



Limiting membrane and diffusion behavior of a geosynthetic clay liner



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ABSTRACT

Bentonite based geosynthetic clay liners (GCLs) are commonly used as chemical containment barriers to minimize liquid flow and contaminant transport. The preference for GCLs relative to other materials for chemical containment include economy of use and superior engineering properties, including low hydraulic conductivity (k) and the ability to exhibit semipermeable membrane behavior that restricts the migration of solutes through the bentonite. In the case of low k (e.g., $\leq 10^{-10}$ m/s), diffusion is likely to be a significant, if not dominant, mechanism of chemical transport. The objective of this study was to evaluate the membrane and diffusion behavior of a GCL in the limit as the salt (KCl) concentration increased to the extent that any observed membrane behavior was destroyed. The results indicated that the observed membrane behavior for the GCL was essentially destroyed when the average KCl concentration across the GCL specimen, C_{ave} , reached 200 mM KCl, which was greater than the value of C_{ave} that would have been predicted on the basis of previous results. The results also indicated that the value of the effective diffusion coefficient, D^* , for Cl^- at this limiting value for C_{ave} of 200 mM KCl was greater than the value of D^* that would have been predicted on the basis of the use of lower values of C_{ave} . However, the differences between the measured and predicted values of D^* were relatively minor ($\leq 11.3\%$), such that the differences likely would be insignificant for most practical applications.

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1. Introduction

Bentonite based geosynthetic clay liners (GCLs) have been used extensively as hydraulic and chemical barriers for a wide variety of containment applications, including liners and covers for solid municipal and hazardous waste landfills, surface impoundments (e.g., lakes and ponds, aeration lagoons, fly ash lagoons, mine tailings impoundments, etc.), canals, storage tanks, and secondary containment of above-grade fuel storage tanks (e.g., Bouazza, 2002; Rowe, 2006, 2007, 2014). The preference for GCLs relative to other materials for such containment applications stems primarily from an economy of use and the possibility of a low hydraulic conductivity, k (e.g., $\leq 10^{-10}$ m/s), although the potential for incompatibility between the bentonite comprising the GCL and the chemicals to be contained can result in significant increases in k , i.e., $\Delta k \gg 0$ (e.g., Shackelford et al., 2000; Jo et al., 2001, 2005; Kolstad et al., 2004; Lee and Shackelford, 2005a, b; Lee et al.,

2005; Shackelford et al., 2010; Ishimori and Katsumi, 2012; Rosin-Paumier and Touze-Foltz, 2012; Liu et al., 2013, 2014, 2015; Mazzieri et al., 2013; Bouazza and Gates, 2014; Makusa et al., 2014). In cases where significant increases in k are deemed likely, consideration has been given to the potential use of chemically modified bentonites in GCLs for the primary purpose of mitigating any such incompatibility in order to maintain a low k (e.g., Onikata et al., 1999; Katsumi et al., 2008; Di Emidio, 2010; Mazzieri et al., 2010; Scalia et al., 2011, 2014; Bohnhoff, 2012; Scalia, 2012; Bohnhoff et al., 2013; Bohnhoff and Shackelford, 2014; Di Emidio et al., 2015; Mazzieri and Di Emidio, 2015). However, even when GCLs are able to maintain a low k upon exposure to chemical solutions, diffusion is likely to be a significant, if not dominant, mechanism of chemical transport through the GCLs due to the low k and the relative thinness of GCLs (typically ≤ 15 mm) (e.g., Shackelford, 2013a, b). As a result, a significant number of studies have focused on the transport, in general, and diffusion, in particular, of chemicals through GCLs both individually and as components of composite barriers (e.g., Lake and Rowe, 2000; Malusis and Shackelford, 2002a; El-Zein et al., 2012; Mendez et al., 2013; Shackelford, 2013a, b; Xie et al., 2013; Hosney and Rowe, 2014; Chen et al., 2015).

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In addition to economy of use and the potential for low k , GCLs have been shown to exhibit semipermeable membrane behavior, which refers to the ability of clays to restrict the migration of dissolved chemical species or solutes (e.g., Malusis and Shackelford, 2002b; Kang and Shackelford, 2011; Malusis et al., 2015; Malusis and Daniyarov, 2016). Solute restriction is a function of the sizes of the clay pores, with smaller pore sizes resulting in greater solute restriction. In bentonites, solute restriction is attributed to electrostatic repulsion of the solutes due to interaction of the electric fields associated with the diffuse double layers (DDLs) of adjacent clay particles in close proximity (Fritz, 1986). Because the sizes of the pores in clays vary, the extent of solute restriction in clays that behave as membranes also varies, such that some pores restrict solute migration while others do not. However, the mass flux resulting from solute migration through clay membranes is reduced relative to that in the absence of solute restriction. As a result, the chemical containment function of a GCL can be enhanced by the existence of membrane behavior (Malusis et al., 2003; Shackelford et al., 2003; Shackelford, 2013a, b; Malusis and Daniyarov, 2016).

