Membrane Behavior of Model Soil–Bentonite Backfills

Sang-Sik Yeo 1; Charles D. Shackelford 2; and Jeffrey C. Evans 3

Abstract: Two model soil-bentonite (SB) backfills were evaluated for their ability to behave as semipermeable membranes. The base soils for the model backfills consisted of natural clay with 89% (dry weight) fines and a mixture of sand with 5% dry sodium bentonite. Specimens of both base soils were mixed with a sufficient amount of 5% sodium bentonite–water slurry to correspond to 100 mm slumps in accordance with standard practice for SB vertical cutoff walls. Membrane behavior was evaluated by measuring the chemico-osmotic efficiency coefficient \( \omega \), resulting from maintaining a 3.88 mM KCl concentration difference across the specimen. The results indicate that both model backfills act as semipermeable membranes, with \( \omega \) ranging from 0.018 to 0.1 for the natural clay backfill and from 0.118 to 0.166 for the sand–bentonite backfill. More significant membrane behavior (higher \( \omega \)) is correlated with higher consolidation stress, lower void ratio, and lower hydraulic conductivity. The benefit of the chemico-osmotic liquid flux due to the existence of membrane behavior in SB vertical cutoff walls is illustrated through an example analysis using the measured results. The results of the study provide the first quantitative evidence that SB vertical cutoff walls can behave as semipermeable membranes.

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CE Database subject headings: Backfills; Bentonite; Containment; Cutoffs; Hydraulic conductivity; Membrane processes; Osmosis; Slurry walls.

Introduction

Membrane behavior refers to restricted passage of solutes during migration through a porous medium. Restricted solute passage also results in chemico-osmotic flow, or the flow of liquid from lower solute concentration (higher water activity) to higher solute concentration (lower water activity) (Shackelford et al. 2003). As noted by Malusis and Shackelford (2002a), membrane behavior in clays has been shown to influence volume change behavior, cause apparent deviations from Darcy’s law in hydraulic conductivity testing, generate anomalous pore-water pressures in low-permeability geologic formations, and affect the rate of solute migration through aquitards.

Membrane behavior is quantified in terms of a chemico-osmotic efficiency coefficient \( \omega \) also commonly referred to as a reflection coefficient (Malusis and Shackelford 2002a, Shackelford et al. 2003). Several different mathematical expressions exist for \( \omega \), depending on the method used to measure \( \omega \) and the associated boundary conditions (e.g., see Malusis et al. 2001a). However, regardless of these different mathematical expressions, \( \omega \) ranges from zero for a material that does not exhibit membrane behavior to unity for a material that restricts the movement of all solutes, commonly referred to as an “ideal” or “perfect” membrane. Because of the wide variation in pore sizes that typically exists in soils, \( \omega \) for most soils that exhibit membrane behavior typically ranges from greater than zero to less than unity (i.e., 0 < \( \omega \) < 1). Regardless of the degree of solute restriction, porous media that exhibit membrane behavior also are referred to as semipermeable membranes, i.e., because all membranes are still permeable to the solvent (e.g., water).

Because the existence and efficiency of membrane behavior is a function of the pore sizes relative to the sizes of the invading solute species, membrane behavior is usually evident only in soils with relatively small pore sizes, such as clayey soils. Also, the existence and efficiency of membrane behavior in clay soils is affected by the factors that tend to affect the pore sizes, including the stress-strain properties, the types and concentrations of salts in the pore water, and the mineralogy of the clay portion of the soil (e.g., McKelvey and Milne 1962; Kemper and Rollins 1966; Olsen 1969; Kemper and Quirk 1972; Marine and Fritz 1981; Fritz and Marine 1983; Fritz 1986; Olsen et al. 1990; Keijzer et al. 1997; Malusis and Shackelford 2002a; Shackelford et al. 2003). For example, membrane behavior in clay soils tends to be enhanced with increasing consolidation stress and/or decreasing void ratio, decreasing concentration and/or valence of the principal salt cations in the pore water of the clay soil, and increasing content of high activity clay minerals, such as sodium montmorillonite.

With respect to the effect of clay mineralogy, the results of numerous studies have shown that membrane behavior can be significant for sodium bentonite (e.g., Kemper and Rollins 1966; Kemper and Quirk 1972; Fritz and Marine 1983; Keijzer et al. 1997; Malusis et al. 2001a; Malusis and Shackelford 2002a). These results are potentially relevant in geoenvironmental containment applications, because several types of clay barriers con-
sist of either sodium bentonite, such as geosynthetic clay liners (GCLs), or soil mixtures containing sodium bentonite, such as compacted sand–bentonite liners and soil–bentonite (SB) vertical cutoff walls. For example, recent results show that a GCL commonly used as a liner or liner component in waste containment applications can behave as a semipermeable membrane, with \( \theta \) values ranging from 0.08 to 0.69 for KCl concentration differences ranging from 3.9 to 47 mM (Malusis et al. 2001a; Malusis and Shackelford 2002a). Because the primary objective of clay soil barriers used for geoenvironmental containment applications is to minimize or prevent the migration of contaminants through the barrier, the existence of membrane behavior resulting from the sodium bentonite content in clay soil barriers represents a potentially significant beneficial aspect that presently is not considered in the design and evaluation of such barriers (Shackelford et al. 2001, 2003).

Based on the aforementioned considerations, the primary objective of this study is to assess the potential for the existence of membrane behavior in SB vertical cutoff walls. Although SB vertical cutoff walls contain sodium bentonite, and sodium bentonite has been shown to exhibit membrane behavior, no assessment of the potential existence or extent of membrane behavior in SB vertical cutoff walls has been performed. The objective is achieved by measuring the chemico-osmotic efficiency coefficient of two model SB backfills in the presence of a potassium chloride (KCl) solution. The results of these measurements are evaluated in terms of the consolidation stress, void ratio, and hydraulic conductivity of the model backfills, as well as the types and amounts of fines in the backfills.

