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Influence of sodium-rich water on the hydraulic conductivity of a model soil-bentonite backfill

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ABSTRACT

The purpose of this study was to investigate the influence of sodium-rich water on the hydraulic conductivity of a model soil-bentonite cutoff wall backfill containing uniform sand and sodium bentonite. Four backfill samples were created by combining the sand with bentonite slurry and additional dry bentonite to achieve a total bentonite content of 5 % by dry mass. Each sample was prepared using mix water with a different NaCl concentration, C_M, ranging from 10 to 500 mM. Backfill specimens were permeated in flexible-wall cells under a low effective confining stress (14 kPa) using permeant liquids with NaCl concentrations, C_P, ranging from 10 to 1,000 mM. Final hydraulic conductivities (k_f) for specimens prepared with a given mix water generally increased with increasing C_P, but the increases were modest (i.e., 1.2x to 2.6x). Increases in C_M had a greater impact on k_f relative to C_P, causing up to a two-fold increase in k_f for C_M = 50 mM, a four- to six-fold increase in k_f for C_M = 100 mM, and a 10- to 55-fold increase in k_f for C_M = 250-500 mM relative to the backfill mixed with C_M = 10 mM. The results illustrate the importance of mix water quality as the dominant factor controlling the initial bentonite fabric and, therefore, the hydraulic conductivity of the backfill.

Keywords: backfill, bentonite, cutoff wall, hydraulic conductivity, sodium

1 INTRODUCTION

Soil-bentonite (SB) cutoff walls, or vertical barriers comprising bentonite-admixed soil, are widely used for seepage control and isolation of contaminated groundwater. The bentonite used in most SB walls constructed in the US is sodium (Na) bentonite. The high swell capacity of Na bentonite enables a low permeability backfill to be formulated from a wide range of native soil textures, including sandy materials with little or no fines, by incorporating only a small amount (typically 1-5 %) of bentonite (Ryan and Day, 2003). However, the swell of Na bentonite can be inhibited by waters containing high concentrations of electrolytes (ions), potentially causing the barrier to have an undesirably high hydraulic conductivity.

Regarding the impact of electrolyte solutions on the hydraulic conductivity (k) of SB cutoff walls, most of the recent attention has been given to solutions containing multivalent cations such as Ca^{2+} (see Bohnhoff et al., 2013; Malusis and McKeehan, 2013; Replogle and Malusis, 2017; Norris et al., 2018). Multivalent cations are known to replace the monovalent Na^+ attracted to the bentonite surface and suppress bentonite swell, resulting in higher k. However, high concentrations of Na^+ in water also can suppress bentonite swell and increase k, which is relevant for applications involving seawater intrusion control (Ahn, 2001; Abdoulhalik et al. 2017; Hussain et al. 2019) or containment of water contaminated by sodium-rich waste liquids (see Chen et al., 2018).

In this study, the influence of sodium-rich water on k of SB backfills was investigated by conducting a series of laboratory k tests on model backfills comprising sand and conventional (untreated) Na bentonite. As described by Norris (2018), the k of sand-based SB backfills can be impacted by high electrolyte concentrations in the mix water used to create the backfill, as well as high electrolyte concentrations in the permeant water. Therefore, the backfills were exposed to a wide range of NaCl concentrations (10-1,000 mM) during backfill mixing and/or permeation. The results are presented and compared against the results from the Norris et al. (2018) study, and the practical implications of the results are discussed.

2 MATERIALS AND METHODS

2.1 Sand and Bentonite

The model backfills tested in this study were created using mortar sand as the base soil, simulating cutoff wall installation in a clean sand aquifer. The mortar sand was obtained from Central Builders Supply (Lewisburg, PA, USA) and is a poorly graded, medium-to-fine sand with 90 percent of the particles passing the #40 (0.425 mm) sieve and <5 % fines. This may be considered a worst-case condition, as most SB walls are created with base soil containing more than 5 % native fines. The sand is similar to that used to produce model backfills in previous studies by Malusis and Di Emidio (2014) and Norris et al. (2018).

