# Membrane behavior of an exhumed geosynthetic clay liner – preliminary analysis

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## ABSTRACT

Membrane behavior of a geosynthetic clay liner (GCL) exhumed from a bottom liner system at a municipal solid waste landfill in California, USA was investigated in this study. The GCL was installed as part of a geomembrane-GCL bottom liner system at a newly constructed cell at the landfill. The cell was not filled subsequent to the construction of the liner system and the geosynthetics were exposed to the atmosphere for 12 years. Preliminary data on the membrane behavior of GCL samples exhumed from the liner system are provided herein. Laboratory multi-stage membrane behavior tests were used to determine membrane efficiency coefficients ($\omega$) of the GCL for potassium chloride source solutions. Measurable membrane behavior was confirmed in the laboratory tests for the exposed GCLs. Even though membrane behavior was shown to still exist in the GCL after 12 years of exposure, values of $\omega$ were very low (0.1 – 5.4 %). Bentonite migration and cation exchange likely contributed to the significantly lower $\omega$ for the exposed GCLs relative to values reported in the literature for virgin/unexposed GCLs.

**Keywords:** landfill, GCL, exhumed, membrane behavior, municipal solid waste

## 1   INTRODUCTION

Geosynthetic clay liners (GCLs) are manufactured barrier materials that typically consist of a layer of bentonite (typically sodium bentonite) encased between two geotextiles (Yesiller and Shackelford, 2011; Koerner 2012). The main advantages of GCLs in containment applications are low thickness, very low hydraulic conductivity, reproducible material properties, high unit attenuation capacity, and relative ease of installation/repair. The main disadvantages of GCLs include changes to bentonite chemistry, seam separation, low overall attenuation capacity, and lack of knowledge on long-term integrity (e.g., NRC, 2007).

Use of composite liner systems consisting of geomembranes placed over GCLs is commonplace for containment barrier applications. Timely cover of the geosynthetics is critical for integrity of the liner systems and performance of the geosynthetic materials. Field investigations and laboratory analyses indicated three main issues, seam separation, cation exchange (i.e., replacement of monovalent cations with divalent cations), and bentonite migration, for GCLs under exposed conditions. Separation of GCL panels underneath exposed geomembrane liners have been reported in multiple field investigations, where panel separations of 50 to 1200 mm occurred (5 to 28% strain) for uncovered liner systems exposed for durations between 2 and 60 months (Koerner and Koerner, 2005; Gassner, 2009; Thiel and Rowe, 2010). While no direct data is available for the bentonite chemistry and associated behavior of fully exposed GCLs from composite liners, GCLs exhumed from composite cover systems (with overlying vegetative/protective layers) after 3 to 7-year service lives in landfills indicated that cation exchange occurred in the exhumed GCLs (Scalia and Benson, 2011). Cation exchange with the underlying soils was higher for the cases with higher water content of soils underlying the installed GCLs. Decreases in swell index and increases in the hydraulic conductivity of the GCLs also were observed in some cases. In a test plot, migration of bentonite and erosion within the GCL occurred with length from the top to the toe of a slope for a GCL underlying a geomembrane under exposed conditions for over 3 years (Take et al., 2015).

In laboratory studies, GCLs have been shown to exhibit membrane behavior, potentially enhancing the long-term performance of the barrier system (e.g., Malusis and Shackelford, 2002; Shackelford et al., 2016). However, experimental research to date has only evaluated membrane behavior for virgin GCL samples that have not been exposed to field conditions. Values of membrane efficiencies reported for virgin GCLs tested under laboratory conditions likely are not representative of GCLs in containment applications after long-term

exposure. Processes that can adversely affect the integrity and performance of GCLs in the field (e.g., cation exchange with subsoil, chemical incompatibility with leachate, downslope bentonite migration) may also result in reduction or destruction of membrane behavior. The persistence and relevance of membrane behavior in GCLs after long-term field exposure remains largely unknown. Therefore, as part of this unique experimental study, samples of GCL exhumed from a liner system after 12-years of field exposure were evaluated for membrane behavior. Samples from both the top and bottom of the liner slope were obtained and tested. Impacts of cation exchange and bentonite migration on membrane efficiencies also were assessed.