### Experimental Materials and Methods

#### Model Backfills

The base soils used for the two model backfills in this study were a natural clay soil and a mixture of silica sand with 5% (dry weight) powdered sodium bentonite. The natural clay soil is referred to as Nelson Farm Clay (NFC), because the soil comes from the Nelson Farm area of Fort Collins, Colo. The silica sand and the sodium bentonite are sold commercially under the trade names Colorado Silica Sand® (Dean Bennett Supply Company, Denver, Colo.) and NATURALGEL® (Wyo-Ben, Inc., Billings, Mont.), respectively. NATURALGEL® is commonly specified for use in slurry trenching, diaphragm walls, and as a soil mixture additive. The physical and chemical properties and mineralogical compositions of the constituent soils used for the model backfills are provided in Table 1.

The grain-size distributions (ASTM D 422) and classifications according to the Unified Soil Classification System (ASTM D 2487) for the NFC, silica sand, and sodium bentonite are shown in Fig. 1. As shown in Fig. 1, NFC is classified as a low plasticity clay (CL) with 89% fines (i.e., \( %<0.075 \) mm) and \( \sim 52\% \) clay-sized particles (i.e., \( %<0.005 \) mm), the sodium bentonite is a high plasticity clay (CH) with 100% fines, and \( \sim 94\% \) clay-sized particles, and the sand is poorly graded (SP) with 100% sand-sized particles. The NFC backfill is characteristic of the backfill that would result from construction of an SB vertical cutoff wall within a geologic formation consisting of a fine-grained soil, whereas the sand–bentonite backfill is characteristic of a cutoff wall constructed within a clean, coarse-grained geologic formation, requiring the addition of dry bentonite to the excavated soil to reduce the hydraulic conductivity of the backfill.

#### Liquids

The primary liquids used in this study consist of de-ionized water (DIW) with an electrical conductivity (EC) at 25°C = 0.26 mS/m and a pH=5.58, and a 3.88 mM potassium chloride (KCl) solution (EC=58.7 mS/m, pH=5.35). A 3.88 mM KCl solution was used in this study because solutions containing salts with a monovalent cation, such as potassium (K\(^+\)), tend to result in more significant membrane behavior than solutions containing salts with divalent cations, such as calcium (Ca\(^{2+}\)), and previous experience with KCl solutions showed that the membrane behavior of a GCL increased with decreasing KCl concentration, with the most significant membrane behavior occurring when a 3.9 mM KCl solution was used (Malusis and Shackelford 2002a; Shackelford et al. 2003). The DIW used in this study classifies as Type IV water per ASTM D 1193. The salt solution was prepared by dissolving KCl (powdered, \( >99.8\% \) pure, certified A.C.S., Fisher Scientific, Fair Lawn, N.J.) in the DIW. Tap water (EC \( =14.5 \text{ mS/m}, \text{pH}=7.12 \)) also was used in preparation of the bentonite-water slurry and as a permeant liquid.

#### Bentonite–Water Slurry

Five percent bentonite–water slurry (i.e., 5% bentonite and 95% tap water by weight) was used as the admixture to the base soils to prepare backfills in accordance with the standard procedure for preparing SB backfills used in vertical cutoff walls (e.g., see Evans 1993). After mixing bentonite with tap water using a high-speed blender (Cuisinart®), the bentonite slurry was allowed to hydrate for 24 h. The relevant properties of the bentonite-water slurry subsequently measured are shown in Table 2.

#### Slump Tests

The bentonite–water slurry was mixed with each of the base soils to form the model backfills. For NFC, only the portion of the NFC that passed a No. 10 sieve (2.00 mm) was used for the mixtures to account for the breakdown of clods that typically occurs during field mixing of SB backfills (Evans 1993). The slurry was added in various proportions to determine the amount of slurry (and corresponding water content) required to provide a 100 mm slump, which is within the consistency of 100–150 mm considered as optimum for SB backfills (Xanthakos 1979; D’Appolonia 1980; Spooner et al. 1984; Millet et al. 1992; Evans 1993). The slump was measured according to ASTM C 143. The initial gravimetric water contents for the sand–bentonite backfill and Nelson Farm Clay (i.e., prior to mixing) were 5 and 10%, respectively. Three slump tests were performed for each mixture at any given water content to evaluate the variability in measured slumps, and the amount of slurry mixed with the backfill was varied to provide a range of slump values and a corresponding range in backfill water content values.

The resulting slump values, \( \Delta H \), are plotted versus the backfill water content \( w_b \) for both the NFC and the sand–bentonite backfills in Fig. 2. As shown in Fig. 2, the values of \( w_b \) required to provide a 100 mm slump are 43.1 and 43.3% for the NFC and sand–bentonite backfills, respectively. At these water contents, the total bentonite contents (by weight) for the NFC and sand–bentonite backfills were 2.12 and 7.20%, respectively. The similarity in the slump test results despite the difference in fines contents may be due, in part, to the significantly greater water absorption capacity per unit mass of clay of the high plasticity...
fines (i.e., bentonite) in the sand-bentonite backfill relative to the water absorption capacity of the low plasticity fines in the NFC.

**Consolidation and Hydraulic Conductivity Tests**

The consolidation behavior and hydraulic conductivity of each backfill were measured using fixed-ring consolidometers (C-252, SOILTEST Inc., Evanston, Ill.), similar to those described by Lambe (1951), Olson and Daniel (1981), and Daniel (1994). The consolidation test procedures conformed with ASTM D 2435, except that falling-head hydraulic conductivity tests were performed at the end of each load increment for a period of at least 24 h prior to adding the subsequent load. Specimens of the backfills were mixed using a 6 quart stand mixer (KitchenAid®) and placed in the consolidometer in a remolded state. The base soil and slurry mixing ratios for the test specimens corresponded to those required for a 100 mm slump. After placement in the consolidation cell, the specimens were allowed to equilibrate for at least 24 h without application of a hydraulic gradient prior to the beginning of the loading sequence.

At the end of each stage of loading stage (i.e., 24 h after loading), the specimens were permeated with tap water using the falling-head procedure (e.g., see Lambe 1951; Olson and Daniel 1981; Daniel 1994). Permeation was continued until at least four consecutive values of hydraulic conductivity \( k_h \) were within \( \pm 25\% \) of the mean value for \( k_h \) or within \( \pm 50\% \) for \( k_h \), in accordance with the termination criteria for the falling-head procedure in ASTM D 5084. Since the \( k_h \) values were measured at the end of each loading increment, the final thickness of the specimen after each increment in load was used to calculate \( k_h \).

