test method in accordance with ASTM D5890. The specimens were first crushed using a pastel and mortar such that 100% passes #100 sieve and at least 65% passes #200 sieve. The tests were conducted using 2 g of oven dried specimens, added into a 100 mL graduated cylinder filled with 90 mL of solution (DI water and six synthetic CCP leachates). The GCL specimen was added into the graduated cylinder with an increment of 0.1 g. After adding 2 g of specimen, the cylinder was filled to 100 mL with the solution (DI water and six synthetic CCP leachates). The SI was measured as the volume of the swollen specimen in the graduated cylinder after 24 h (in mL/2 g).

2.5. Polymer quantification

Loss on ignition (LOI) tests per ASTM D7348 was used as an indicator of polymer loading in B-P GCL (Scalia et al., 2014; Tian et al., 2016; Gustitus et al., 2021). The Na-B and B-P composites were extracted from GCLs, grounded to pass a No. 20 sieve (ASTM E11; ASTM 2013) and dried for 24 h in the oven at 105 °C to remove the moisture in test specimens. Approximately 3 g of oven-dry B-P composite was used for each LOI test. Oven-dried specimens were ignited at 550 °C for 4 h. After 4 h, the material was removed from the furnace to determine the LOI of Na-B and B-P. The LOI test should also conduct with pure polymer additives used in the B-P GCLs to determine the burning rate. Then, the initial polymer loading in B-P GCL can be calculated by discounting the LOI of pure Na-B (e.g., 1.6%) from the LOI of B-P composite and considering burning rate of polymer additives, as proposed in Gustitus et al. (2021).

The polymer added in B-P-12.7 GCL is the same as the B-P GCL used in Scalia et al. (2014), which has a burning rate of 74.7% (loss of mass). The polymer loading in B-P-12.7 GCL was calculated by discounting the LOI of pure Na-B and considering incomplete burning of polymer additive. However, the polymers in B-P-0.5, B-P-1.5, B-P-3.4, B-P-3.7, and B-P-5.5 GCLs are proprietary, so the LOI of pure polymer additives were unavailable. Polymer loadings in these GCLs were calculated based on the assumption that polymer only contained organic compounds and can be fully combusted during LOI test. The polymer loading in B-P GCLs by discounting the LOI Na-B GCL (e.g., 1.6%). This approach may underestimate the polymer loading if the polymer is not fully degraded during the LOI test.

The residual polymer loading in B-P GCLs after permeation were also determined using LOI test via ASTM D7348. Similar procedures were applied as the determination of initial polymer loading. The Na-B and B-P composites were extracted from three spots on the circular GCL samples after permeation. The LOI of Na-B GCL that permeated with different CCP leachates were carefully measured, because the LOI of Na-B after permeation may be different than that of original Na-B due to the cation exchange process. Increasing mole fraction of Ca^{2+} in