## 2 BACKGROUND

### 2.1 Membrane behavior in GCLs

Membrane behavior is the ability of clays to behave as semipermeable membranes, selectively restricting aqueous miscible chemical species from entering the pores. This phenomenon, also referred to as anion exclusion in the soil sciences, exists when two adjacent clay particles are sufficiently close such that the electric fields associated with the particles overlap and result in electrostatic repulsion of charged inorganic solutes (e.g., Fritz, 1986; Shackelford et al., 2003). The existence of membrane behavior, or the process of chemical restriction, also may result in chemico-osmosis, whereby liquid counterflows from lower solute concentration (higher water activity) to higher solute concentration (Shackelford et al., 2003). For clay barriers used in containment systems, the existence of membrane behavior can benefit the barrier performance by reducing the total contaminant flux into the environment.

Membrane behavior is quantified by the membrane efficiency coefficient, ω, which represents the relative magnitude of chemical restriction of the material. Values of ω range from 0 to 1, with 0 representing no solute restriction (i.e., no membrane behavior) and unity representing complete chemical restriction (i.e., perfect semipermeable membrane behavior) (e.g., Mitchell, 1991). Typically, ω is dependent on the type and amount of clay minerals in the soil, the pore sizes, and the type and concentrations of ionic species in the porewater (Kemper and Rollins, 1966; Bresler, 1973; Olsen et al., 1990; Mitchell, 1991; Malusis et al., 2001).

Previous studies have demonstrated the existence of semipermeable membrane behavior in GCLs (e.g., Malusis and Shackelford, 2002; Kang and Shackelford, 2011; Shackelford et al., 2016). Closed-system apparatuses, such as described in Malusis and Shackelford (2001), often are used to measure ω for GCLs and other bentonite-based specimens. In the closed-system approach (Fig. 1), a solution with higher concentration typically is circulated across the top boundary of the specimen while a solution of lower concentration is circulated across the bottom boundary,

creating a concentration gradient across the specimen. If the specimen exhibits membrane behavior, chemico-osmotic flow will try to occur from the lower concentration boundary to the higher concentration boundary. However, the closed-system apparatus prohibits chemico-osmotic flow from occurring, and as a result a hydraulic pressure difference ($\Delta P$) develops across the specimen.
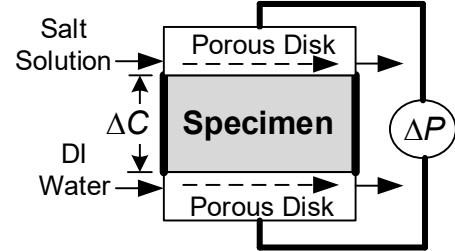


Fig. 1. Conceptual depiction of cell used in a closed-system apparatus to measure membrane behavior of a GCL specimen.

Values of $\Delta P$ measured in response to the applied concentration gradient are used to quantify ω, in accordance with Equation 1 (Groenevelt and Elrick, 1976; Malusis et al., 2001):

$$\omega = \Delta P / \Delta \pi \qquad (1)$$

where $\Delta \pi$ is the osmotic pressure difference, which can be regarded as the theoretical maximum value of $\Delta P$ that would result across an ideal semipermeable membrane subjected to a particular solute concentration difference ($\Delta C$) across the specimen. The value of $\Delta \pi$ is determined using the van't Hoff equation (e.g., Malusis et al., 2001):

$$\Delta \pi = \nu R T \Delta C \qquad (2)$$

where ν is the number of ions per molecule of the salt (e.g., 2 for KCl), R is the universal gas constant (8.314 J/mol·K), T is the absolute temperature (assumed to be 293 K for room temperature), and $\Delta C$ is the difference in solute concentration across the specimen (in M).

Although results of prior research demonstrate GCLs can exhibit substantial membrane behavior, with ω as high as 70% (e.g., Malusis and Shackelford, 2002), critical knowledge gaps remain. Specifically, membrane behavior studies to date have only been performed on virgin GCLs in a laboratory setting. Experimental evaluation of the significance and persistence of membrane behavior in GCLs after field exposure has not been conducted. Thus, the relevance of membrane behavior to the long-term containment performance of barrier systems with GCLs is not understood.

### 2.2 Field site

GCL samples were obtained from a geomembrane-GCL composite bottom liner system that was installed at a municipal solid waste landfill in San Luis Obispo, California (USA) in a temperate climate. The geomembrane consisted of a 1.5 mm-thick black HDPE

material and the GCL consisted of a needle-punched nonwoven-nonwoven product with sodium bentonite. The liner system was constructed in 2004 and the landfill cell remained unfilled for 12 years, until 2016 (Fig. 2), when the liner system was removed along the south and east slopes for expansion of the cell. The cell had 2H:1V slopes and the length of the slopes ranged from approximately 24 m (south slope) to 30 m (east slope).