The bentonite used in this study is CETCO Premium Gel (Hoffman Estates, Illinois), a powdered sodium bentonite that contains approximately 70% montmorillonite/smectite and meets the requirements for viscosity and filtrate loss in Section 9 of the American Petroleum Institute (API) specification 13A. The cation exchange capacity (CEC) ranges from 70-82 $cmol_c$/kg, with sodium comprising 75 % of the exchangeable cations (Malusis and Barlow 2020).

2.2 Electrolyte Solutions

The electrolyte solutions listed in Table 1 were used as mix waters to create the backfills and/or as permeant liquids to evaluate the hydraulic conductivity of the backfills. The solutions were prepared by mixing deionized water with anhydrous, enzyme grade NaCl and $CaCl_2$ (Fisher Scientific, Fair Lawn, NJ). All of the solutions contained 0.5 mM $CaCl_2$ (hardness = 50 mg/L as $CaCO_3$), but the NaCl concentrations varied from 10 mM to 1,000 mM to capture a wide range of concentrations that may be encountered in the field. The solution containing 0.5 mM $CaCl_2$ and 10 mM NaCl (Solution 1) is similar to the tap water supplied to the geotechnical laboratory on the Bucknell University campus and is representative of potable water commonly used as mix water on soil-bentonite cutoff wall projects.

Table 1. *Summary of electrolyte solutions used in study.*

Solution No.	Concentrations (mM) NaCl	$CaCl_2$	*RMD* ($M^{0.5}$)	*I* (mM)	*EC* (mS/m)
1	10	0.5	0.45	12	132
2	50	0.5	2.24	52	597
3	100	0.5	4.47	102	1,118
4	250	0.5	11.2	252	2,592
5	500	0.5	22.4	502	5,245
6	1000	0.5	44.7	1002	10,540

Notes: *RMD* = ratio of the molarities of monovalent to divalent cations = $M_M/(M_D)^{0.5}$; *I* = Ionic Strength; *EC* = Electrical Conductivity

2.3 Backfill Preparation

Five model backfills were created in this study, as summarized in Table 2. Each backfill was prepared by combining the sand, dry bentonite, and bentonite-water slurry in a benchtop mixer (Hobart Corporation, Troy, OH). Slurries were prepared by blending the bentonite and one of the mix waters in Table 1 (excluding Solution 6) for 10 minutes in a Hamilton Beach (Glen Allen, VA) blender at the highest speed, simulating a colloidal shear mixer used in the field. The bentonite content in each slurry was modified as needed to obtain a Marsh funnel viscosity (ASTM D6910) of at least 35 s. The slurries with the three lowest NaCl concentrations in the mix water (10, 50, and 100 mM) were prepared with 5 % bentonite by weight, whereas the slurries mixed with the 250 mM and 500 mM NaCl solutions required more bentonite (7 % and 11 %, respectively) to achieve the required minimum viscosity. The proportions of sand, dry bentonite, and slurry were adjusted for each mixture so that each backfill contained a total bentonite content of 5 % by dry weight and exhibited a slump (ASTM C143) within the range of 125±12.5 mm. This slump range is within the typical range specified for SB backfill (Evans, 1993).

2.4 Hydraulic Conductivity (*k*) Testing

Flexible-wall *k* tests were performed on specimens of each model backfill using Method C (falling headwater-rising tailwater method) of ASTM D5084. The procedures and apparatus were the same as those described by Malusis et al. (2009), and involved the use of cylindrical acrylic sleeves placed around the flexible membrane to provide lateral support to the backfill before consolidation. Each specimen was consolidated under a low effective confining stress of 14 kPa (2 psi) for approximately 48 hours before the start of permeation. This confining stress was chosen to simulate the low effective stresses that typically develop in a shallow (<10 m deep) SB cutoff wall (e.g., see Ruffing et al., 2010; Malusis et al., 2017; Evans and Ruffing, 2019; Malusis and Barlow, 2020; Evans et al., 2021).

