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Advances in Solute Diffusion through Bentonite Polymer Composites

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ABSTRACT

Geosynthetic clay liners (GCLs) comprising bentonite-polymer composite (BPC) have been developed for use in containment applications where sodium bentonite (NaB) is likely to undergo chemical interactions that degrade barrier performance (e.g., an increase in hydraulic conductivity, *k*, for solutions of high ionic strength and/or extreme pH). Modelling the performance of a barrier system within the hydrogeological setting requires knowledge of both the *k* and the diffusion coefficients of the GCL. Although diffusion coefficients for NaB-GCLs for inorganic and organic contaminants have been well studied, understanding of the impact of polymer amendment on diffusion coefficients of BPC-GCLs remains limited. This paper provides a synthesis of data in the literature as well as ongoing studies focused on diffusion of inorganic contaminants through BPC. Based on experimental results to date, BPC may exhibit significantly lower *k* than NaB (e.g., by orders of magnitude) for a given solution, without also exhibiting lower diffusion coefficients than NaB. In fact, for some cases, diffusion coefficients of BPC were double to triple those of the NaB, whereas for other BPC-leachate combinations the opposite was true. Relative to NaB, the diffusion coefficients of BPCs are more variable and complex, due in part to how contaminants diffuse through and interact with the polymer hydrogel versus the bentonite fraction. Thus, reliable estimation of diffusion coefficient values of BPC-GCLs is more challenging than for NaB-GCLs and may require case-specific testing. Data for diffusion coefficients of BPC-GCLs for a range of leachates, BPC types, and test conditions (e.g., hydration, stress) is needed to allow for accurate transport analysis of barrier systems that utilize BPC-GCLs. In this regard, recent advances and future research directions also are identified within the paper.

Keywords: bentonite polymer composite, diffusion, geosynthetic clay liner

1 INTRODUCTION AND BACKGROUND

1.1 Bentonite Polymer Composites (BPC)

Sodium bentonite (NaB) is a naturally occurring clay commonly used in engineered barrier systems, such as geosynthetic clay liners (GCLs). NaB exhibits low hydraulic conductivity (*k*), high swell, and low diffusion coefficients when exposed to dilute aqueous solutions (Malusis et al., 2003). However, in some applications NaB may undergo chemical interactions that degrade barrier performance (e.g., when exposed to solutions with high ionic strength, multivalent cations, and/or extreme pH) resulting in increased *k*, reduced swell, and increased diffusion coefficients (e.g., Egloffstein, 2002; Jo et al., 2005; Petrov and Rowe, 1997; Shackelford et al., 2000; Lake and Rowe, 2000).

Geosynthetic clay liners containing enhanced bentonites, such as bentonite polymer composite (BPC), have been developed to be more resistant to chemical interactions. Examples of common BPC types include bentonites that have been dry-mixed with hydrophilic polymers or have been modified with sodium polyacrylic acid (Na-PAA) (Scalia et al., 2018; Rowe and Jefferis, 2022). The polymers used in BPC-GCLs often are proprietary and may be linear, branched, or cross-linked. The polymers also may be electrically neutral, cationic, or anionic (most common).

Substantial experimental research has been conducted to advance understanding of transport properties of BPC, with a primary focus on evaluating k for aggressive solutions (e.g., Ashmawy et al., 2002; Scalia et al., 2014; Tian et al., 2016a,b, 2019; Ozhan, 2018; Salemi et al., 2018; Prongmanee et al., 2018; Prongmanee and Chai, 2019; Chen et al., 2019; Yu et al., 2019; Chai and Prongmanee, 2020; Gustitus and Benson, 2020; Li et al., 2021; Zainab et al., 2021; Norris et al., 2022a; Wireko et al., 2022,). BPC and BPC-GCLs have been reported to maintain low k even when exposed to solutions with high ionic strength and extreme pH (Scalia et al., 2018). Scalia et al. (2014) hypothesized that the primary mechanism for maintaining low k in BPC was pore-clogging by polymer hydrogel. Scanning electron microscope imaging of BPC performed by Tian et al. (2016b, 2019) and Li et al. (2021) further supported this hypothesis. Studies also have shown that the measured k of BPC may be highly impacted by the degree of polymer elution that occurs during permeation (e.g., Scalia et al., 2014; Tian et al., 2019; Rowe & Hamdan, 2021; Wireko & Abichou, 2021; Norris et al., 2022a,b).