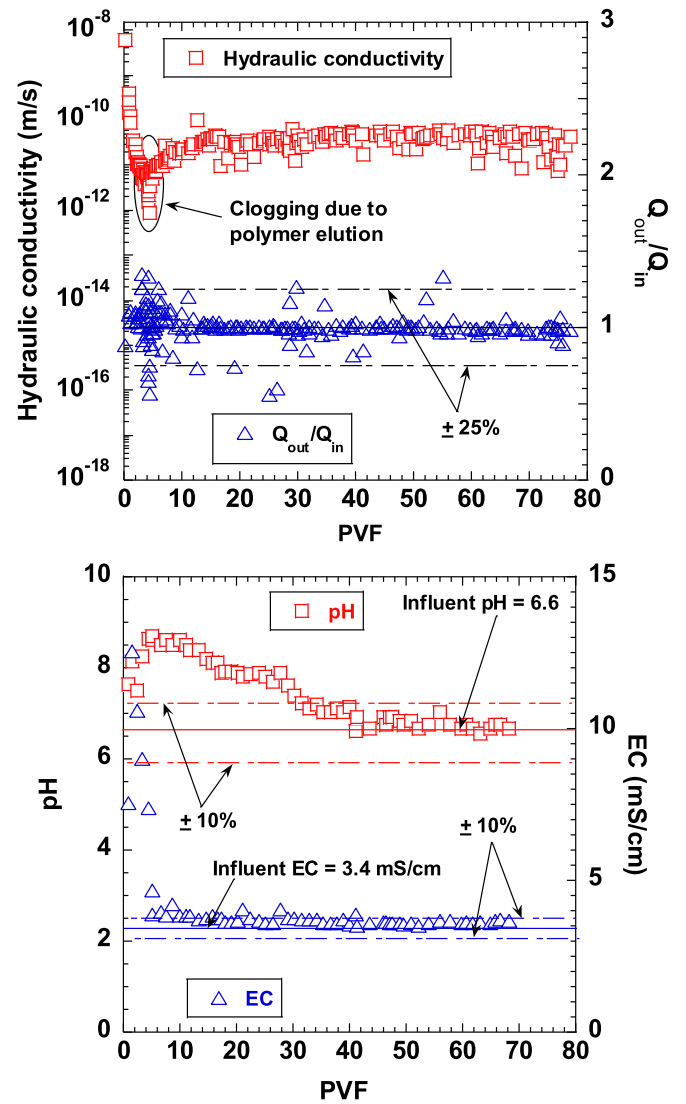


Fig. 3. Hydraulic conductivity, ratio of outflow to inflow (Q_{out}/Q_{in}), pH, and EC from test on B-P-0.5 permeated with LRMD-96 leachate.

bentonite results in increase of LOI (Gustitus et al., 2021). The residual polymer loading of B-P GCL samples were calculated by discounting the LOI of Na-B GCL samples that permeated with same leachates.

2.6. Total organic carbon analysis

The mass of polymer eluted from the B-P GCLs during permeation was quantified by the total organic carbon (TOC) concentration (mg/L) in the effluents measured using a Shimadzu TOC-VWS analyzer, following the APHA method (Rice et al., 2012). Calibration standard solutions were prepared by diluting 1000 mg/L anhydrous potassium hydrogen phthalate to various concentrations with DI water. Prior to analysis, samples were diluted 25 times to keep the carbon content within the range of the standard solutions. After dilution, 40 mL of each sample was placed into the glass vials for analysis. Samples were injected into analyzer and acidified to a pH of 2–3 with phosphoric acid. Inorganic carbon component of the sample was removed by sparging of a gas free of carbon dioxide (CO_2) through sample. Remaining total carbon content was oxidized to CO_2 by oxidizing reagent (containing sodium persulfate and phosphoric acid). After the gas was cooled, dehydrated, and passed through halogen scrubber. Finally, the CO_2 gas was detected in a non-dispersive infrared (NDIR) gas analyzer.

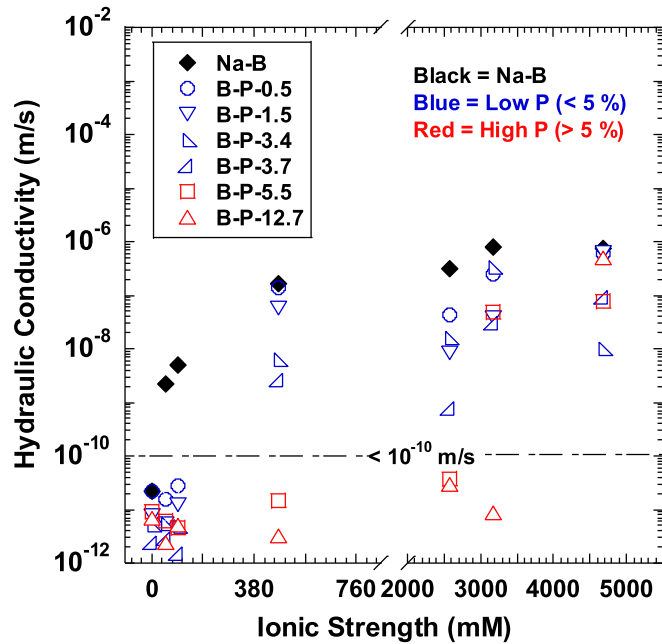


Fig. 4. Hydraulic conductivity of Na-B and B-P GCLs to synthetic CCP leachates as a function of ionic strength.

3. RESULTS and DISCUSSIONS

A summary of the hydraulic conductivity tests performed on the Na-B and B-P GCLs using the synthetic CCP leachates is shown in Table 3. Test durations were up to 500 days. All Na-B GCL specimens permeated with the synthetic leachates achieved both hydraulic and chemical equilibrium. Hydraulic conductivity of all the B-P GCLs permeated with TFGDS-473, LR-2577, HI-3179, and HR-4676 met both the hydraulic and chemical equilibrium termination criteria, whereas all the B-P GCLs permeated with TCCP-50, and LRMD-96 reached hydraulic equilibrium but not chemical equilibrium due to low hydraulic conductivity and limited pore volume of flows (PVFs). Tests of B-P GCLs to TCCP-50 and LRMD-96 are still ongoing to evaluate their long-term hydraulic conductivity.

