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Membrane efficiency and diffusive tortuosity of a dense prehydrated geosynthetic clay liner

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ABSTRACT

In this study, a series of membrane/diffusion tests were conducted on specimens of a dense prehydrated geosynthetic clay liner (DPH GCL) subjected to KCl solutions (source concentration, $C_o = 8.7-160$ mM) in rigid-wall cells. The source KCl solutions and de-ionized water (DIW) were circulated across the top and bottom specimen boundaries, respectively, and membrane efficiency coefficients, ω, were measured based on the differential pressures induced across the specimens due to prevention of liquid flux. Also, effective salt-diffusion coefficients, D_s^*, and apparent tortuosity factors, τ_a, were determined for each specimen based on the steady-state diffusive Cl^- flux measured at the exit (bottom) boundary. The DPH GCL specimens exhibited higher ω and lower D_s^* (and, likewise, lower τ_a) relative to conventional (granular, non-prehydrated) GCL specimens tested under similar conditions. The results were consistent with the lower hydraulic conductivities, k, measured for the DPH GCL specimens and are attributed primarily to the higher bentonite dry densities in the DPH GCL specimens (1.1 Mg/m^3) relative to the conventional GCL specimens (~0.4 Mg/m^3), although differences in bentonite texture (powdered versus granular), bentonite type (treated versus untreated), and testing apparatus (rigid wall versus flexible wall) may have been contributing factors. Both the DPH GCL and the conventional GCL exhibited similar trends of decreasing τ_a with increasing ω. The τ_a values are considered to be a function of both a matrix tortuosity factor, τ_m, that accounts for the geometry of the interconnected pores, and a restrictive tortuosity factor, τ_r, that accounts for solute exclusion due to membrane behavior. Whereas the DPH GCL exhibited a lower τ_m relative to the conventional GCL, both GCLs exhibited similar trends of decreasing τ_r with increasing ω. The relationship between τ_r and ω for both GCLs is reasonably well represented by $\tau_r = 1 - \omega$, an expression that has been proposed for clay membranes in previous theoretical and experimental studies.

1. Introduction

The potential for engineered soil and geosynthetic barriers containing bentonite clay to act as semipermeable membranes, restricting the passage of charged solutes (ions) while allowing relatively unrestricted flow of water, has been reported in numerous studies conducted over the past 15 years (e.g., Malusis et al., 2001; Malusis and Shackelford, 2002a,b; Yeo et al., 2005; Henning et al., 2006; Di Emidio, 2010; Kang and Shackelford, 2010, 2011; Mazzieri et al., 2010; Bohnhoff and Shackelford, 2013; Shackelford, 2013; Malusis and Daniyarov, 2014; Di Emidio et al., 2015). The results of these studies indicate that, among the different types of bentonite-rich barriers used in geoenvironmental containment applications, geosynthetic clay liners (GCLs) are most likely to exhibit significant membrane behavior over a wide range of ion concentrations and overburden stresses, given the high bentonite content (~100%) in these barriers.

Numerous experimental studies have been conducted to investigate the hydraulic and contaminant transport properties of conventional GCLs consisting of loose, granular or powdered Na bentonite held between two geotextiles (e.g., Boardman and Daniel, 1996; Petrov and Rowe, 1997; Petrov et al., 1997; Lin and Benson, 2000; Lake and Rowe, 2000; Shackelford et al., 2000; Jo et al.,

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2001; Malusis and Shackelford, 2002a,b; Shackelford and Lee, 2003; Benson et al., 2010; Lange et al., 2010; Shackelford et al., 2010; Kang and Shackelford, 2011; Rowe and Hosney, 2013; Bouazza and Gates, 2014; Hosney and Rowe, 2014; Liu et al., 2014, 2015; Makusa et al., 2014), such as the GCL shown in Fig. 1a. However, a relatively new GCL product known as a dense, prehydrated GCL (DPH GCL; see Fig. 1b) has emerged as a potentially attractive alternative to conventional GCLs. Whereas conventional GCLs are typically ~10 mm thick and contain naturally dry (non-prehydrated) bentonite, the DPH GCL in Fig. 1b contains a 4–6 mm vacuum extruded layer of powdered bentonite that has been factory prehydrated with a treatment solution designed to improve the flexibility and resilience of the bentonite (Di Emidio, 2010). The combination of the smaller manufactured thickness and the higher bentonite mass per unit area (i.e., ~5 kg/m^2 for the DPH GCL versus ~4 kg/m^2 for the conventional GCL) yields higher bentonite dry densities for DPH GCLs (~1.1 Mg/m^3) relative to conventional GCLs (~0.4 Mg/m^3). As a result, DPH GCLs have been shown to exhibit extremely low hydraulic conductivities, k, to water (1×10^{-12} to 4×10^{-12} m/s; e.g., see Kolstad et al., 2004; Di Emidio, 2010; Mazzieri and Di Emidio, 2015), approximately an order of magnitude lower than for conventional GCLs. Also, studies by Kolstad et al. (2004) and Katsumi et al. (2008) indicate that DPH GCLs exhibit little or no degradation in k when permeated with highly concentrated salt solutions.