Hanson and Yesiller (2017; 2019) reported field conditions (prior to laboratory testing) during the exhumation. The observations for the GCL included:
(1) GCL seams along both slopes had separated, at a total of eight locations along 7 out of 43 seams with gaps 20-220 mm in width and 1.7 to 17 m in length;
(2) the GCL near the top was relatively dry for both slopes, whereas the GCL was wet, in particular along the bottom of the east slope;
(3) bentonite migration occurred along both slopes, with significant accumulation at the bottom of the east slope;
(4) loss of bentonite along upper parts of the slope as well as along the bottom of the south slope near the sump in the cell with low to essentially no bentonite remaining between the geotextile sheets.



Fig. 2. Exhumed liner system.

# 3 MATERIALS AND METHODS

## 3.1 Exhumed GCL samples

The GCL originally installed at the landfill was a Bentomat DN product manufactured by CETCO. The water content, swell index, bentonite mass per unit area, and hydraulic conductivity were determined using ASTM D2216, ASTM D5890, ASTM D5993, and ASTM D5887 test methods, respectively. The average properties of the GCL reported by the manufacturer were: water content of 9.1%, swell index of 26.0 mL/2g, bentonite mass per unit area of 4.3 kg/m$^2$, and hydraulic conductivity of $5 \times 10^{-11}$ m/s (maximum). The properties of the GCL determined in conformance testing were: water content of 26.0%, bentonite mass per unit area of 4.9 kg/m$^2$, and hydraulic conductivity of $2.8 \times 10^{-11}$ m/s (under a consolidation pressure of 69 kPa and using a gradient range of 87-119). Water content of the GCL rolls in the field was determined to be 20.3%.

GCL samples used for membrane behavior analysis were obtained from a long strip sample (27 m long parallel to slope, 0.3 m wide) exhumed along the corner of the east and south slopes. In particular, sub-samples obtained from the strip near the top and bottom of the slope were used. The strip sample was cut from the GCL panel using a utility knife. The sample was rolled with the upper side placed inside the roll and immediately wrapped in plastic and placed in air-tight bags following exhumation, and transported to the laboratory. The square-shaped sub-samples (300 mm x 300 mm) were removed using a precision knife for membrane behavior analysis. A small amount of water was sprayed along the perimeter of the target location for the removal of the sub-samples to prevent loss of bentonite along the cut edges of the sample. The amount of added water was monitored and accounted for in the determination of the mass of the GCL sample and water content of the GCL.

The water contents of the GCL samples were 16 and 55% at the top and the bottom of the slope, respectively. The swell index was 25.5 mL/2g for the top sample and 9.5 mL/2g for the bottom sample. Bentonite mass per unit areas of 3.9 and 4.9 kg/m$^2$ were measured for the top and bottom samples, respectively. The hydraulic conductivities ($k$) of the top (average GCL thickness of 6.0 mm) and bottom samples (average GCL thickness of 11.7 mm) were $7.7 \times 10^{-7}$ and $1.2 \times 10^{-8}$ m/s, respectively. The top of slope sample was relatively dry with a swell index slightly lower and mass/area lower than the originally installed GCL. The bottom of slope sample was relatively wet with a swell index lower than and mass/area similar to the installed GCL. The hydraulic conductivities were several orders of magnitude higher than the values determined in manufacturer's and conformance tests, which may have been related to the low mass/area for the top sample and low swell in the bottom sample.

## 3.2 Chemical Solutions

Potassium chloride (KCl) solutions were used to allow for comparison of the results with literature data for similar tests on virgin GCLs. The liquids included de-ionized water (DIW) and KCl solutions (certified A.C.S.) with target KCl concentrations of 5, 10, 20, and 50 mM. This concentration range is consistent with solution ranges used in the membrane behavior literature for GCLs (see Shackelford 2013). The electrical conductivity ($EC$), pH, and temperature of the solutions were measured with a pH/Conductivity meter with probes for: pH and EC. The concentration of Cl$^-$ was confirmed using the same meter with a Cl$^-$ ionic selective electrode. The Cl$^-$ concentrations of randomly selected solutions were also confirmed with a discrete nutrient analyzer with photospectrometer.