Table 2. *Bulk backfill samples prepared in study (total bentonite content = 5 % by dry weight).*

Backfill No.	Slurry Mix Water Concentrations (mM) NaCl	$CaCl_2$	X_{BS} (%)	X_{DB} (%)	Backfill Water Content (%)	Backfill Slump (mm)
1	10	0.5	1.76	3.24	40.2	130
2	50	0.5	1.71	3.29	39.7	121
3	100	0.5	1.55	3.45	36.7	123
4	250	0.5	2.07	2.93	33.0	127
5	500	0.5	3.15	1.85	31.1	121

Notes: X_{BS} = dry weight percentage of bentonite contributed to backfill by slurry; X_{DB} = dry weight percentage of dry bentonite added to backfill

In total, 16 backfill specimens were tested in this study, as shown in Table 3. The concentrations of NaCl in the slurry mix water and the permeant water are designated in Table 3 as C_M and C_P, respectively. Specimens created using mix water with a given C_M were permeated with water containing a different C_P that was equal to or higher than C_M. In general, the tests were conducted until the following termination criteria were achieved: (1) steady hydraulic conductivity was observed over time; (2) a minimum of two pore volumes of flow were passed through the specimen; and (3) the electrical conductivity (*EC*) of the effluent was within ±10 % of the *EC* of the influent. The only exception was the control test on Specimen 1a (C_M = C_P = 10 mM), which was terminated after one pore volume of flow.

Table 3. *Summary of backfill test specimens subjected to hydraulic conductivity testing.*

Backfill No.	Specimen ID	C_M (mM)	C_P (mM)	Initial Porosity (---)	Initial Dry Unit Weight (Mg/m^3)
1	1a	10	10	0.52	1.27
	1b	10	100	0.54	1.23
	1c	10	250	0.53	1.25
	1d	10	500	0.52	1.27
	1e	10	1000	0.55	1.21
2	2a	50	50	0.53	1.25
	2b	50	100	0.54	1.23
	2c	50	250	0.53	1.24
	2d	50	500	0.52	1.27
3	3a	100	100	0.51	1.31
	3b	100	250	0.50	1.33
	3c	100	500	0.51	1.31
4	4a	250	250	0.49	1.37
	4b	250	500	0.48	1.39
	4c	250	1000	0.49	1.36
5	5a	500	500	0.48	1.39

Notes: C_M = NaCl concentration in slurry mix water; C_P = NaCl concentration in permeant water

3 RESULTS AND DISCUSSION

Measured *k* values for all of the test specimens are plotted as a function of pore volumes of flow in Figure 1, and final *k* values (k_f) based on the average of the final four measurements are summarized in Table 4. Steady *k* was achieved after approximately two pore volumes of flow or less for all of the specimens, except for specimen 3c (C_M = 100 mM, C_P = 500 mM), which required approximately four pore volumes of flow to achieve steady k. The specimens mixed with the simulated potable water (C_M = 10 mM) had the lowest range of hydraulic conductivities and, for $C_P \leq 250$ mM, exhibited k_f equal to or lower than the maximum k of $\leq 1.0 \times 10^{-9}$ m/s typically specified for SB cutoff walls in geoenvironmental containment applications. All but one of the remaining specimens exhibited $k_f \geq 10^{-9}$ m/s.