3.1. Hydraulic conductivity of B-P-0.5 GCL to LRMD-96 leachate

Hydraulic conductivity of the B-P-0.5 permeated with LRMD-96 leachate as a function of PVF is shown in Fig. 3. Hydraulic conductivity of the B-P-0.5 to LRMD-96 decreased dramatically between 4 and 6 PVF. This decrease in hydraulic conductivity can be attributed to clogging of the influent and effluent tubes by eluted polymer. Tian et al. (2019) had similar observations that polymer eluted from B-P GCLs during permeation. To prevent clogging, the influent and effluent tubes were regularly flushed using the permeant solutions (Wireko and Abichou, 2021). Hydraulic conductivity of the B-P-0.5 to LRMD-96 however, increased from 10^{-12} m/s to 10^{-11} m/s between 6 and 14 PVF and remained constant afterward. At approximately 6 PVF the EC of the effluent levelled off and fall within the range associated with chemical equilibrium based on EC termination criteria as per ASTM D6766 (i.e., effluent EC within 10% of the influent EC). The test reached chemical equilibrium around 40 PVF (i.e., EC and pH of the effluent levelled off and fell within the $1 \pm 10\%$ of the EC and pH in the influent).

3.2. Effect of ionic strength on hydraulic conductivity

Hydraulic conductivity of the GCLs permeated with DI water and the synthetic CCP leachates is shown in Fig. 4 as a function of ionic strength of the permeant solutions. Hydraulic conductivity of the Na-B GCL was

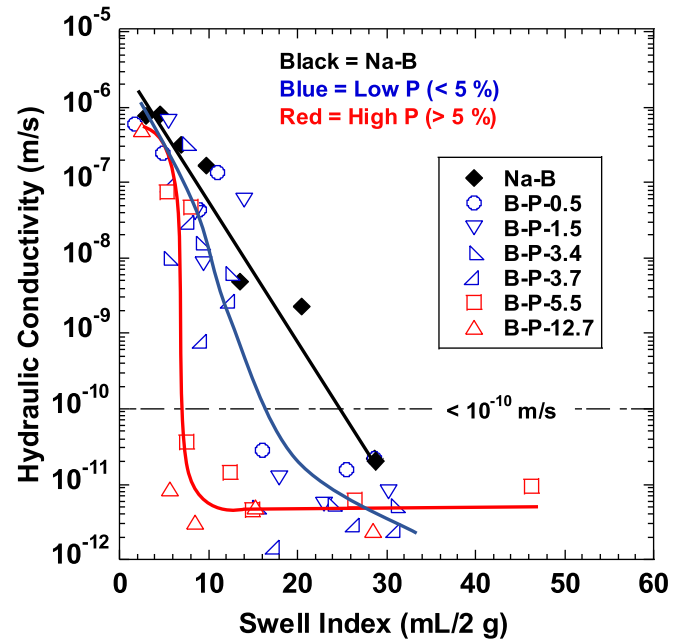


Fig. 5. Hydraulic conductivity of Na-B and B-P GCLs to synthetic CCP leachates and DI water versus swell index.

low (2.2×10^{-11} m/s) when permeated with DI water, whereas the hydraulic conductivity of Na-B GCL to all the CCP leachates used in this study ($I = 50$ mM–4676 mM) were greater than 1×10^{-10} m/s. Hydraulic conductivity of the B-P GCLs to the CCP leachates varied with different polymer loading and ionic strength. All six B-P GCLs had low hydraulic conductivity ($< 1 \times 10^{-10}$ m/s) when permeated with the TCCP-50 leachate (ranging from 2.4×10^{-12} m/s to 2.6×10^{-11} m/s) and LRMD-96 (ranging from 1.5×10^{-12} m/s to 2.7×10^{-11} m/s) leachate.

Hydraulic conductivity of the B-P GCLs varied with polymer loading when the ionic strength of CCP leachates increased to 473 mM–4676 mM. Hydraulic conductivity of B-P GCLs with polymer loading less than 5% (i.e., B-P-0.5, B-P-1.5, B-P-3.4, and B-P-3.7) were $> 1 \times 10^{-10}$ m/s when permeated with TFGDS-473, LR-2577, HI-3179 and HR-4676, whereas B-P-5.5 maintained low hydraulic conductivity when permeated with CCP leachates with ionic strength up to 2577 mM, and hydraulic conductivity of B-P-12.7 was in the range of 10^{-12} m/s to all the CCP leachates ($I = 50$ mM–3179 mM), except HR-4676.

3.3. Swell index

The relationship between hydraulic conductivity of the GCLs and swell index is shown in Fig. 5. The hydraulic conductivity of Na-B GCL decreased significantly with the increase in swell index. The swell index of Na-B in HR-4676 was 2.0 mL/2 g, which yielded a high hydraulic conductivity (7.6×10^{-7} m/s). The hydraulic conductivity of Na-B GCL decreased to 2.2×10^{-11} m/s ($< 1 \times 10^{-10}$ m/s) at a swell index of 28.7 mL/2 g when permeated with DI water. The hydraulic conductivity of Na-B GCLs has a strong inverse relationship with swell index since bentonite swelling is primarily responsible for the hydraulic conductivity of Na-B GCLs (Jo et al., 2001; Kolstad et al., 2004; Lee et al., 2005; Katsumi et al., 2008; Scalia et al., 2014; Tian et al., 2016; Chen et al., 2018).