The purpose of this study was to evaluate the membrane behavior of a DPH GCL and the influence of this behavior on DPH GCL diffusion coefficients and tortuosity factors. Membrane behavior of a GCL typically is quantified in terms of a membrane efficiency coefficient, ω, also called the reflection coefficient, which normally is expected to vary from zero to unity ($0 \le \omega \le 1$). An ideal membrane ($\omega = 1$) would not allow any ions to enter the pores, whereas a non-membrane ($\omega = 0$) would not restrict ion transport. In general, GCLs act as non-ideal membranes, with ω varying over $0 \le \omega < 1$ depending on factors such as the dry density (or porosity), applied stress, and the types and concentrations of ions attempting to pass through the pores (e.g., Malusis and Shackelford, 2002a,b; Shackelford and Lee, 2003; Di Emidio, 2010; Kang and Shackelford, 2011). When $\omega > 0$, the containment performance of a GCL is enhanced by processes that limit solute migration, including hyperfiltration, chemico-osmosis, and restricted diffusion (Malusis et al., 2003).

Regarding diffusion, the influence of membrane efficiency on diffusion of inorganic salts through GCLs and GCL-type specimens has been investigated in a few studies (Malusis and Shackelford, 2002b; Di Emidio, 2010; Dominijanni et al., 2013; Malusis et al., 2015; Bohnhoff and Shackelford, 2015). The mass or molar flux, J_d, of the cation and anion of a binary salt migrating through a clay membrane under purely diffusive conditions (i.e., no hydraulic or chemico-osmotic liquid movement) may be represented by Fick's law, as follows (Malusis et al., 2012):

$$J_{dc} = -nD_s^*\frac{\partial C_c}{\partial x};\ J_{da} = -nD_s^*\frac{\partial C_a}{\partial x} \tag{1}$$

where n is total porosity and C_c and C_a are the concentrations of the cation and anion, respectively. The effective salt-diffusion coefficient, D_s^*, is defined as the product of the salt-diffusion coefficient in free solution, D_{so}, and the apparent tortuosity factor, τ_a (i.e. $D_s^* = \tau_a D_{so}$), which accounts for the reduction in diffusive solute flux that occurs in a porous medium relative to free solution (see Shackelford and Daniel, 1991). Furthermore, τ_a can be expressed as the product of a matrix tortuosity factor, τ_m, and a restrictive tortuosity factor, τ_r, as follows (Malusis and Shackelford, 2002b,c; Malusis et al., 2015):

$$\tau_a = \tau_m\tau_r = \tau_m\prod_{i=1}^{N}\tau_i \tag{2}$$

where τ_m accounts for tortuosity associated with the geometry of the interconnected pores and τ_r accounts for any number (N) of other mechanisms (represented by τ_i) that restrict diffusion, such as solute exclusion via membrane behavior and solute drag near particle surfaces (e.g., Kemper et al., 1964; Shackelford and Daniel, 1991; Shackelford and Moore, 2013).

Because τ_m is associated solely with the geometric interconnectivity of the pores, τ_m generally is considered constant for a given arrangement of soil particles and, therefore, independent of solute concentration. In contrast, τ_r for GCLs exhibiting membrane behavior (i.e., $\omega > 0$) has been shown to be concentration dependent, due to the implicit linkage between solute concentration and ω (Malusis and Shackelford, 2002b; Dominijanni et al., 2013; Malusis et al., 2015). Theoretically, τ_r (and, therefore, τ_a and D_s^*) will approach zero as the solute concentration decreases and ω approaches the ideal condition (i.e., $\omega = 1$), given that an ideal membrane would completely exclude solute migration. However, higher solute concentrations cause shrinkage of the diffuse double layers surrounding the bentonite particles and a decrease in ω, such that $\tau_r \to 1$ as $\omega \to 0$, assuming that all other potentially restrictive effects are insignificant. Both pore scale modeling (e.g., Dominijanni, 2005; Dominijanni and Manassero, 2012; Dominijanni et al., 2013) and the aforementioned experimental studies (e.g., see Malusis et al., 2015) indicate that τ_r for conventional GCLs may be approximated as a simple, linear function of ω, as follows:

$$\tau_r = 1 - \omega \tag{3}$$

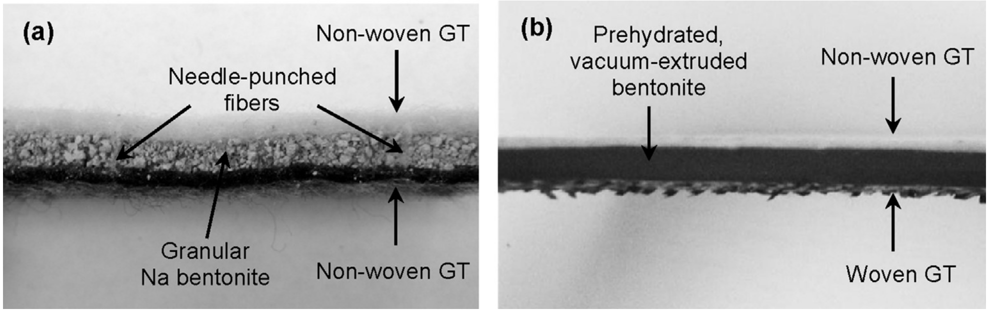

Fig. 1. Photographs of GCL cross sections (GT = Geotextile): (a) conventional GCL (Bentomat® DN); (b) DPH GCL.