1.2 Diffusion in Sodium Bentonite (NaB) and BPC

Far fewer experimental studies to date have focused on evaluating diffusion coefficients of BPC (e.g., Bohnhoff et al., 2014; Bohnhoff and Shackelford, 2015; Tong et al., 2021). However, diffusion is known to be a significant if not dominant transport mechanism for transport through NaB and BPC-GCLs (e.g., when $k < 10^{-9}$ m/s), such that modelling long-term performance of barrier systems requires knowledge of both the k and diffusion properties (Shackelford 2014). Measurement of diffusion coefficients of bentonites and GCLs can be performed using a range of well-established techniques (e.g., steady-state through-diffusion, column, and half-cell methods), but generally has not been as standardized or commonly performed as hydraulic conductivity testing. Most of the diffusion data in the literature for NaB and BPC used in modern GCLs has been obtained using the through-diffusion test method (e.g., Figure 1a) or the dialysis leaching test (DLT) method (Figure 1b). Results presented subsequently in Section 2 were from studies utilizing these two test method types.

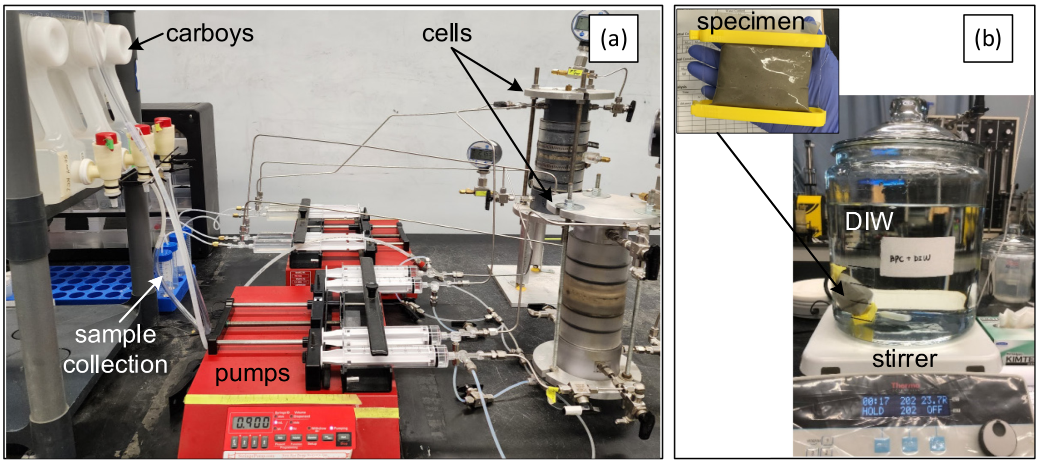

Figure 1. *Photographs of typical testing methods used to measure diffusion coefficients of BPC reported in the literature: (a) through-diffusion method; (b) dialysis leaching test (DLT) method.*