**Chemico-Osmotic Tests**

The chemico-osmotic tests performed in this study involved the same testing procedure and apparatus as described in detail by

<table>
<thead>
<tr>
<th>Property</th>
<th>Standard</th>
<th>Nelson Farm clay</th>
<th>Silica sand</th>
<th>Bentonite</th>
</tr>
</thead>
<tbody>
<tr>
<td>Specific gravity, ( G_s )</td>
<td>ASTM D 854</td>
<td>2.70</td>
<td>2.62</td>
<td>2.72</td>
</tr>
<tr>
<td>Liquid limit (LL), (%)</td>
<td>ASTM D 4318</td>
<td>32.3</td>
<td>N.A.</td>
<td>497</td>
</tr>
<tr>
<td>Plasticity index (PI), (%)</td>
<td>ASTM D 4318</td>
<td>14.5</td>
<td>N.A.</td>
<td>454</td>
</tr>
<tr>
<td>Principal minerals (%)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Montmorillonite</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cristobalite</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Mixed-Layer Illite/Smectite</td>
<td></td>
<td>22</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Quartz</td>
<td></td>
<td>40</td>
<td>99.8</td>
<td>6</td>
</tr>
<tr>
<td>Plagioclase Feldspar</td>
<td></td>
<td>13</td>
<td></td>
<td>5</td>
</tr>
<tr>
<td>Calcite</td>
<td></td>
<td>13</td>
<td></td>
<td>2</td>
</tr>
<tr>
<td>Other</td>
<td></td>
<td>12</td>
<td></td>
<td>4</td>
</tr>
<tr>
<td>Cation exchange capacity (CEC)</td>
<td></td>
<td>10.1</td>
<td>N.A.</td>
<td>86.1</td>
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<td>Exchangeable metals (meq/100 g)</td>
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<td></td>
<td></td>
<td></td>
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<tr>
<td>Ca</td>
<td></td>
<td>4.2</td>
<td>N.A.</td>
<td>4.4</td>
</tr>
<tr>
<td>Mg</td>
<td></td>
<td>3.1</td>
<td></td>
<td>8.1</td>
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<tr>
<td>Na</td>
<td></td>
<td>0.3</td>
<td></td>
<td>77.5</td>
</tr>
<tr>
<td>K</td>
<td></td>
<td>0.3</td>
<td></td>
<td>0.99</td>
</tr>
<tr>
<td>Sum</td>
<td></td>
<td>7.9</td>
<td></td>
<td>90.99</td>
</tr>
<tr>
<td>Soluble salts (mg/kg)</td>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ca(^{2+})</td>
<td></td>
<td>35.3</td>
<td>1.59</td>
<td>47.0</td>
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<tr>
<td>Mg(^{2+})</td>
<td></td>
<td>8.30</td>
<td>0.46</td>
<td>14.1</td>
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<tr>
<td>Na(^{+})</td>
<td></td>
<td>22.7</td>
<td>8.61</td>
<td>2.097</td>
</tr>
<tr>
<td>K(^{+})</td>
<td></td>
<td>3.40</td>
<td>0.52</td>
<td>61.2</td>
</tr>
<tr>
<td>CO(_{3}^{−})</td>
<td></td>
<td>&lt;0.4</td>
<td></td>
<td>&lt;0.4</td>
</tr>
<tr>
<td>HCO(_{3}^{−})</td>
<td></td>
<td>78.7</td>
<td>28.0</td>
<td>674</td>
</tr>
<tr>
<td>SO(_{4}^{−})</td>
<td></td>
<td>37.5</td>
<td>&lt;0.03</td>
<td>3.610</td>
</tr>
<tr>
<td>CI(^{−})</td>
<td></td>
<td>20.9</td>
<td>0.49</td>
<td>238</td>
</tr>
<tr>
<td>NO(_{3}^{−})</td>
<td></td>
<td>53.3</td>
<td>1.97</td>
<td>217</td>
</tr>
<tr>
<td>Soil pH</td>
<td>ASTM D 4972</td>
<td>8.3</td>
<td>7.1</td>
<td>7.9</td>
</tr>
<tr>
<td>Soil electrical conductance (EC)</td>
<td></td>
<td>36.0</td>
<td>1.5</td>
<td>193</td>
</tr>
</tbody>
</table>

\( ^{a}\)N.A.= not applicable.

\( ^{b}\)Based on x-ray diffraction analyses performed by Mineralogy Inc., Tulsa, Ok.

\( ^{c}\)Procedures described in Shackelford and Redmond (1995).

\( ^{d}\)Measured using saturated soil paste.

**Table 1. Physical and Chemical Properties and Mineralogical Compositions of Constituent Soils Used for Model Backfills**
This procedure involves circulating electrolyte solutions with different salt (solute) concentrations at the same constant rate through porous stones adjacent to the top and bottom boundaries of a specimen to establish a concentration difference across the specimen. The specimen is contained in a rigid, circular acrylic cell, and the porous stones are contained within an adjustable top piston and rigid-base pedestal, both sealed to the cell with O-rings. The top piston can be adjusted, e.g., via a load frame, but is locked in place during the test to prevent specimen expansion and to control the thickness, i.e., void ratio, of the specimen, and circulation through the porous stones is controlled such that no net volume change can occur. As a result, the cell and the circulation plumbing represent a closed system during the test, e.g., see Malusis et al. 2001a.

The established concentration difference induces a chemico-osmotic pressure difference, $D_P$, directed from the low concentration side (bottom) to the high concentration side (top) of the specimen, as a result of prohibiting chemico-osmotic flux of solution through the specimen. The concentration difference also results in solute diffusion from the high concentration side (top) to the low concentration side (bottom) of the specimen. If desired, the circulation outflow from the top and bottom porous stones can be collected simultaneously for measurement of solution chemistry (EC, pH, solute concentrations) to evaluate the diffusive solute flux. Malusis et al. (2001a) and Malusis and Shackelford (2002a) provide more extensive details regarding the testing apparatus and procedures.