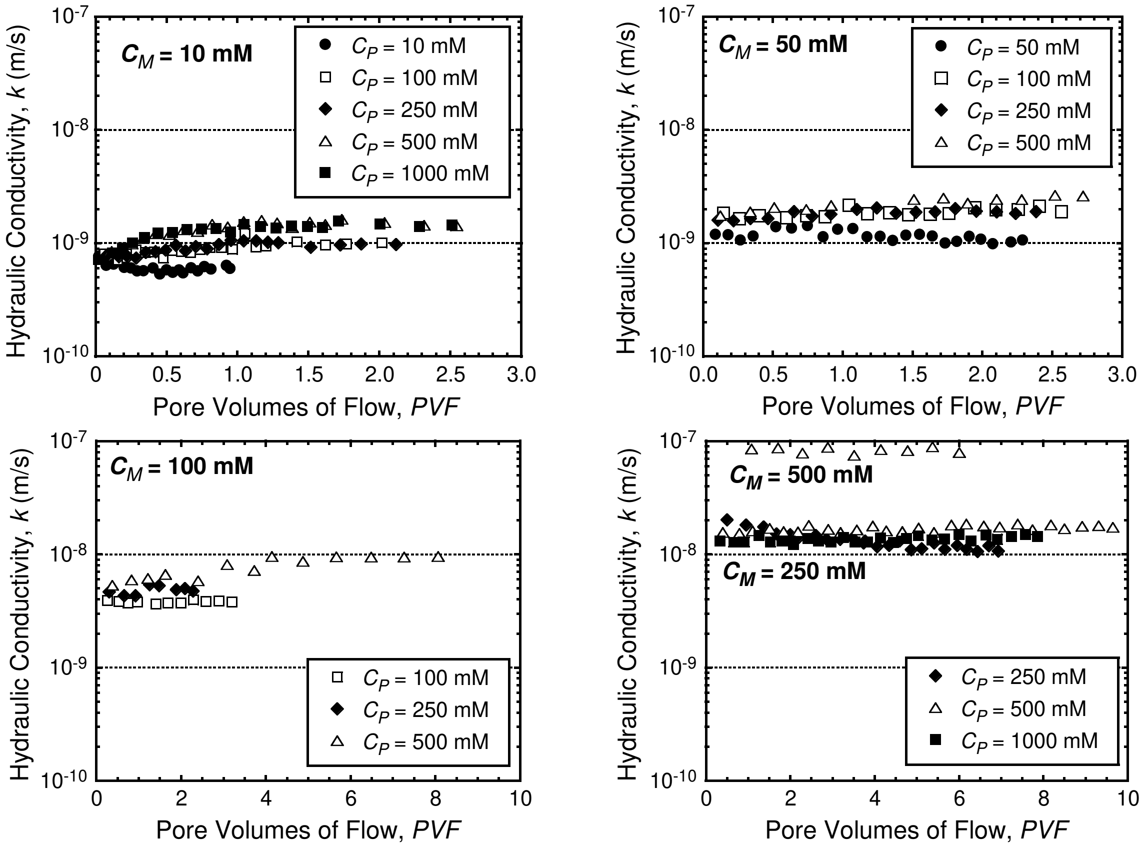

Figure 1. *Hydraulic conductivity versus pore volumes of flow for model sand-bentonite backfill specimens tested in study.*

Table 4. *Summary of hydraulic conductivity test results.*

Specimen ID	C_M (mM)	C_P (mM)	k_f (m/s)	k_f/k_m (---)	k_f/k_{10} (---)	Test Durations	
						t (days)	***PVF***
1a	10	10	6.5×10^{-10}	1.0	1.0	39	1.0
1b	10	100	1.0×10^{-9}	1.5	1.0	54	2.0
1c	10	250	9.8×10^{-10}	1.5	1.0	54	2.1
1d	10	500	1.5×10^{-9}	2.3	1.0	48	2.6
1e	10	1000	1.5×10^{-9}	2.3	1.0	49	2.5
2a	50	50	1.0×10^{-9}	1.0	---	48	2.3
2b	50	100	2.0×10^{-9}	2.0	2.0	36	2.6
2c	50	250	1.9×10^{-9}	1.9	1.9	37	2.5
2d	50	500	2.6×10^{-9}	2.6	1.7	31	2.8
3a	100	100	4.0×10^{-9}	1.0	4.0	21	3.2
3b	100	250	4.9×10^{-9}	1.2	5.0	12	2.5
3c	100	500	9.5×10^{-9}	2.4	6.3	27	8.1
4a	250	250	1.1×10^{-8}	1.0	11.2	14	6.9
4b	250	500	1.7×10^{-8}	1.5	11.3	14	9.7
4c	250	1000	1.4×10^{-8}	1.3	10.0	14	7.9
5a	500	500	8.3×10^{-8}	1.0	55.3	5	6.0