Extensive detail of the through-diffusion method utilizing syringe pumps to provide a concentration gradient, such as pictured in Figure 1a, is available in the literature (e.g., see Malusis et al., 2001). Details of the DLT method (Figure 1b) are available in Tong et al. (2019) and Adeleke et al. (2023), and are summarized herein for convenience due to the more recent development of the technique. In the DLT method, specimens are prepared by mixing air-dried bentonite with the source salt solution to a target gravimetric water content. The mixture then is placed into a dialysis membrane bag (MWCO 6-8,000 Daltons, Spectra/Por 1, Spectrum Laboratories, Inc., Rancho Dominguez, CA), and sealed with clamps on each end. To begin diffusion testing, the bag is submerged in DIW (typically 2.5 – 7.0 L) in a covered glass jar on a temperature-controlled magnetic stir plate. Due to the chemical gradient between the DIW and the specimen, solute diffusion occurs from inside the specimen outward through the bag and into the DIW. The water in the jar is sampled for chemical analysis and replaced with fresh DIW at regular intervals. The mass and volume of the specimen are measured at each interval using a scale and volumetric measuring device. These measurements are used to determine apparent diffusion coefficients and the average concentrations in the specimen over time.

Data regarding the impact of polymer amendment on diffusion properties of BPC remains limited, hindering understanding of transport mechanisms and prediction of long-term barrier performance. This paper provides a synthesis of data in the literature as well as from ongoing studies focused on diffusion of inorganic contaminants through BPC.

2 SOLUTE DIFFUSION THROUGH BPC

This section presents data from previous and ongoing studies of solute diffusion through BPC. Specifically, diffusion coefficients for chloride are summarized and compared for tests performed on NaB and BPC specimens using either the through-diffusion method (Fig. 1a) or the DLT method (Fig. 1b). Properties of the bentonites associated with the results presented in Sections 2.1 and 2.2 are summarized in Table 1. For the material ID, the first number represents the material type (e.g., BPC1 and BPC2 are different BPC types) and the second number represents the polymer content (e.g., BPC1-4 has 4% polymer by mass).

Table 1. *Sodium bentonite (NaB) and bentonite polymer composite (BPC) properties.*

Material ID	Material Description[a]	CEC[b] (cmol$^+$/kg)	Bound Cations (%)[b]				Liquid Limit (%)
			Na^+	K^+	Ca^{2+}	Mg^{2+}	
NaB1	NaB (CG-50)[c]	80.4	43	1.5	41	14.5	253[g]
NaB2	NaB (MX-80)[c]	88.6	42.7	12.3	40.5	4.5	336[g]
NaB3	NaB (CG-50 Bentomat)[d]	47.7	31.0	0.8	20.8	6.4	478[g]
NaB4	NaB[e]	78.3	47	2	36	15	420[g]
BPC1-4	NaB (CG-50) dry mixed with 4 % polymer (Resistex)[c]	115.8	63.8	3.5	24.7	8	403[h]
BPC1-8	NaB (CG-50) dry mixed with 8 % polymer (Resistex)[c]	98.2	58.6	1.6	30.8	9	451[h]
BPC2-4	NaB (CG-50) dry mixed with 4 % polymer (Universal)[c]	159.6	52	14	20	14	421[h]
BPC2-8	NaB (CG-50) dry mixed with 8 % polymer (Universal)[c]	136.7	42.1	2.2	32.8	22.9	550[h]
BPC3-20.8	In situ polymerized NaB with 20.8 % polymer[f]	142.6	90	2	6	2	-
BPC4-3.2	BPC-20.8 dry mixed with NaB4; 3.2 % polymer total[e]	-	-	-	-	-	-

[a] All NaB and BPC materials provided by CETCO (Hoffman Estates, IL, USA).
[b] Determined from tests performed in accordance with ASTM D 7503.
[c] Data from ongoing/unpublished studies by authors.
[d] Data from Malusis and Shackelford (2002).
[e] Data from Tong et al. (2021).
[f] Data from Bohnhoff and Shackelford (2013) and Scalia et al. (2014).
[g] Determined from tests performed in accordance with ASTM D 4318.
[h] Determined via fall cone method (BS 1377-2).