In comparison with Na-B, the hydraulic conductivity of the B-P GCLs with polymer loading lower than 5% (B-P-0.5, B-P-1.5, B-P-3.4, B-P-3.7) decreased faster as the swell index increased. For example, the hydraulic conductivity of the B-P GCLs (with polymer loading $< 5\%$) were lower than 1×10^{-10} m/s with swell index > 15.5 mL/2 g, whereas hydraulic conductivity of the Na-B GCL was $> 1 \times 10^{-10}$ m/s with swell

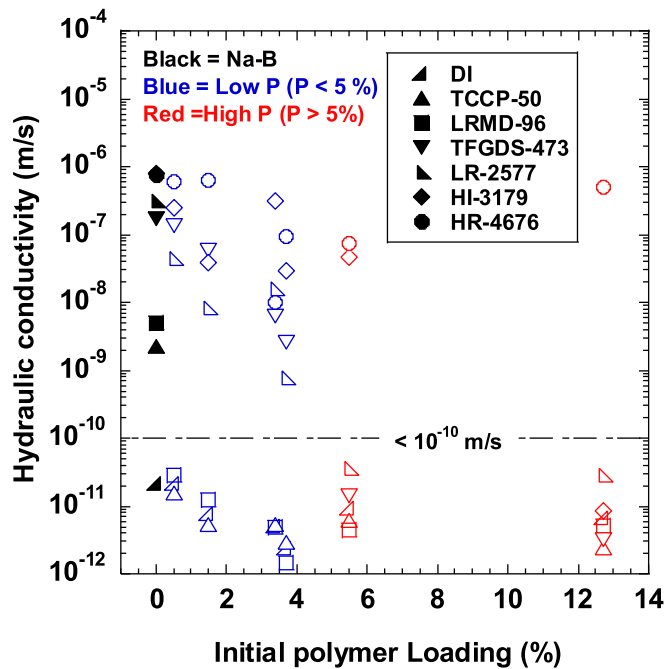


Fig. 6. Hydraulic conductivity of Na-B and B-P GCLs permeated with synthetic CCP leachates as a function of polymer loading. (Note: polymer loadings were determined by LOI test and Na-B GCL was plot at zero polymer loading.)

index of 20.5 mL/2 g. These observations indicate that both swelling of Na-B and polymer clogging mechanism contribute towards the low hydraulic conductivity of B-P GCLs with low polymer loading (<5%), as proposed in Tian et al. (2019).

The hydraulic conductivity of B-P GCLs with polymer loading higher than 5% showed decoupling trend with swell index, i.e., both low ($<1 \times 10^{-10}$ m/s) and high ($>1 \times 10^{-10}$ m/s) hydraulic conductivities were observed at a similar SI (≈ 6 mL/2 g), suggesting that hydraulic conductivity of B-P GCLs with high polymer loading is primarily controlled by polymer clogging rather the Na-B swelling (Tian et al., 2019). Similar findings were observed by previous researchers. Scalia et al. (2014)

observed that B-P GCL when permeated with 500 mM CaCl_2 solutions reported low hydraulic conductivity ($<1 \times 10^{-10}$ m/s) even with a swell index of 7 mL/2 g.

3.4. Initial polymer loading

The hydraulic conductivity of the B-P GCLs to CCP leachates is shown in Fig. 6 as a function of polymer loading. Na-B is also shown in Fig. 6 with zero polymer loading (e.g., black symbols). Blue symbols represent B-P GCLs with polymer loading less than 5% and red symbols for B-P GCLs with polymer loading higher than 5%. Na-B GCL could not maintain low hydraulic conductivity to all synthetic leachates. B-P GCLs with low polymer loading (e.g., <5%) were chemically compatible with TCCP-50 and LRMD-96, but not synthetic leachates with an ionic strength ≥ 470 mM. In contrast, B-P GCLs with higher polymer loading (>5%) maintained low hydraulic conductivity to most of the synthetic CCP leachates. For example, the hydraulic conductivity of B-P-5.5 to TCCP-50, LRMD-96, TFGDS-473, and LR-2577 was comparable to the hydraulic conductivity to DI water. Also, B-P-12.7 had low hydraulic conductivity to HI-3179 (with $I = 3179$ mM).