Notes: C_M = NaCl concentration in slurry mix water; C_P = permeant liquid NaCl concentration; k_f = final hydraulic conductivity; k_f/k_m = ratio of k_f to k_f for $C_P = C_M$ for specimens mixed with the same C_M; k_f/k_{10} = ratio of k_f to k_f for C_M = 10 mM for specimens permeated with the same C_p

The k_f of backfills mixed with a given C_M generally increased with increasing C_P, as expected due to the higher ion concentrations entering the pores of the specimens during permeation. However, the increases were relatively modest. For example, k_f for the backfill mixed with C_M = 10 mM NaCl but permeated with C_P = 500 or 1,000 mM NaCl was 1.5×10^{-10} m/s, which is higher than k_f for the control specimen ($C_M = C_P$ = 10 mM NaCl) by a factor of only 2.3 (i.e., $k_f/k_m \leq 2.3$ in Table 4). Likewise, values of k_f/k_m of specimens mixed with C_M = 50 mM and 100 mM NaCl were 2.6 or lower, and k_f/k_m for specimens mixed with C_M = 250 mM NaCl were 1.5 or lower.

The mix water NaCl concentration (C_M) had a greater impact on k_f than the permeant water NaCl concentration (C_P). For example, backfill specimens mixed with C_M = 10 mM and permeated with $C_P \geq$ 250 mM exhibited $k_f = 9.8 \times 10^{-10}$ to 1.5×10^{-9} m/s, whereas k_f for specimens mixed with C_M = 250 mM and permeated with $C_P \geq$ 250 mM were approximately 10 to 11 times higher (1.1×10^{-8} - 1.7×10^{-8} m/s, k_f/k_{10} = 10.0 - 11.3 in Table 4). Also, the specimen mixed with C_M = 500 mM and permeated with C_P = 500 mM (specimen 5a) exhibited the highest k_f (8.3×10^{-8} m/s), approximately 55 times greater than k_f for the specimen mixed with C_M = 10 mM and permeated with C_P = 500 mM ($k_f = 1.5 \times 10^{-9}$ m/s, k_f/k_{10} = 55.3 in Table 4). Thus, specimens mixed with a lower C_M but permeated with the same C_P exhibited lower k_f, presumably because a lower C_M allowed for improved swell of the bentonite and creation of a more dispersed fabric in the backfill during hydration while the backfill was being mixed. A better initial fabric created during mixing with lower C_M resulted in smaller changes in k_f after the specimens were confined and permeated with solutions having higher C_P.

The results of this study are consistent with those of a previous study by Norris et al. (2018) in which similar model backfills (total bentonite content = 5.5-5.7 %) were prepared with mix waters containing a low NaCl concentration (10 mM) but a range of $CaCl_2$ concentrations from 0.5 to 25 mM ($CaCl_2$ concentrations in the permeant liquids ranged from 0.5 to 50 mM). The values of k_f from this study and from the Norris et al. (2018) study are plotted as a function of C_M and C_P in Figure 2. The trends obtained in both studies are remarkably similar, in that k_f increased significantly with increasing C_M but varied modestly for a given C_M over a range of C_P. In both cases, the results illustrate the importance of mix water quality as the dominant factor controlling the initial fabric and, therefore, the hydraulic conductivity of the backfill. Use of higher quality mix water allows for better osmotic swell of the bentonite during hydration, because the high concentration gradient between the interlayer water (higher salt concentration) and pore water (lower salt concentration) promotes chemico-osmotic flow of water from the pores to the interlayer region, resulting in more swell and lower k. When permeating with higher salt concentration liquids, the reverse is true, i.e., chemico-osmotic water flow occurs from the interlayer regions into the pores, favouring shrinkage and higher k. However, since the salt concentration in the interlayer region is already high, this reverse effect is limited, resulting in only modest increases in k.