In the absence of volume change, the chemico-osmotic efficiency coefficient $\omega$ is defined as follows (Malusis et al. 2001a):

$$\omega = \frac{\Delta P}{\Delta \pi}$$

where $\Delta \pi$=theoretical chemico-osmotic pressure difference across an ideal semipermeable membrane (i.e., $\omega=1$) subjected to an applied difference in solute concentration (e.g., Olsen et al. 1990). The value for $\Delta \pi$ in Eq. (1) can be calculated based on the source salt concentrations at the specimen boundaries in accordance with the van’t Hoff expression or (Malusis et al. 2001a; Malusis and Shackelford 2002a)

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Table 2. Measured Properties of 5% Bentonite–Water Slurry

<table>
<thead>
<tr>
<th>Property</th>
<th>Standard</th>
<th>Bentonite–water slurry</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density (g/cm$^3$)</td>
<td>Xanthakos (1979)</td>
<td>1.025</td>
</tr>
<tr>
<td>Viscosity (Marsh-cone time) (s)</td>
<td>API-RP-13B-1$^a$</td>
<td>40</td>
</tr>
<tr>
<td>pH</td>
<td>ASTM D 4972$^b$</td>
<td>9.15</td>
</tr>
<tr>
<td>Electrical conductivity (EC) (mS/m) at 25°C</td>
<td></td>
<td>94.7</td>
</tr>
</tbody>
</table>


$^b$Measured using saturated soil paste.
\[ \Delta \pi = vRT\Delta c = vRT(c_{ob} - c_{ot}) \]  

where \( v \) = number of ions per molecule of the salt (\( v = 2 \) for KCl); \( R \) = universal gas constant (8.314 J mol \(^{-1} \) K \(^{-1} \)); \( T \) = absolute temperature (K), \( \Delta c \) = salt concentration difference; \( c_{ob} \) = initial (source) concentration of solute species at the bottom of the specimen; and \( c_{ot} \) = initial (source) concentration of solute species at the top of the specimen.

As noted by Malusis et al. (2001a) and Malusis and Shackelford (2002a), using the source solute concentrations versus using the average solute concentrations across the top and bottom boundaries to calculate \( \Delta \pi \) in accordance with Eq. (2) results in conservative (low) estimates of \( \omega \), but obviates the need to measure solute concentrations in the circulation outflow. For example, based on the results of tests performed by Malusis and Shackelford (2002a) using a GCL and similar testing conditions (e.g., salt, boundary concentrations, specimen thickness), the \( \omega \) values calculated using \( \Delta \pi \) as defined by Eq. (2) were approximately 6% lower than those using \( \Delta \pi \) defined in terms of the average solute concentrations across the top and bottom boundaries.

Specimens of the model backfills were prepared using a 6 quart stand mixer (KitchenAid®) and placed on the base pedestal in the cell in a remolded state resulting in a nominal diameter of 71.1 mm. The base soil and slurry mixing ratios for the test specimens corresponded to those required for a 100 mm slump. The specimens were separated from the porous stones by filter paper (Whatman No. 41) and a nonwoven geotextile (Geotex® 401, SI solutions, Chattanooga, Tenn.), to prevent clogging of the porous stones and to prevent squeezing of the specimen between the cell wall and the side of the loading piston.

The mass of each backfill placed in the test cell was based on the amount of the backfill required to provide a desired void ratio for a given initial thickness. This consideration resulted in 74.0 g of NFC backfill being placed for an initial thickness of 10 mm corresponding to an initial void ratio of 1.008, and 73.5 g of sand–bentonite backfill being placed for initial thicknesses of 11 mm corresponding to an initial void ratio of 1.212. The desired thickness was obtained by compressing the top piston after placing the assembled cell in a load frame (Model 55760, GEOTEST Co., Evanston, Ill.). After completion of compression, the top piston was locked in place to prevent further volume expansion of the specimen during the test due to swelling of the bentonite.

The osmotic cell was connected to a triaxial control panel, and the specimen in the osmotic cell was permeated under backpressure (hydraulic gradient <30) with DIW before testing to saturate the specimen and to enhance the potential for membrane behavior by removing excess soluble salts. As shown in Fig. 3, this permeation was continued until the EC of the effluent from the specimen was \( \approx 60\% \) of the measured EC for the 3.88 mM KCl source solution of 58.7 mS/m. Although the duration of permeation with DIW is arbitrary, this extent of reduction in the EC is consistent with that of previous studies involving evaluation of the membrane behavior of GCLs (e.g., Malusis et al. 2001a; Malusis and Shackelford 2002a; Shackelford and Lee 2003).

After permeation, the system was closed, and DIW was circulated at the top and bottom boundaries of the specimen for 5 days to establish a steady baseline pressure difference. The chemico-osmotic tests then were initiated by circulating the 3.88 mM KCl solution through the top piston (i.e., \( c_{ob} = 3.88 \text{ mM KCl} \)), while continuing circulation of DIW in the base pedestal (i.e., \( c_{ot} = 0 \)). This concentration difference across the specimen results in \( \Delta \pi \) of \(-18.9 \text{ kPa} \) for an ideal membrane in accordance with Eq. (2). The circulation outflows from the top and bottom of the specimen were collected and analyzed for EC using a probe (Hach conductivity/TDS meter Models 44600, HACH Co., Loveland, Colo.) to check the imposed boundary conditions. No attempt was made to maintain the steady pressure difference for a sufficient period to achieve steady-state solute diffusion, because such test durations were expected to range from 30 to 60 days, or longer, for each test condition based on the results previous studies (e.g., Malusis and Shackelford 2002b; Shackelford and Lee 2003). As a result, the circulation outflows from the specimen were not sampled for solute concentrations.