The ability of GCLs to restrict solute migration also is known to decrease as the salt concentration within the pore water of the bentonite contained within the GCL increases (Malusis and Shackelford, 2002b; Shackelford et al., 2003; Shackelford, 2011, 2013a, b; Malusis and Daniyarov, 2016). This trend of decreasing solute restriction with increasing solute concentration occurs because, as the concentration of the solutes in the pore water increases, the thicknesses of the DDLs and associated electric fields controlling solute restriction decrease, resulting in progressively larger pores and correspondingly lesser solute restriction (Fritz, 1986). In this regard, knowledge of the solute concentration at which solute restriction (i.e., membrane behavior) ceases to exist, commonly referred to as the threshold concentration (Shackelford et al., 2003), is important from the viewpoint of knowing whether or not the beneficial aspects of membrane behavior will be prevalent.

Unfortunately, very few experimental studies have been conducted using solute concentrations that were sufficiently high so as to result in complete destruction of membrane behavior, such that the threshold concentrations typically have been determined by extrapolation. However, such extrapolations are suspect, as the trend in solute restriction (membrane behavior) versus logarithm of the average solute concentration has been shown to become non-linear in the limit as the solute concentration approaches the threshold concentration (e.g., Kemper and Rollins, 1966; Dominijanni et al., 2013).

For example, Kemper and Rollins (1966) measured osmotic (membrane) efficiency coefficients, designated as σ , for specimens of a sodium bentonite at several different volumetric water contents, θ (0.80, 0.84, 0.91), exposed to several different salts (NaCl, CaCl₂, Na₂SO₄, CaSO₄) over a range of average salt concentrations, C_{ave} , across the specimens. In general, values of σ range from zero for no solute restriction (no membrane behavior) to unity for complete or 100% solute restriction corresponding to ideal membrane behavior. Kemper and Rollins (1966) established that σ decreased as C_{ave} increased for any given salt and value of θ . More importantly, they performed their tests using a wide range of salt concentrations (e.g., $0.0006 \text{ N} \leq C_{ave} \leq 0.2 \text{ N}$), which allowed them to measure values of σ that were either very close to or at zero. However, their study did not include any results pertaining to diffusion of the salts through the bentonite.

Shackelford and Lee (2003) measured the membrane and diffusion behaviors of a specimen of a GCL bounded by a 5 mM calcium chloride (CaCl₂) solution on one side and de-ionized water (DIW) on the other side. They observed an initial development of a

chemico-osmotic pressure difference, ΔP , of 19.3 kPa, which is directly proportional to the membrane efficiency coefficient, suggesting the existence of significant membrane behavior in the GCL. However, the magnitude of ΔP subsequently decreased to zero as the test progressed. The decrease in ΔP was attributed to reduction in the solute restriction resulting from continual diffusion of Ca²⁺ into the pores of the bentonite, which led to the progressive collapse of the DDLs surrounding the bentonite particles and a concomitant increase in the sizes of the pores. They concluded that the eventual destruction of the initially observed membrane behavior correlated well with the time required to achieve steady-state diffusion of Ca²⁺. However, their test involved only one concentration (5 mM) and one salt (CaCl₂).

Meier et al. (2014) evaluated the persistence of membrane behavior for a GCL specimen by exposing the specimen to increasingly higher concentrations of potassium chloride (KCl) until any observed membrane behavior was essentially destroyed. They found that the membrane behavior of the GCL specimen persisted until the specimen was exposed to a KCl concentration of 400 mM (i.e., $C_{ave} = 200 \text{ mM KCl}$), which was well beyond the salt concentrations used in any previous study evaluating the membrane behavior of GCLs. They concluded that the results of their study represented the first evaluation of the persistence of membrane behavior of a GCL exposed to simple monovalent salt solutions. However, their study did not include an evaluation of the diffusion of the KCl.

The purpose of this paper is to extend the results previously reported by Meier et al. (2014) by including the diffusion behavior of the same GCL with respect to the same boundary concentrations of KCl. The results reported in this paper are believed to provide the first experimental evidence pertaining to quantifying the limiting membrane and diffusion behavior of a GCL.

2. Materials and methods

2.1. Materials

The GCL evaluated in this study was Bentomat[®] (Colloid Environmental Technologies Company (CETCO), Hoffman Estates, Illinois, USA). The mineralogy, cation exchange capacity (CEC), and index properties of the bentonite in the GCL were reported by Malusis and Shackelford (2002b) and are summarized in Table 1. The mineralogy includes 71% smectite (montmorillonite), 15% quartz, 7% mixed layer illite/smectite minerals, and 7% other minerals. The measured CEC was 47.7 meq/100 g, with 53% of the exchange complex comprising exchangeable sodium. The liquid limit (LL) and plastic limit (PL) measured in accordance with ASTM D4318 were 478% and 39%, respectively. Based on the Unified Soil Classification System (USCS, ASTM D2487), the bentonite classified as a high plasticity clay (CH). Further details on the chemical and physical properties of the bentonite in the GCL can be found in Malusis and Shackelford (2002a) and Meier et al. (2014).

The liquids used in this study included DIW and solutions of KCl (certified A.C.S.; Fisher Scientific, Fair Lawn, NJ) with target (desired) KCl concentrations, designated as C_{ot} , of 20, 35, 50, 100, 200, and 400 mM. The measured values of the electrical conductivity (EC) and pH for the DIW were approximately zero ($\leq 0.2 \text{ mS/m}$) and 7.0, respectively, whereas the measured values of EC and pH for each of the KCl solutions are summarized in Table 2. The actual, measured concentrations of KCl for each of the KCl solutions also are summarized in Table 2. As noted in Table 2, the measured concentrations of KCl were higher than the target concentrations of KCl. Only the measured concentrations were used in any relevant calculations.

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