2.1 Results from Testing with Calcium Chloride Solutions

Results from diffusion testing with $CaCl_2$ solutions are presented in Figure 2. The data are from Tong et al. (2018, 2021), Adeleke et al. (2023), and studies currently underway by the authors. All of the results presented in Figure 2 were obtained using the DLT method (see Tong et al. (2019) for details of the method). Plots (a), (b), and (c) represent data from tests performed with dilute (10-20 mM), medium (40-

50 mM), and aggressive (100 mM) $CaCl_2$ source concentrations, respectively. Apparent diffusion coefficients (D_a) for chloride are compared for NaB without polymer amendment (NaB1, shown as crosses) and five different BPCs (see Table 1 for BPC information).

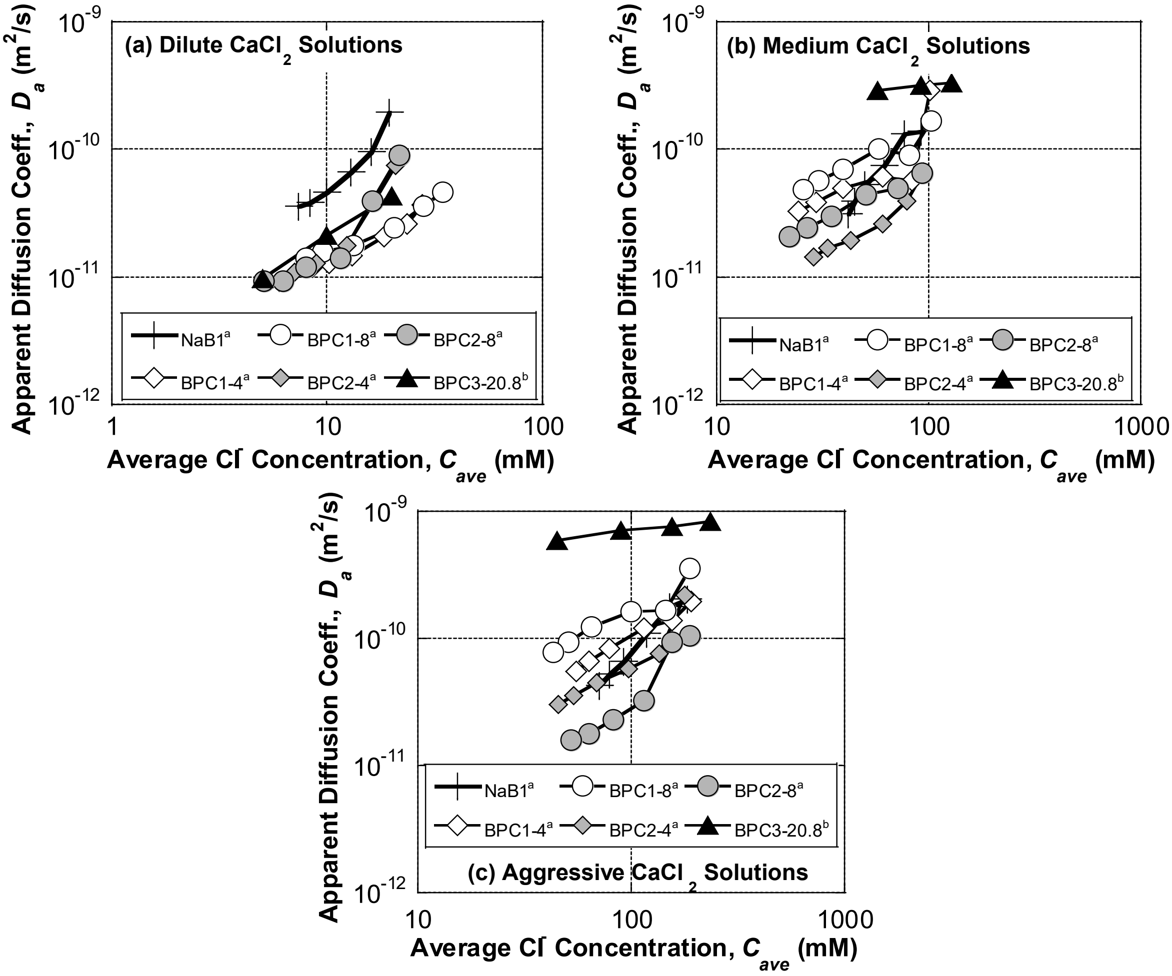

Figure 2. *Diffusion coefficients for Cl^- versus average Cl^- concentration within the bentonite for tests performed with source $CaCl_2$ solutions ranging from (a) dilute (10 - 20 mM), to (b) medium (40 – 50 mM), and (c) aggressive (100 mM). [[a]data from Adeleke et al. (2023); [b]data from Tong et al. (2018, 2021)]*

Several observations can be drawn from Figure 2. Firstly, for dilute $CaCl_2$ solutions (Figure 2a), the BPCs consistently exhibit lower (better) D_a than the unamended NaB. When the average chloride concentration (C_{ave}) in the BPC is ≤ 10 mM, the D_a values for all five BPC types are similar and fall between 9 x 