After the induced chemico-osmotic pressure had reached steady state, the specimen was compressed an additional 1 mm to provide a new (lower) void ratio, and the same boundary liquids were circulated again until a new steady state in induced chemico-osmotic pressure difference had been reached resulting in a new \( \omega \) value. This procedure was repeated two times to provide a total of three measured \( \omega \) values corresponding to three different specimen thicknesses and three different void ratios for each of the two backfills. Thus, the specimen thicknesses for the NFC backfill were 10, 9, and 8 mm, whereas the specimen thicknesses for the sand-bentonite backfill were 11, 10, and 9 mm. At the end of the test, the final thickness of the specimen was checked, the cell was disassembled, the water content of the specimen was measured, and the void ratio was calculated assuming saturated conditions.

**Results**

**Consolidation and Hydraulic Conductivity**

Results of the consolidation tests (loading stage only) and hydraulic conductivity measurements performed on test specimens of the model backfills prepared at 100 mm slumps are shown in Fig. 4. Traditional plots of the void ratio (\( e \)) versus the logarithm of the

![Fig. 3. Electrical conductivity of effluent resulting from permeating specimens of two model backfills with de-ionized water prior to chemico-osmotic stage of tests](image)


consolidation stress ($\sigma$), or $e - \log \sigma$ curves, for both model backfills are presented in Fig. 4(a). As expected, stress history is not evident in the $e - \log \sigma$ curves because the specimens were prepared in a remolded condition. For a given stress, the void ratio for the sand–bentonite backfill is higher than that for the NFC backfill, probably due to the significantly greater swelling capacity of the high plasticity bentonite in the sand–bentonite backfill relative to the low plasticity fines in the NFC backfill, and the better gradation associated with the NFC backfill.

However, the trends in the $e - \log \sigma$ curves for both backfills are similar, with a coefficient of compression $C_e$ for the NFC backfill of 0.270 and a $C_e$ for the sand–bentonite backfill of 0.212.

The measured hydraulic conductivity $k_h$ values for both backfills are plotted as a function of the consolidation stress $\sigma$ in Fig. 4(b). Despite the difference in compositions of the two backfills, the magnitude of $k_h$ at a given $\sigma$ and the trend in $k_h$ versus $\sigma$ are almost identical. For a given $\sigma$, the greater swelling capacity of the high plasticity bentonite in the sand–bentonite backfill apparently is sufficient to achieve the same hydraulic conductivity as that for the NFC backfill that contains a much higher percentage of low plasticity fines, but a lower percentage of bentonite.

The results in Figs. 4(a and b) are combined in Fig. 4(c) as plots of the logarithm of hydraulic conductivity versus void ratio, or $e - \log k_h$. As suggested by Lambe and Whitman (1969), a semilog linear trend is evident in the $e - \log k_h$ data for both backfills, with a better correlation (i.e., higher coefficient of determination, $r^2$) between $e$ and $\log k_h$ for the NFC backfill than for the sand–bentonite backfill. Also, as shown in Fig. 4(c), a lower void ratio for the NFC backfill is required to achieve the same $k_h$ as that for the sand–bentonite backfill.

**Membrane Efficiency**

The EC values measured in the circulation outflows from the top and bottom boundaries during the chemico-osmotic stage of the tests are shown in Fig. 5. The EC values in the circulation outflow from the top boundaries of the specimens for the NFC and sand–bentonite backfills were relatively constant at approximately 56.4 and 52.3 mS/m, respectively. However, the EC values for the circulation outflows from the bottom of the specimens increased from ~8.5 mS/m at the start of the test for both backfills to ~19 mS/m for the NFC backfill and ~15 mS/m for the sand–bentonite backfill by the end of the test (i.e., 18 days). These measured EC values reflect the imposed boundary conditions and associated solute transport in the tests.

Specifically, the slightly lower EC in the circulation outflow from the top boundary relative to the EC for the 3.88 mM KCl solution of 58.7 mS/m is consistent with loss of solute mass from the source solution due to solute diffusion into the specimen, whereas the increase in EC in the circulation outflows from the bottom boundary is consistent with a gain in solute mass due to diffusion through the specimen. Also, the slight increase in EC in the circulation outflow from the bottom boundary that occurs with each change (decrease) in void ratio is consistent with an increase in diffusive solute mass flux due to an increase in concentration gradient resulting from the decrease in thickness of the specimen (i.e., $\Delta e$ remains constant but the thickness decreases). Finally, the higher final EC of ~19 mS/m for the NFC backfill relative to that of ~15 mS/m for the sand–bentonite backfill suggests that
Fig. 6. Measured induced chemico-osmotic pressure difference for model backfills (e=void ratio; DIW=de-ionized water)

Table 3. Test Results

<table>
<thead>
<tr>
<th>Type of backfill</th>
<th>Bentonite contents (%)</th>
<th>Effective chemico-osmotic pressure difference, $-\Delta P_s$ (kPa)</th>
<th>Chemico-osmotic efficiency coefficient, $\omega$</th>
<th>Consolidation stress, $\sigma$ (kPa)</th>
<th>Hydraulic conductivity, $k_h$ (m/s)</th>
<th>Chemico-osmotic permeability coefficient, $k_p$ (m/s)$^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nelson Farm clay</td>
<td>0.212</td>
<td>0.034</td>
<td>0.018</td>
<td>12</td>
<td>$6.0 \times 10^{-10}$</td>
<td>$1.1 \times 10^{-11}$</td>
</tr>
<tr>
<td></td>
<td>0.052 1.008</td>
<td>0.46</td>
<td>0.024</td>
<td>92</td>
<td>$1.7 \times 10^{-10}$</td>
<td>$4.1 \times 10^{-12}$</td>
</tr>
<tr>
<td></td>
<td>0.377 0.605</td>
<td>0.46</td>
<td>0.024</td>
<td>480</td>
<td>$4.9 \times 10^{-11}$</td>
<td>$1.2 \times 10^{-12}$</td>
</tr>
<tr>
<td>Sand–bentonite</td>
<td>0.720</td>
<td>2.23</td>
<td>0.118</td>
<td>10</td>
<td>$7.2 \times 10^{-10}$</td>
<td>$8.5 \times 10^{-11}$</td>
</tr>
<tr>
<td></td>
<td>0.503 1.012</td>
<td>2.78</td>
<td>0.147</td>
<td>100</td>
<td>$1.5 \times 10^{-10}$</td>
<td>$2.2 \times 10^{-11}$</td>
</tr>
<tr>
<td></td>
<td>0.448 0.812</td>
<td>3.14</td>
<td>0.166</td>
<td>910</td>
<td>$3.1 \times 10^{-11}$</td>
<td>$5.1 \times 10^{-12}$</td>
</tr>
</tbody>
</table>

$^a\omega=\Delta P/\Delta \pi$, where $\Delta \pi$ theoretical maximum chemico-osmotic pressure difference of $-18.9$ kPa corresponding to an initial difference in source KCl concentration of 3.88 mM and a temperature of 20°C.