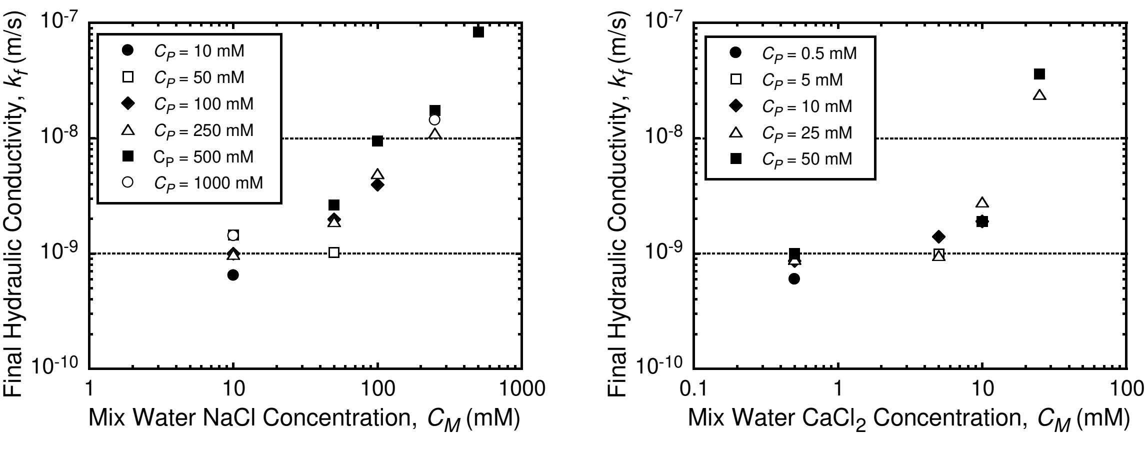

Figure 2. *Final hydraulic conductivities, k_f, versus mix water NaCl concentration, C_M, for backfill specimens in this study (left); k_f versus mix water $CaCl_2$ concentration, C_M, for similar specimens (5.5-5.7 % bentonite) mixed and permeated with $CaCl_2$ solutions (right; data from Norris et al. 2018).*

There are two primary differences in the results from this study relative to the results from Norris et al. (2018). First, the range of $CaCl_2$ concentrations used by Norris et al. (2018) was substantially lower than the range of NaCl concentrations used in this study, since predominantly divalent ($CaCl_2$) solutions have greater impact on the fabric and swell of bentonite at lower concentrations relative to predominantly monovalent (NaCl) solutions. Second, the impacts on k_f caused by mixing the backfill with hard water (elevated Ca^{2+}) could not be reversed by permeating with solutions having lower C_P relative to C_M. For example, Norris et al. (2018) observed that k_f for backfill specimens mixed and permeated with 10 mM or 25 mM $CaCl_2$ solutions were similar to those for replicate specimens mixed with 10 mM or 25 mM $CaCl_2$ but permeated with solutions containing lower C_P. Cation exchange of the divalent Ca^{2+} for the monovalent Na^+ initially on the exchange complex of the bentonite resulted in changes to the bentonite fabric that could not be reversed by permeating with more dilute solutions.