The lower hydraulic conductivity ($<1 \times 10^{-10}$ m/s) of B-P GCLs to CCP leachates compared with Na-B GCL can be attributed to polymer clogging mechanism, as proposed in Tian et al. (2016). B-P GCLs contained super absorbent polymers, such as Na-carboxymethylcellulose, Na-polyacrylate and Na-polyacrylamide, that can absorb high amount of water and form three dimensional structures (Scalia et al., 2014; Di Emidio et al., 2015; Tian et al., 2019). The water molecules attached to the polymer hydrogels hydraulically act as solid and immobile water, which are similar to water molecules attached to bentonite during osmotic swell (Tian et al., 2016, 2019). The polymer hydrogel can clog the intergranular void spaces of bentonite, resulting in narrower and tortuous flow paths and low hydraulic conductivity. However, leachates with high ionic strength can inhibit the water absorption capacity and swelling of polymer hydrogels (Tian et al., 2019), which limit capability of polymer hydrogel to clog intergranular pore space of bentonite. More polymer hydrogel is required to maintain the low hydraulic conductivity of B-P GCL when permeated with aggressive leachates. Therefore, B-P GCLs with higher polymer loading have better chemical compatibility than B-P GCLs with low polymer loading (Tian et al., 2016, 2019; Chen et al., 2019).

Type of Polymer

Linear-polymer



Crosslinked-polymer

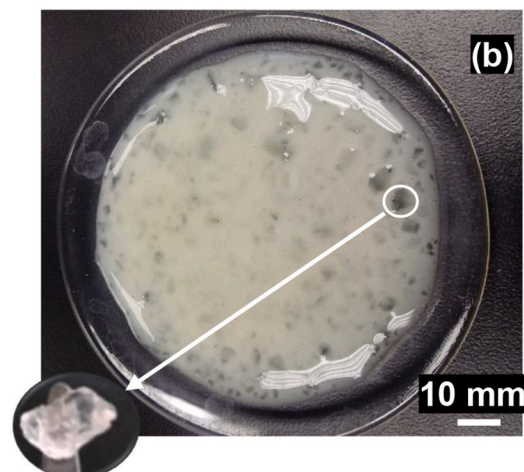


Fig. 7. Images of two different types of polymers found in B-P GCLs: (a) B-P-3.7 and (b) B-P-3.4.

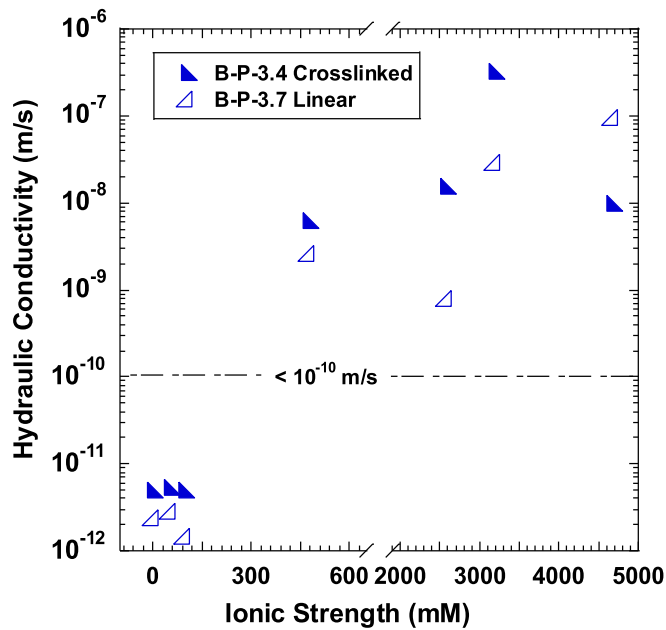


Fig. 8. Hydraulic conductivity of B-P-3.4 and B-P-3.7 GCLs to CCP leachates.

3.5. Polymer type

Visual examination of the B-P specimens after the swell index test showed that the B-P GCLs used in this study may contain two different types of polymers. B-P-0.5, B-P-1.5 and B-P-3.7 formed a viscous gel after hydration in leachates as shown in Fig. 7a, whereas B-P-3.4, B-P-5.5 and B-P-12.7 formed small granules of non-viscous hydrogels after hydration, which are likely to be crosslinked polymer (Fig. 7b). Chimmampam et al. (2011) reported that linear polymers result in viscous solutions after hydration and the viscosity of these solutions can vary depending on molar mass and charge density of polymer and the quality of hydrating medium. Ahmed (2015) reported that crosslinked hydrogels can swell and retain a significant amount of water without dissolving in water.