$^b$Calculated from $k_p=\omega \cdot k_h$. 

in the hydraulic properties of the porous stones at the opposite ends of the specimen (Malusis et al. 2001a; Malusis and Shackelford 2002a). Such differences cause the head loss through the porous stones from the point of circulation inflow to the point of measurement of the pressure difference (i.e., center of the stone) to be different, resulting in $-\Delta P \neq 0$. As a result, an effective chemico-osmotic pressure difference $-\Delta P_s$, representing the difference between the measured $-\Delta P$ for $-\Delta \pi > 0$ and the measured $-\Delta P$ for $-\Delta \pi = 0$, is used in Eq. (1) to calculate $\omega$ (e.g., see Malusis et al. 2001a; Malusis and Shackelford 2002a).

As indicated in Fig. 6, replacement of the DIW with the 3.88 mM KCl solution as the liquid circulated through the top boundary results in an abrupt increase in $-\Delta P$ demonstrating the existence of membrane behavior. The postpeak degradation in $-\Delta P$ is consistent with diffusion of the KCl into the specimen, and subsequent compression of adsorbed layers of cations resulting in partial degradation in the initially observed membrane behavior (Fritz 1986; Malusis and Shackelford 2002a). This degradation in membrane behavior has been observed to increase with an increase in the source salt concentration, such that complete destruction of the membrane behavior eventually may occur if a sufficiently high source salt concentration is used (e.g., see Shackelford and Lee 2003). However, in the present case, the 3.88 mM KCl source solution apparently is sufficiently low such that only partial degradation occurs and residual membrane behavior is maintained, as previously shown by Malusis and Shackelford (2002a). As expected, this trend in $-\Delta P$ with time is repeated each time the specimen was compressed to a new void ratio, since compression results in smaller pores and a corresponding increase in membrane efficiency.

The effective (net) chemico-osmotic pressure differences, $-\Delta P_s$, based on the results shown in Fig. 6, and the corresponding chemico-osmotic efficiency coefficients $\omega$ at steady state are summarized as a function of void ratio in Table 3. The consolidation stress and hydraulic conductivity corresponding to each void ratio based on the results of the consolidation tests shown in Fig. 4(a) and the semilog linear regression equations shown in Fig. 4(c), respectively, are also summarized in Table 3.

Finally, the chemico-osmotic permeabilities $k_p$, defined as the product of $\omega$ and $k_h$ (i.e., $k_p=\omega k_h$; Barbou and Fredlund 1989), are also shown in Table 3. These values for $k_p$ are based on the implicit assumption that the measured values for $k_h$ based on permeation with tap water are not different than those that would result from permeation with the 3.88 mM KCl solution...
Previous results based on permeation of a GCL containing sodium bentonite sandwiched between two geotextiles with a 3.9 mM KCl solution and a processed tap water indicated virtually no difference in the measured hydraulic conductivity values (Malusis and Shackelford 2002a). Thus, although the postpeak partial degradation in membrane behavior is consistent with compression of the adsorbed layer of cations resulting from diffusion of KCl into the specimen, any such compression in the case of permeation with the KCl solution apparently was not sufficient to significantly affect the hydraulic conductivity of the specimens, especially for the relatively low bentonite contents of the backfills evaluated in this study.

Discussion

Correlation of $\omega$ with Stress, Void Ratio, and Hydraulic Conductivity

The measured $\omega$ values at steady state summarized in Table 3 are correlated with the consolidation stress $\sigma$, the void ratio $e$, and the hydraulic conductivity $k_h$, in Fig. 7. In general, for the same range in $\sigma$, $e$, and $k_h$, the $\omega$ values for the sand–bentonite backfill are approximately 600% greater than the $\omega$ values for the NFC backfill. The higher $\omega$ values for the sand–bentonite backfill are consistent with the observation previously noted that membrane behavior tends to be significant in sodium bentonite or soil mixtures containing sodium bentonite, and the difference in $\omega$ values between the two model backfills is consistent with the greater significance of high plasticity clays relative to low plasticity clays in inducing membrane behavior. Therefore, despite the significantly greater content of the fines (silty soils) associated with the NFC backfill (i.e., 89 versus 5%), and the similarity in hydraulic conductivities for the two backfills (see Table 3), the greater amount of sodium bentonite in the sand–bentonite backfill relative to the NFC backfill (i.e., 7.20 versus 2.12%) apparently is sufficient to cause an ~sixfold increase in the membrane efficiency of the sand–bentonite backfill relative to the NFC backfill. This observation also is consistent with the greater significance of the adsorbed layers of cations in governing the pore sizes between clay particles dominated by sodium montmorillonite relative to those dominated by larger, less active clay minerals, such as kaolinite (Mitchell 1993).

As shown in Fig. 7, $\omega$ for both model backfills is semilog linearly correlated with $\sigma$ and $k_h$, and linearly correlated with $e$, with $\omega$ increasing slightly with increase in log $\sigma$ and decrease in $e$ or log $k_h$. In general, the correlation between $\omega$ and $\sigma$, $e$, or $k_h$ is better (i.e., higher $r^2$) for the sand–bentonite backfill than for the NFC backfill, primarily because the membrane efficiency of the NFC backfill remained constant as $e$ decreased from 0.805 to 0.605, corresponding to an increase in $\sigma$ from 92 to 480 kPa, respectively (see Table 3).