## 3.3 Membrane Behavior Testing

Two multi-stage membrane behavior tests were performed to evaluate the membrane efficiencies of the GCL specimens over a range of KCl concentrations. The testing apparatus and procedure were the same as that

described in Malusis et al. (2001) and Shackelford et al. (2016). The apparatus included: (1) a hydraulic control system with an infusion/withdraw double syringe flow pump to circulate solutions; (2) an acrylic 71-mm-diameter rigid-wall cell as specimen holder; pressure transducers to monitor pressures at the top and bottom boundaries of the specimen; and (3) stainless steel tubing and connections throughout the system to prevent corrosion or volume change. Since the hydraulic control system forces all inflow and outflow rates to be identical, flow across the specimen or volume change cannot occur (i.e., the setup is a closed system). Thus, the volume of the GCL specimens does not change during the multi-stage test. Additional details regarding the apparatus and test conditions are provided in Malusis et al. (2001).

In preparation for testing, the exhumed GCL samples were cut to diameters of approximately 71 mm using a razor blade. To avoid loss of bentonite particles during cutting, a few drops of de-ionized water were added to wet the perimeter during cutting. The mass of the added water was measured and accounted for in the calculation of the initial water content of the GCL specimens. After the specimens were cut, the dimensions (i.e., diameter and length) and weight of the specimens were re-measured. The initial thicknesses of the top and bottom GCL specimens were 6.8 mm and 11.8 mm, respectively. The GCL specimen was sandwiched by two layers of porous disks, inside the acrylic rigid-wall cell (Fig. 1). The top and bottom pedestals were locked in place immediately after assembly.

After the test cell was assembled, a DIW permeation stage was conducted to saturate the specimens. The cumulative permeation times and pore volumes of flow (PVF) for the top and bottom GCL specimens were 11.4 and 28.4 days, and 8.2 and 12.7 PVF, respectively. The final $EC$ of the effluent from the top and bottom GCLs was 27.8 mS/m and 9.9 mS/m, respectively. A DIW circulation stage (i.e., circulation of DIW across the boundaries, and not through the specimen) then was conducted to flush excess salts and establish a stabilized, baseline $\Delta P$ when $\Delta C = 0$ ($\Delta P_o$). Ideally, $\Delta P_o$ should be near zero as no differential chemical concentration should exist across the specimen. However, very low values of $\Delta P_o$ often are observed in membrane behavior testing, partially due to slight differences in the hydraulic resistance of the top and bottom porous disks (e.g., Malusis et al., 2001; Malusis and Shackelford, 2002; Shackelford et al., 2016). The measured $\Delta P_o$ values for the top of slope and bottom of slope specimens were 0.299 kPa and -0.234 kPa, respectively.

After completion of baseline testing to establish $\Delta P_o$ values, the membrane behavior test stages were performed with fresh KCl solutions and DIW circulating across the porous disks at the top and the bottom boundaries of the specimen, respectively. The start of the first salt solution stage is designated as test time ($t$) time "0", such that times during the baseline DIW circulation

stage are t < 0. Throughout testing, inflow and outflow concentrations at each boundary of the specimen were recorded. Due to diffusion of solutes from the top to bottom boundary of the specimen, concentrations in the top outflow ($C_{ot}$) were lower than that in the top inflow ($C_t$). Accordingly, concentrations in the bottom outflow ($C_{ob}$) were higher than that in the bottom inflow ($C_b$). The average boundary concentrations, i.e., $C_{t\_ave} = (C_{ot} + C_t)/2$ and $C_{b\_ave} = (C_{ob} + C_b)/2$, were used to calculate the concentration difference across the specimen in Equation 2. The average solute concentration in the entire specimen, $C_{ave}$, was the average of the average boundary concentrations: $C_{ave} = (C_{t\_ave} + C_{b\_ave})/2$.

Termination criteria for the DIW and KCl stages included achieving steady-state effluent concentrations and $\Delta P$. After the tests were terminated, the dimensions and mass per unit area of the specimen were remeasured.