10^{-12} and 2 x 10^{-11} m^2/s. As C_{ave} increases above 10 mM, D_a values of the different BPC types begin to diverge. For the medium and aggressive solutions (Figures 2b,c), the D_a values for BPC type 1 (BPC1-4 and BPC1-8) were similar to or higher than D_a of the unamended NaB. This occurs despite BPC1-4 and BPC1-8 exhibiting lower k than NaB1 in concurrent hydraulic conductivity testing being performed as part of the same study. In Figures 2b,c, the higher values of D_a for BPC3-20.8 relative to the other materials may be due in part to the higher porosity (n) of those specimens, and thus may not be appropriate for performance comparisons; i.e., values of n for BPC3-20.8 exceeded 0.92, whereas n of the other specimens was typically < 0.9.

To examine potential impacts of the source solution on the D_a results shown in Figure 2, example data sets also are plotted together for different initial $CaCl_2$ source concentrations in Figure 3. As shown in Figure 3, for a given BPC material, values of D_a followed a similar trend with increasing C_{ave}, regardless of the initial source solution concentration used for the individual test.

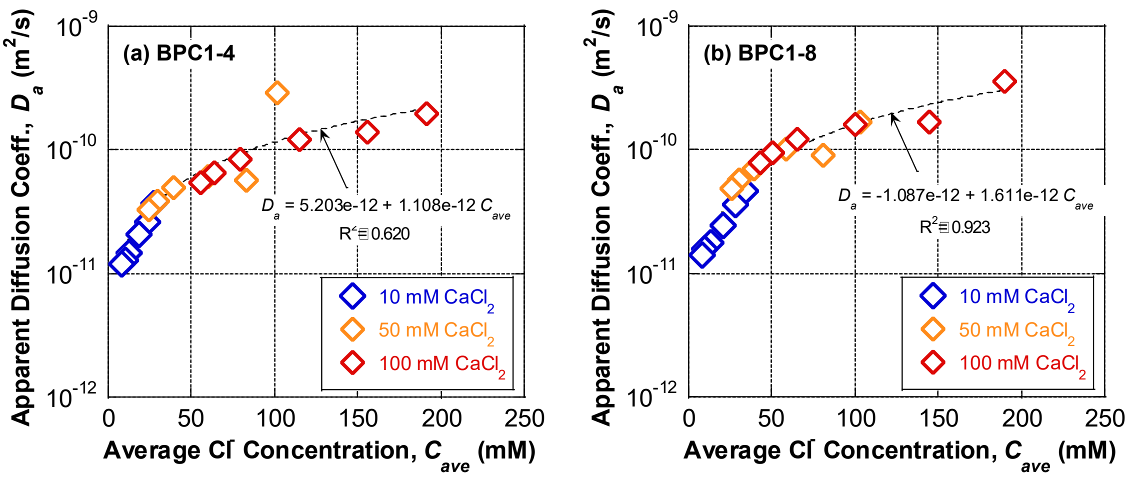

Figure 3. *Diffusion coefficients for Cl^- versus average concentration for two BPC materials: (a) BPC1-4; (b) BPC1-8. Tests were performed with different source solution $CaCl_2$ concentrations, as denoted in the figure legends.*