Comparison between B-P GCLs with different polymer type (B-P-3.4, crosslinked and B-P-3.7, linear) but similar polymer loading is shown in Fig. 8. The two GCLs showed similar trends of low ($< 1 \times 10^{-10}$ m/s) or high ($> 1 \times 10^{-10}$ m/s) hydraulic conductivity to the same CCP leachates, which suggests that polymer type may not have a significant impact on the hydraulic conductivity of B-P GCLs in comparison to polymer loading, as seen in Fig. 6.

These different types of polymer may affect the mechanism of controlling the hydraulic conductivity of B-P GCLs. The linear polymers forming a viscous glue-like substance may help to keep the bentonite particles together, whereas the crosslinked polymer forming small three-dimensional hydrogels help to clog the void spaces present between the bentonite particles resulting in low hydraulic conductivity of the B-P GCLs. Further studies are needed to evaluate the effect and behavior of these two different types of polymers.

3.6. Polymer elution

The temporal trend of hydraulic conductivity of B-P-0.5 to LR-2577 leachate ($I = 2577$ mM) is shown in Fig. 9a. The hydraulic conductivity of the B-P GCL decreased to 8.9×10^{-10} m/s and remained constant for approximately 10 PVF but after 10 PVF the hydraulic conductivity of the GCL specimen increased drastically to 8.2×10^{-8} m/s. Dye test was conducted on the B-P-0.5 GCL specimen to determine the possibility of side wall leakage or preferential flow. The result showed a spot with concentrated pink color (Fig. 9b), indicating the faster flow pathways for

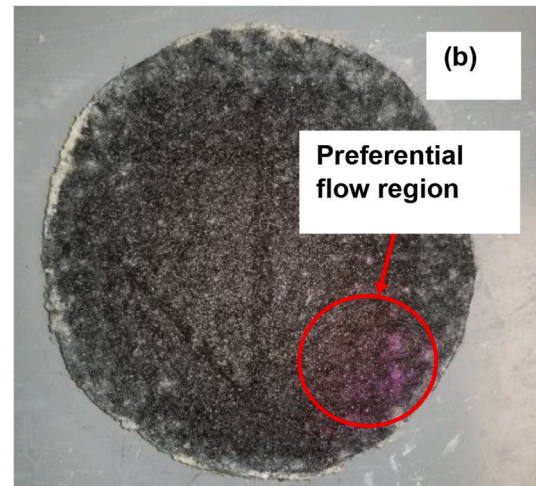
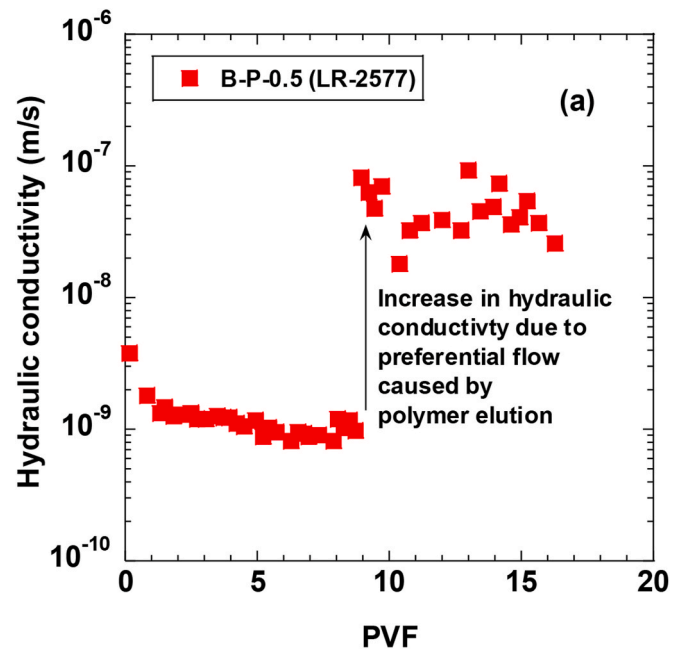


Fig. 9. (a) Temporal behavior of B-P-0.5 GCL permeated with LR-2577 leachate (b) Dye test conducted on B-P-0.5 GCL to LR-2577 leachate indicating preferential flow on the effluent side.

synthetic leachate of B-P-0.5 GCL specimen. The increase in hydraulic conductivity of B-P GCLs can be attributed to polymer elution during permeation, leading to open flow channels and high hydraulic conductivity (Tian et al., 2019).

The polymer elution was estimated by measuring the TOC in the effluent. TOC concentration in the effluent is shown in Fig. 10 as a function of PVF for the B-P-5.5 and B-P-12.7 GCLs permeated with LRMD. The TOC was highest within a few PVF after permeation began and gradually decreased as permeation continued, illustrating that polymer was eluted out from the B-P GCLs during permeation. For examples, the TOC in the effluent of B-P-12.7 was approximately 3100 mg/L at 1.6 PVF and decreased to 5.3 mg/L at 12 PVF. TOC in the effluent were higher for the B-P-12.7 GCL than the B-P-5.5 GCL, indicating greater elution for the B-P GCL with higher polymer loading.