## 4   RESULTS AND DISCUSSION

The measured $C_t$ and $C_b$ for the top and bottom specimens over the test durations are presented in Figs. 3a and 3b, respectively. During the DIW circulation stage (i.e., the baseline stage to establish $\Delta P_o$), soluble salts initially existing in the specimen diffused outward. At the start of each new test stage when the salt solution at the top boundary was increased (e.g., 5 mM), there was an immediate increase in the salt concentration at the top outflow, and a slower increase in concentration in the bottom outflow due to increased solute diffusion across the specimen. The trends in Fig. 3 are consistent with trends in the literature for virgin GCL tests.
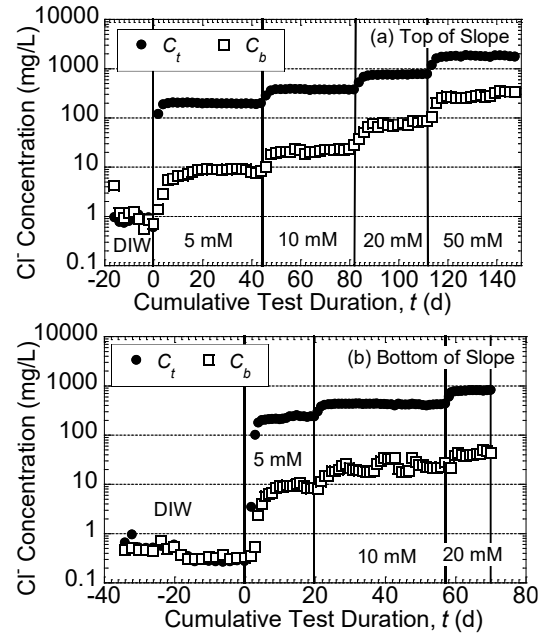


Fig. 3. Chloride concentration in the effluent from the top ($C_t$) and bottom boundary ($C_b$) with cumulative test duration.

The effective differential pressures, $\Delta P_e$ ($\Delta P_e = \Delta P - \Delta P_o$), for the top of slope and bottom of slope specimens

are in Table 1 and Figs. 4a,b. As the KCl concentrations were increased, the measured $\Delta P_e$ remained relatively low (i.e., < 3.3 kPa) for the top specimen. The $\Delta P_e$ values for the bottom specimen were also relatively low (i.e., < 10 kPa) except some scattered data during the 10 mM KCl stage. The scatter in the pressures during the 10 mM stage in Fig. 4b are attributed to malfunction of the flow pump, as well as potential microbial activity.

Table 1. Summary of results from membrane behavior testing.

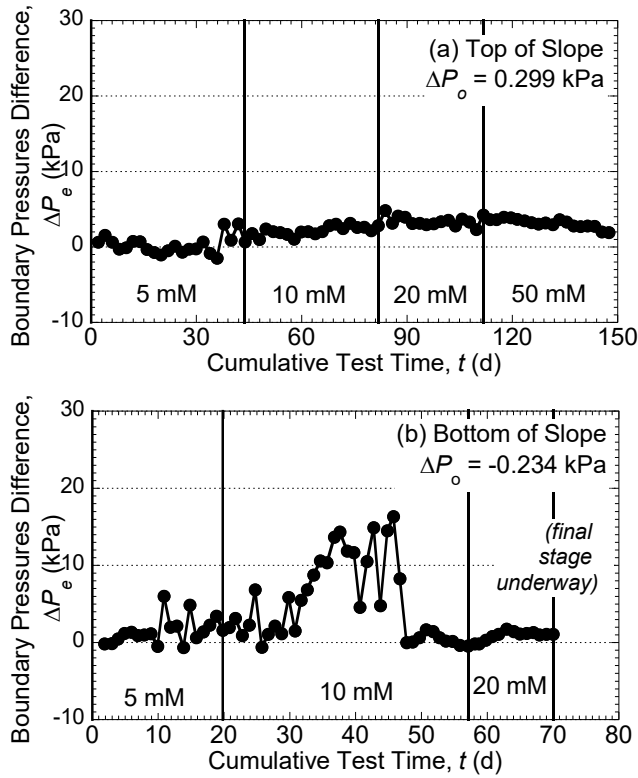| GCL Sample | $C_o$ (mM) | Final $\Delta P_e$ (kPa) | $\omega$ (%) |
|---|---|---|---|
| Top | 5 | 0.87 | 2.98 |
| | 10 | 2.65 | 5.43 |
| | 20 | 3.25 | 3.65 |
| | 50 | 2.88 | 1.97 |
| Bottom | 5 | 1.48 | 5.26 |
| | 10 | 0.04 | 0.07 |
| | 20 | 1.05 | 0.91 |





Fig. 4. Measured effective differential pressures ($\Delta P_e$) during each membrane behavior test stage for the (a) top of slope and (b) bottom of slope specimens.