In contrast to the Norris et al. (2018) study, the backfills in this study were not impacted significantly by exchange of Ca^{2+} for Na^+, as the mix waters and permeant waters had low concentrations of Ca^{2+} (i.e., 0.5 mM). Thus, high k_f resulting from mixing with high C_M was reversible by permeating with a more dilute solution to flush the Na^+ from the pores of the backfill. To illustrate this effect, the permeant water in the tests conducted on specimens 2d (C_M = 50 mM), 3c (C_M = 100 mM), and 5a (C_M = 500 mM) was changed to the simulated potable water (C_P = 10 mM NaCl) after the first permeation stage with C_P = 500 mM NaCl. As shown in Figure 3, k decreased sharply to below the k_f for specimen 1a (C_M = C_P = 10 mM; $k_f = 6.5 \times 10^{-10}$ m/s) in all cases. Although the tests were terminated before k became steady in the second permeation stage, the results indicate that k of these specimens was actually *improved* by permeating with C_P = 10 mM after permeating with C_P = 500 mM. The 500 mM NaCl concentration likely was sufficiently high to cause some of the Na^+ in the permeant water to exchange with divalent exchangeable cations (comprising up to 25 % of the exchangeable cations initially on the exchange complex of the bentonite) by mass action during the first stage of permeation, which caused the bentonite to become further enriched with bound sodium and resulted in more swell and lower k as the high NaCl concentrations were flushed from the pores. This hypothesis is supported by the results of Chen et al. (2018), who observed enrichment of bound Na^+ in sodium bentonite GCL specimens after permeation with trona ash leachate containing 645 mM Na^+.

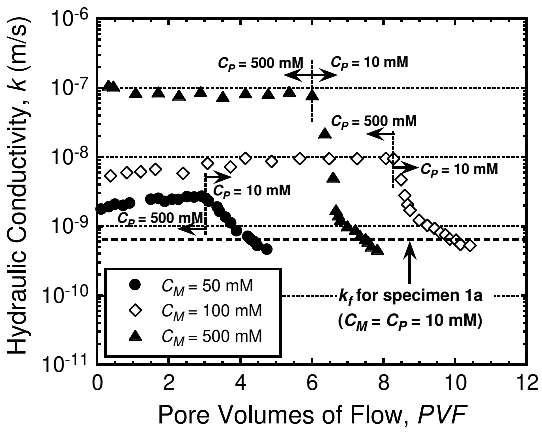

Figure 3. *Changes in hydraulic conductivity of specimens 2d (C_M = 50 mM NaCl), 3c (C_M = 100 mM* NaCl*), and 5a (C_M = 500 mM NaCl) after changing permeant liquid from C_P = 500 mM NaCl to C_P = 10 mM NaCl.*

From a practical perspective, the results of this study provide a conservative view of the impact of sodium-rich water on k of SB backfill, as the model backfills contained predominantly sand (< 5 % native fines) and, therefore, required an appreciable percentage of bentonite (5 %) and higher quality mix water to achieve k values in the vicinity of 10^{-9} m/s. As mentioned previously, the base soil used to create SB backfill typically will contribute more than 5 % native fines to the mixture, such that the backfill requires less bentonite to achieve a low k and, therefore, is more resilient when exposed to high salt concentrations.

To illustrate the beneficial influence of native fines, the authors conducted four additional k tests on model backfill specimens created from a loamy base soil with moderate to low plasticity (fines content = 44-57 %, liquid limit = 16-18 %, plasticity index = 6-7 %, classification = SC-SM or CL-ML based on ASTM D2487) using C_M = 10 mM NaCl or 500 mM NaCl. This base soil was the same base soil used to construct the large-scale SB cutoff wall tested in the field study by Malusis and Barlow (2020). The additional backfills were prepared and tested in the same manner as described above for the sand-based model backfills, but contained a lower bentonite content. The backfill mixed with C_M = 10 mM NaCl contained 1.9 % total bentonite (1.4 % bentonite from slurry and 0.5 % dry bentonite), whereas the backfill mixed with C_M = 500 mM NaCl contained 2.6 % total bentonite (all from slurry). The results, shown in Figure 4, illustrate that all three specimens mixed with C_M = 10 mM exhibited similar *k*, and all k values were at or below 10^{-9} m/s regardless of C_P. The *k* of the specimen mixed and permeated with C_M = C_P = 500 mM NaCl was only slightly higher than 10^{-9} cm/s. In both cases, *k* is controlled predominantly by the presence of the native fines rather than the bentonite, rendering the backfills more compatible with high NaCl concentrations relative to the sand-based backfills.