Overall, for specimens exposed to $CaCl_2$ solutions, higher polymer content does not appear to consistently correlate to better (lower) D_a values, despite corresponding improvements in k with increased polymer loading reported in the literature. In fact, the opposite appears to be true in Figure 2, whereby BPC with 8 % polymer typically exhibited higher D_a than the same BPC type with only 4 % polymer. These results support expectations that the presence of hydrogel in the bentonite pores impacts solute diffusion mechanisms and rates very differently from the concomitant impacts to fluid flow and advective flux.

2.2 Results from Testing with Potassium Chloride Solutions

Results from diffusion testing with KCl solutions are presented in Figures 4 and 5. The data for BPC and NaB2 are from Bohnhoff and Shackelford (2015), Tong and Sample-Lord (2022), and additional studies currently underway by the authors. Data for NaB3 are from Malusis and Shackelford (2002).

Figure 4 shows effective diffusion coefficients (D^*) for chloride obtained using the through-diffusion method (see Malusis et al. 2001 for method details). The first observation from Figure 4 is that although NaB2 and NaB3 are both sodium bentonites that were not amended with polymer, the D^* values for NaB2 are notably lower (better) than those for NaB3. However, in reviewing the bentonite properties in Table 1, this difference can be expected given the higher CEC and bound sodium percentage for NaB2 relative to NaB3. A second observation from Figure 4 is that, similar to tests performed with $CaCl_2$ solutions that were shown in Figure 2, the addition of polymer does not consistently result in lower D^* values relative to unamended NaB. However, the n of the BPC has a clear and significant impact on D^*; this is evidenced by the four sets of results in Figure 4 for the same BPC (BPC3-20.8) tested at different specimen n. Values of D^* for BPC3-20.8 approximately triple as n increases from 0.8 to 0.94.

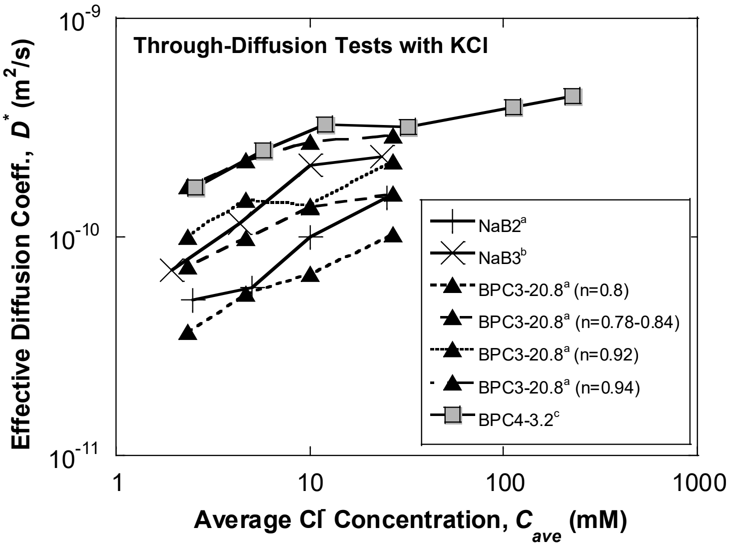

Figure 4. *Effective diffusion coefficients for Cl^- versus average Cl^- concentration within the bentonite for through-diffusion tests performed with KCl solutions. [[a]data from ongoing/unpublished studies; [b]data from Malusis and Shackelford (2002); [c]data from Tong and Sample-Lord (2022)]*

Results in Figure 5 for D_a were obtained using KCl source solutions (50 and 100 mM) and the previously described DLT method. Values of D_a for chloride are compared for two types of unamended NaB and three different BPCs (see Table 1). Observations from Figure 5 are consistent with those from Figure 4 for the through-diffusion results; i.e., (1) NaB with higher CEC exhibits lower diffusion coefficients, and (2) diffusion coefficients of BPC may be lower, similar to, or higher than those of unamended NaB even for cases when *k* of the BPC was lower than that of NaB.