Values of $\omega$ for the top of slope and bottom of slope specimens were calculated based on the final $\Delta P_e$ values for each stage. As shown in Fig. 5, the GCL specimens from the top and bottom of the slope still exhibited measurable membrane behavior after over a decade of field exposure. However, the values of $\omega$ for both specimens were very low (< 6 %) as demonstrated in the figure.
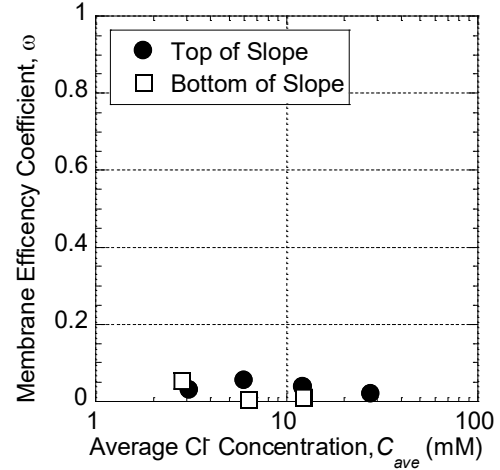


Fig. 5. Membrane efficiency coefficients ($\omega$) as a function of average concentration in the pore-water of the specimens ($C_{ave}$).

Values of $\omega$ for the top GCL decreased from 2.98 % to 1.97 % as $C_{ave}$ increased from 3.1 to 27.7 mM. For the bottom specimen, $\omega$ values decreased from 5.26 % to only 0.07 % as $C_{ave}$ increased from 2.85 to 6.41 mM. It should be noted that for the 5 mM test stage for the top GCL and the 10 mM test stage for the bottom GCL, scatter in the measured $\Delta P$ may have resulted in less reliable assessment of $\omega$ relative to the other concentration stages. This trend of decreasing $\omega$ with increasing $C_{ave}$ is consistent with trends reported in the literature for membrane behavior of virgin GCLs exposed to KCl solutions (e.g., Malusis and Shackelford, 2002; Kang and Shackelford, 2011; Shackelford et al., 2016). Decreasing $\omega$ with increasing source salt concentration has been attributed to compression of diffuse double layers with increasing pore-water concentrations due to diffusion of KCl from the upper boundary into the accessible pores (Fritz 1986; Shackelford et al. 2003).

All of the $\omega$ values for the exposed GCLs were significantly lower than the range of $\omega$ values that have been reported in the literature for virgin GCLs, for the same $C_{ave}$. For example, Malusis and Shackelford (2002) performed membrane behavior tests on virgin Na-bentonite GCLs using the same test method and similar range of KCl concentrations (3.9, 8.7, 20, and 47 mM) as used in this study. At the lowest concentration ($C_{ave}$ ~ 2 mM), Malusis and Shackelford (2002) reported $\omega$ for the virgin GCL was 63 %. This is more than an order of magnitude higher than the $\omega$ values measured for the exposed GCLs from the top and bottom of the slope (2.98 % and 5.26 %, respectively) at similar $C_{ave}$.

The low membrane efficiencies of the exhumed GCLs were in line with the observed variations (i.e., increases) in the hydraulic behavior of the exhumed GCLs. The low membrane efficiency of the specimens from the top of the slope may have resulted from bentonite migration downslope. The final dry bentonite content of the top of slope specimens was only 3.6 kg/m$^2$

(27% lower than the original values from conformance testing). The differences observed in hydraulic and also swelling behavior of the GCL for the bottom of the slope locations were in line with the low measured membrane efficiency, all potentially resulting from exchange of the sodium in the bentonite. Further testing is underway to determine the membrane efficiency of the virgin GCL as well as cation exchange properties of the exhumed GCLs to provide further insight into the observed behavior.

## 5 CONCLUSIONS

The study presented herein represents the only known experimental analysis to quantify membrane behavior of exhumed GCLs after field exposure. Laboratory multi-stage membrane behavior tests were performed on GCL samples that were exhumed from the top and bottom of the slope of a landfill bottom liner, 12 years after the initial installation. The GCLs exhibited measurable membrane behavior that generally decreased with increasing $C_{ave}$ when tested with KCl solutions. However, the values of $\omega$ were very low (0.1 – 5.4 %) relative to values reported in the literature for virgin GCLs, likely due to bentonite migration and cation exchange that occurred in the field.

The results of this preliminary study suggest: (1) measurable membrane behavior can persist in GCLs even after a decade of field exposure; and (2) predictions of impacts of membrane behavior on containment performance based on data for virgin GCLs may be inaccurate due to cation exchange and bentonite migration that can occur in the field. Testing is currently underway for virgin samples of the same GCL that have not been exposed to field conditions, to allow for more direct assessment of the effects of field exposure on membrane behavior.

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