Comparison of Results with Literature

The measured $\omega$ values from this study are compared with those from the literature in terms of $\sigma$, $e$, and $k_h$ in Fig. 8. In Fig. 8(a), the results from this study are compared with those from Olsen (1996) and Malusis and Shackelford (2002a): (a) consolidation stress; (b) void ratio; and (c) hydraulic conductivity.
(1969) for kaolin clay in terms of the consolidation stress $\sigma$. In general, $\omega$ for the kaolin clay is more sensitive to $\sigma$ than is $\omega$ for either of the two model backfills. For example, $\omega$ for the kaolin clay is similar to that for the NFC backfill at $\sigma \approx 10$ kPa, and approaches $\omega$ for the sand–bentonite backfill as $\sigma$ approaches $\sim 200$ kPa. For $\sigma$ greater than $\sim 200$ kPa, $\omega$ for the kaolin clay is greater than that for the sand–bentonite backfill. This difference can be attributed to the greater compressibility for the kaolin clay ($C_c \approx 0.523$) relative to that for either the NFC backfill [$C_c = 0.270$; Fig. 4(a)] or the sand–bentonite backfill [$C_c = 0.212$; Fig. 4(a)]. Thus, for a given increment in stress, the change in void ratio for the kaolin clay will be approximately twice that for the model backfills evaluated in this study.

In Fig. 8(b), the results of this study are compared in terms of void ratio $e$, with those from Olsen (1969) for kaolin clay and those from Malusis and Shackelford (2002a) for a GCL. The $\omega$ value is significantly more sensitive to void ratio for the kaolin clay relative to either of the model backfills or the GCL, indicating that void ratio plays a much greater role in determining the value of $\omega$ for the kaolin clay. The greater influence of void ratio on $\omega$ for the kaolin clay likely reflects the lesser role of the adsorbed layer of cations in determining the interparticle pore spaces available for solute migration for clays comprised primarily of kaolinite relative to those comprised primarily of montmorillonite, where physico-chemical interactions resulting from the adsorbed layer of cations play a much more dominant role in determining the microfabric and behavior of the clay (Santamaria et al. 2002).

As shown in Fig. 8(b), $\omega$ values for the GCL are significantly greater than those for either model backfill, and the range in void ratios for the GCL is also greater. The significantly higher $\omega$ and $e$ for the GCL is consistent with the relative sodium bentonite contents, since all of the soil in the GCL is sodium bentonite.

As shown in Fig. 8(c), the higher $\omega$ values for the GCL also are associated with significantly lower (i.e., $\sim 10 \times$) hydraulic conductivity $k_h$ values than those for either model backfill. These lower $k_h$ values for the GCL again can be attributed, in part, to the significantly greater sodium bentonite content in the GCL relative to either model backfill.

**Chemico-Osmotic Permeability Coefficients**

The chemico-osmotic permeability coefficients $k_c$ for both model backfills are shown as a function of the consolidation stress $\sigma$ and void ratio $e$ in Fig. 9. The $k_c$ values are also shown for comparison. As shown in Fig. 9(a), $k_c$ decreases with increasing $\sigma$ for both model backfills, since $k_c$ decreases with increasing $\sigma$ to a much greater extent than $\omega$ increases with increasing $\sigma$. Also, although the $k_c$ values at a given $\sigma$ for both model backfills are almost identical, the $k_c$ values are different due to the difference in $\omega$ at a given stress (see, e.g., Fig. 9(a)). Finally, $k_c$ for the NFC backfill is lower than $k_c$ for the sand–bentonite backfill at a given $\sigma$. This difference is consistent with the expectation of a lower chemico-osmotic liquid flux $q_\sigma$ associated with lower membrane efficiency (i.e., lower $\omega$) such that, in the limit as $\omega$ approaches zero, $q_\sigma$ also approaches zero (i.e., $q_\sigma \to 0$ as $\omega \to 0$).

As shown in Fig. 9(b), the $k_c$ values for the model backfills are reasonably close at a given void ratio, even though $k_c$ for the sand–bentonite backfill is lower than $k_c$ for the NFC backfill at the same given void ratio. This closeness in $k_c$ with void ratio suggests that void ratio controls membrane behavior to a greater extent than does consolidation stress; i.e., regardless of the stress, $k_c$ will be similar if the void ratio is similar. However, given the limited number of backfills evaluated in this study, further results are required before this observation is conclusive.

**Practical Significance**

The results of this study indicate that membrane behavior may be present in SB vertical cutoff walls. The significance of these results can be illustrated with the aid of an example analysis showing the influence of chemico-osmosis due to membrane behavior on the total liquid flux through an SB vertical cutoff wall, as illustrated schematically in Fig. 10. The total liquid flux through a cutoff wall that behaves as a semipermeable membrane $q$ at steady state includes a hydraulic component of liquid flux $q_h$ in response to the difference in hydraulic head, and a chemico-osmotic component of liquid flux $q_\sigma$ in response to a difference in solute concentration (e.g., Barbour and Fredlund 1989; Malusis et al. 2001b) or

$$q = q_h + q_\sigma = k_h\Delta h + k_c\Delta \rho = k_h\left(-\frac{\Delta h}{L}\right) + (\omega k_h)\frac{\Delta \rho}{\gamma_w L}$$

(3)

where $\Delta \rho = $head loss across the wall; $L$=thickness of the wall; $\gamma_w =$unit weight of water (i.e., 9.81 kN/m$^3$ assuming dilute solutions); and $\Delta \sigma =$theoretical difference in chemico-osmotic pressure resulting from the difference in solute concentrations across the barrier. As shown in Fig. 10, $q_\sigma$ is in the opposite direction of $q_h$ when the contaminated ground water accumulates behind the wall such that $q_h$ is directed in the outward (positive $x$) direction, whereas $q_\sigma$ is in the same direction as $q_h$ when the contaminated ground water is the same. $q_\sigma$ is drawn down (e.g., by pumping) such that $q_h$ is directed in the inward (negative $x$) direction. Also, note that $q \to 0$ as $\omega \to 0$ such that Eq. (3) reduces to Darcy’s law in the limit when $\omega = 0$. 