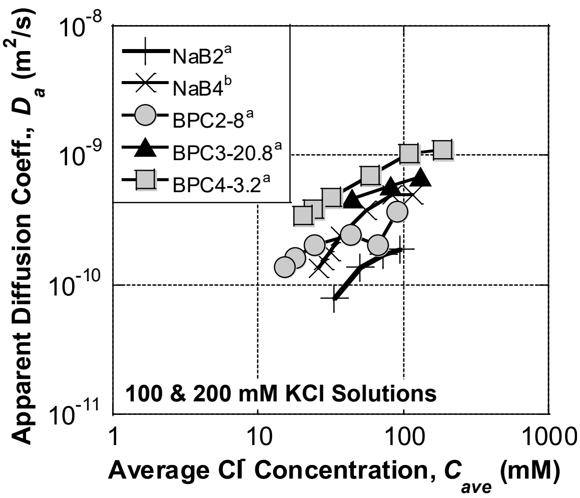

Figure 5. *Apparent diffusion coefficients for Cl^- versus average Cl^- concentration within the bentonite for tests performed with source KCl solutions of 100 and 200 mM. [[a]data from ongoing studies; [b]data from Tong et al. (2021)]*

Similar to the overall observations from testing with $CaCl_2$ solutions, higher polymer content does not necessarily correlate to better (lower) diffusion coefficient values for KCl solutions, as demonstrated in Figures 4 and 5. Further, the results in Figure 4 suggest that diffusion coefficients of NaB and BPC may be more sensitive to changes in specimen porosity than polymer content.

3 FUTURE DIRECTIONS

Based on the literature to date and results summarized in Figures 2 through 5, BPC may exhibit significantly lower *k* than NaB (e.g., by orders of magnitude) for a given solution, without also exhibiting lower diffusion coefficients than the NaB. In fact, for some cases, diffusion coefficients of BPC may be double to triple those of unamended NaB, whereas for other BPC-leachate combinations the opposite occurs. Solute diffusion through BPC is controlled not only by electrostatic interactions and diffusive pathways within the bentonite fraction but also by diffusion behavior through the polymer network in the pore spaces. Thus, relative to unamended NaB, the diffusion coefficients of BPCs are more variable and complex, and thus more challenging to predict without associated experimental data. Similar to leachate compatibility testing performed for evaluating hydraulic conductivity of GCLs, quantifying diffusion coefficients for BPC-GCLs also requires case-specific testing.

Future research needs include further experimental evaluation of diffusion coefficients of BPC and BPC-GCLs for: (1) a range of leachate chemistries, (2) different BPCs (polymer types, polymer loadings, preparation methods), and (3) variable test conditions (e.g., pre-hydration conditions, effective stress, degree of saturation, temperature). Such data are necessary to allow for accurate transport analysis and modelling long-term performance of barrier systems that utilize BPC-GCLs.

4 CONCLUSIONS

Modelling transport of contaminants through GCLs comprising bentonite polymer composite (BPC) requires knowledge of the hydraulic conductivity, *k*, and diffusion coefficients of the GCL. Diffusion coefficients have been studied for traditional Na-bentonite (Na-B), but data is limited for BPC-GCLs. The data from literature and ongoing studies summarized in this paper indicate that, although BPC may exhibit significantly lower *k* than NaB, diffusion coefficients may not follow the same trend. The diffusion coefficients for BPC are complex due to the presence of the polymer, and can be highly sensitive to porosity and solution strength during testing. Similar to leachate compatibility testing performed for evaluating *k* of GCLs, case-specific testing is also recommended for determining diffusion coefficients of BPC-GCLs.

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