Loss on ignition (LOI) tests were conducted to quantify the polymer content of B-P GCL after permeation. The comparison of initial and residual polymer loading of B-P-0.5, B-P-1.5, B-P-3.4, B-P-3.7, and B-P-5.5 GCLs to TFGDS-473, LR-2577, and HI-3179 leachates are shown in Fig. 11. The initial polymer loading of all five B-P GCLs were higher

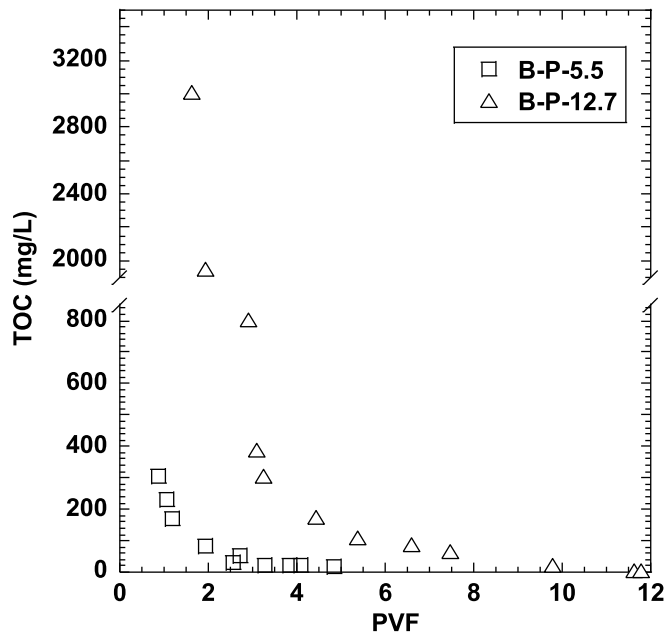


Fig. 10. TOC in the effluent of B-P-5.5 and B-P-12.7 GCLs during the permeation with LRMD-96 leachate as a function of PVF.

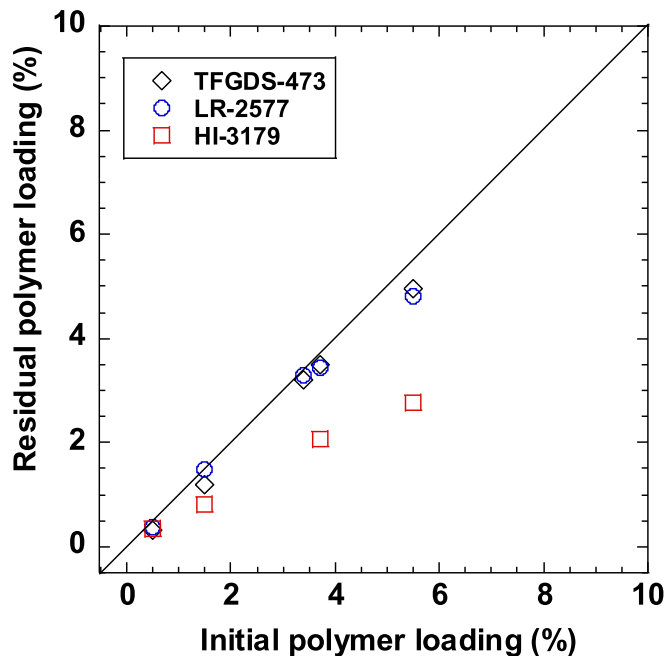


Fig. 11. Initial polymer loading of B-P GCLs versus residual polymer loading of B-P GCLs after hydraulic conductivity test. (Note: polymer loadings were determined by LOI test).

than the residual polymer loading, illustrating that polymer was eluted from B-P GCL during permeation. In addition, the polymer elution increased as the permeant solution become more aggressive. The polymer loading of B-P-3.7 GCL decreased from 3.7% to 3.5% after permeated with TFGDS, whereas polymer loading of B-P-3.7 GCL decreased to 2.1% to HI-3179 leachate. B-P GCLs permeated with HI-3179 leachate eluted more, ranging from 30% to 50% of the initial polymer content, whereas only a small fraction (1%–26%) of polymer eluted from B-P GCLs permeated with LR-2577 leachate. Similar observation has been reported in Tian et al. (2019) that B-P GCL