![Fig. 9. Chemico-osmotic permeability coefficients and hydraulic conductivity values for two model backfills as function of: (a) consolidation stress and (b) void ratio](image-url)
For this example analysis, both the thickness of the wall (L) and the head loss across the wall are conveniently assumed to be 1 m resulting in a unit hydraulic gradient directed either outward (i.e., \( \Delta h = -1 \) m, \( i_0 = 1 \)) or inward (i.e., \( \Delta h = 1 \) m, \( i_0 = -1 \)). Values for \( k_h \) and \( \omega \) are based on the results of this study for the two model backfills (Table 3). Finally, in order to be consistent with the conditions for which the \( \omega \) values were measured, \( \Delta \pi \) is taken as \(-18.9 \) kPa corresponding to a 3.88 mM KCl concentration difference across the wall. This value for \( \Delta \pi \) results in a chemico-osmotic gradient, \( i_\pi \), for a 1 m thick wall of \(-1.93 \).

The results of the example analysis are shown in Fig. 11 in the form of the ratio of \( q \) to \( q_{h0} \), or \( q / q_{h0} \), as a function of consolidation stress \( \sigma \), and the depth within the wall based on a saturated unit weight of 17.5 kN/m\(^3\) for the backfill. In the absence of membrane behavior, \( \omega \) would be zero such that \( q / q_{h0} = 100\% \). However, as shown in Fig. 11, the chemico-osmotic liquid flux results in a reduction of the total liquid flux through the wall (i.e., \( q / q_{h0} < 100\% \)) in the case of the outward hydraulic gradient scenario, and contributes to the inward hydraulic liquid flux (i.e., \( q / q_{h0} > 100\% \)) in the case of the inward hydraulic gradient scenario. This reduction or contribution tends to increase with increasing \( \sigma \), because \( \omega \) tends to increase with increasing \( \sigma \), and is greater in the case of the sand–bentonite backfill–slurry mixture, because both \( \omega \) and \( k_h \) are greater at a given \( \sigma \) for the sand–bentonite backfill relative to the NFC backfill [see Figs. 7(a) and 10(a)].

For example, at the highest stress of 910 kPa for the sand–bentonite backfill, the total liquid flux through the wall is only about 68% of the liquid flux that would be predicted neglecting membrane behavior in the case of a unit outward hydraulic gradient. In the case of a unit inward hydraulic gradient, the total liquid flux through the wall is about 132% of the liquid flux that would be predicted neglecting membrane behavior. Thus, in either case, the existence of membrane behavior in the wall enhances the containment efficiency of the wall by enhancing the resistance to outward directed flux of contaminated ground water. Also, as noted by Shackelford et al. (2001), another beneficial aspect of membrane behavior for low permeability containment barriers in which diffusion dominates contaminant transport results from the reduction in diffusive mass flux due to solute restriction.

The effect of chemico-osmotic liquid flux also can have other important ramifications. For example, the reduction in liquid flux that results when an outward hydraulic gradient is applied across the wall also can affect the field measurement of \( k_h \) for vertical cutoff walls (e.g., when a well placed just outside the wall is pumped), as neglecting the existence of membrane behavior could result in an underestimation of \( k_h \) in the case where the wall exhibits membrane behavior.

The practical extension of the results of this example analysis may be limited. For example, arching of placed backfill with rigid sidewalls can cause shear stresses at the trench walls resulting in consolidation stresses in SB vertical cutoff walls that typically are lower than geostatic stresses (Evans et al. 1995; Filz 1996; Filz et al. 1999). As a result, arching would cause a reduction in the membrane efficiency of the wall. Also, the backfills tested in this study were permeated with DIW to flush soluble salts from the pore water thereby enhancing the likelihood of membrane behavior. Although a similar flushing process could be induced in the field, e.g., by placing the wall within “clean” ground water ahead of a migrating contaminant plume, the presence of high soluble salts concentrations in the pore water of the backfill may reduce or eliminate the membrane efficiency of the wall.

On the other hand, the results of this analysis do not include the potentially beneficial aspects of membrane behavior associated with the formation of the filter cake on the sides of the trench. Also, as previously discussed, the \( \omega \) values reported in this study likely are somewhat lower than the true \( \omega \) values (by \(~6\%) due to the use of the source solute concentrations to calculate \( \Delta \pi \) in accordance with Eq. (2) (i.e., relative to the use of the average solute concentrations across the top and bottom boundaries).

**Summary and Conclusions**

Two model backfills were evaluated for their ability to behave as semipermeable membranes. The base soils for the model backfills consisted of a natural clay with 89% (dry weight) fines and a mixture of sand with 5% dry sodium bentonite. Specimens of both backfills were mixed with a sufficient amount of 5% sodium bentonite–water slurry to correspond to 100 mm slumps in accordance with standard practice for SB vertical cutoff walls. The final bentonite contents were 2.12% for the natural clay backfill and 7.20% for the sand–bentonite backfill.

The results indicate that both backfills act as semipermeable membranes, with chemico-osmotic efficiency coefficients \( \omega \) ranging from 0.018 to 0.024 for the natural clay backfill and from 0.118 to 0.166 for sand–bentonite backfill. The difference in the
range of \( \omega \) values is attributed to the significantly higher amount of high-swelling bentonite in the sand–bentonite backfill (i.e., 7.20 versus 2.12\%). For each backfill, the higher \( \omega \) values are correlated with higher consolidation stress, lower void ratio, and lower hydraulic conductivity. The results of this study provide the first quantitative indication that SB vertical cutoff walls may act as semipermeable membranes.

The measured results of this study were used in an example analysis to illustrate the potential significance of membrane behavior in a 1 m thick SB vertical cutoff wall. The results of the analysis show that chemico-osmotic liquid flux due to membrane behavior can reduce the total liquid flux through an SB vertical cutoff wall in the case of a unit outward hydraulic gradient to as low as 68\% of that which would occur in the absence of membrane behavior. In the case of a unit inward hydraulic gradient, the results of the analysis also show that chemico-osmotic liquid flux can contribute as much as 32\% of the hydraulic liquid flux to the total liquid flux through the wall. Therefore, the existence of membrane behavior in an SB vertical cutoff wall would potentially enhance the performance of the wall.

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References