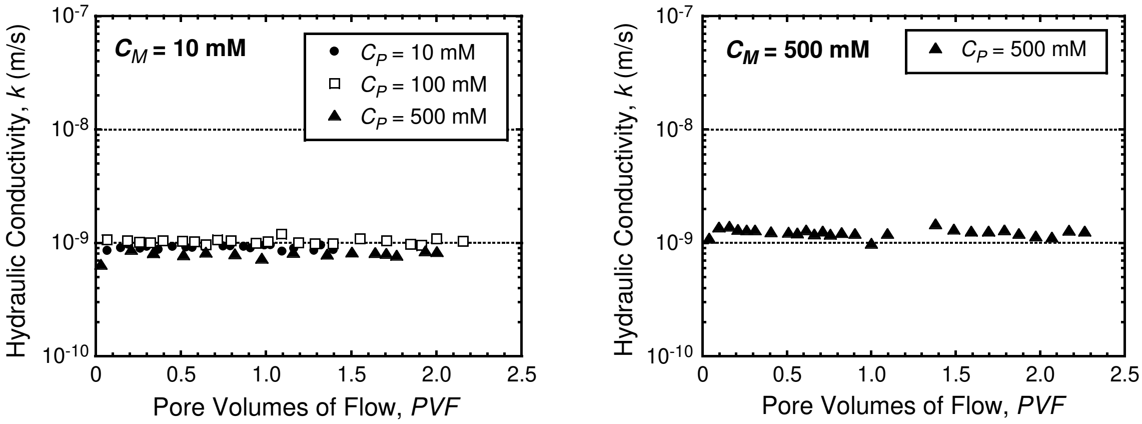

Figure 4. *Hydraulic conductivity versus pore volumes of flow for loamy model backfills (44-57 % native fines, 1.9-2.6 % bentonite):* C_M *= 10 mM NaCl (left),* C_M *= 500 mM NaCl (right).*

4 CONCLUSIONS

The results of this study show that sand-based SB backfills containing 5 % conventional Na bentonite and <5 % native fines can exhibit a low hydraulic conductivity (*k*) of ~10^{-9} m/s when permeated with water having elevated NaCl concentrations (i.e., up to 1,000 mM NaCl) under low effective confining stresses (14 kPa) representative of those in shallow (<10 m deep) SB cutoff walls. The quality of the mix water appears to be the most important factor for achieving low *k*. The backfills in this study exhibited modest increases in *k* (i.e., within a factor of 2.6) with increasing NaCl concentration in the permeant water. However, high NaCl concentrations in the mix water had a much greater impact on *k* relative to high NaCl concentrations in the permeant water. Specimens prepared with mix waters having NaCl concentrations of 100-500 mM exhibited *k* values that were 4 to 55 times higher (for the same permeant liquid) than the *k* values for specimens prepared with simulated potable mix water (10 mM NaCl). Use of higher quality mix water enhances the osmotic swell of the bentonite, resulting in lower *k*. The results also illustrate that (1) increases in backfill *k* caused by high sodium concentrations in the mix water and/or permeant water can be reversed by flushing the sodium from the pores of the backfill, provided that appreciable exchange of divalent cations for bound sodium has not occurred in the bentonite, and (2) sand-based model backfills provide a conservative (worst-case) assessment of the influence of sodium-rich water on *k* due to the absence of appreciable native fines.

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