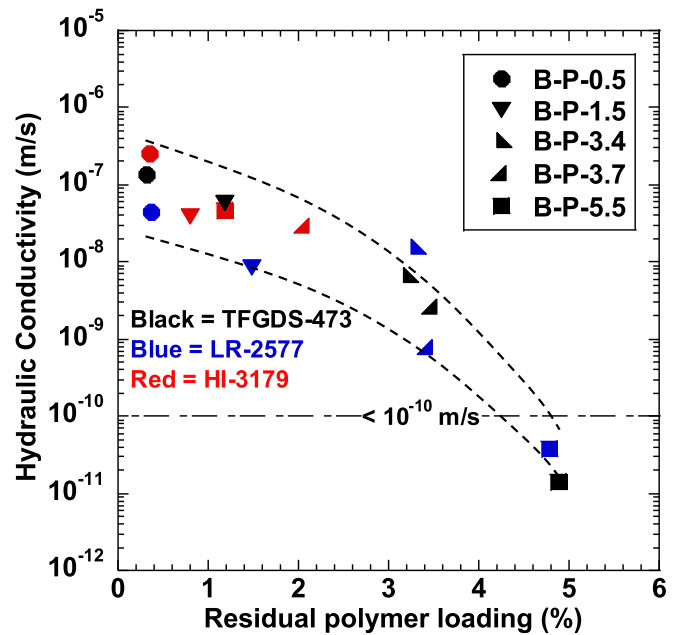


Fig. 12. Hydraulic conductivity of B-P GCLs to synthetic CCP leachates as a function of residual polymer loading after permeation. (Note: polymer loadings were determined by LOI test).

permeated with aggressive leachate results more polymer elution.

The hydraulic conductivity of B-P GCLs permeated with TFGDS-473, LR-2577, and HI-3179 leachates were shown in Fig. 12 as a function of residual polymer loading after permeation. The test results illustrated that hydraulic conductivity of the B-P GCLs to synthetic CCP leachates corresponded to the residual polymer loading after permeation, i.e., B-P GCL with high residual polymer loading maintained low hydraulic conductivity to synthetic CCP leachates, whereas hydraulic conductivity of B-P GCLs with low residual polymer loading were higher than 1×10^{-10} m/s. B-P-5.5 had high polymer loading initially, however, as the polymer eluted during permeation with HI leachate, the B-P-5.5 GCL yielded to a high hydraulic conductivity (4.7×10^{-8} m/s). In contrast, the B-P-5.5 GCLs with less polymer elution when permeated with TFGDS-473 and LR-2577 had low hydraulic conductivity. These results indicate that preventing polymer elution is of particular importance to maintain low hydraulic conductivity of B-P GCLs to CCP leachates.

4. Summary and conclusions

Hydraulic conductivity of Na-B and B-P GCLs to six synthetic CCP leachates ($I = 50$ mM–4676 mM) was evaluated in this study. The B-P GCLs used in this study contained a dry mixture of sodium bentonite and proprietary polymer with polymer loading ranging from 0.5% to 12.7%. Hydraulic conductivity tests were conducted at an effective stress of 20 kPa and were continued until reaching the hydraulic and chemical equilibrium as per ASTM D6766. Tests were also conducted with DI water as control. Based on the findings from these tests, the following conclusions and recommendations are drawn:

- Na-B GCL had high hydraulic conductivity to all six synthetic CCP leachates with an ionic strength ranging from $I = 50$ mM–4676 mM, illustrating that Na-B GCL may not be adequate to manage CCP leachates.
- B-P GCLs with polymer loading $< 5\%$ had low hydraulic conductivity ($< 1 \times 10^{-10}$ m/s) when permeated with the TCCP-50 leachate ($I = 50$) and LRMD-96 leachate ($I = 50$), but had high hydraulic conductivity ($> 1 \times 10^{-10}$ m/s) when permeated with TFGDS-473, LR-2577, HI-3179 and HR-4676. On the contrary, B-P GCLs with

polymer loading 5.5% can maintain low hydraulic conductivity (3.7×10^{-11} m/s) to LR-2577, but not to HI-3175 and HR-4676 leachates, whereas the B-P-12.7 GCLs can maintain low hydraulic conductivity to HI-3175.

- The test results show that B-P GCLs with higher polymer loading have better chemical compatibility. B-P GCL with sufficient polymer loading (>5%) can be used to manage aggressive leachate. Leachate specific tests are needed to evaluate the long-term hydraulic conductivity of B-P GCL.
- Hydraulic conductivity of B-P GCLs can be affected by polymer elution, as polymer elution may open flow channels for preferential flow paths, resulting in increasing of hydraulic conductivity. Further study is needed to evaluate the long-term performance of B-P GCL to CCP leachates to ensure the durability of B-P GCLs.
- Test result indicated that effect of polymer loading on hydraulic conductivity is more significant than the type of polymer hydrogel. Further study is needed to investigate.

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