

MODELING CONTAMINANT TRANSPORT THROUGH CLAY MEMBRANE BARRIERS

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Abstract: The migration of solutes in fine-grained soils of low hydraulic conductivity has become an important consideration with respect to the design and performance of waste containment barriers that consist of these soils (e.g., compacted clay liners, vertical cutoff walls). In this regard, solute transport analyses for engineered barriers typically are performed using solutions to the advective-diffusive equation. However, advective-diffusive transport theory represents a limiting case of the more general coupled flux transport theory in which the coupling terms (e.g., chemico-osmosis) are assumed to be negligible. In the case of clay soils that act as semi-permeable membranes restricting the passage of solutes, the use of advective-diffusive theory may not be appropriate. This presentation includes simulations of a coupled solute transport model that can be used to simulate transient, one-dimensional transport through reactive (i.e., ion exchanging) clay soil membranes. Simulation results will be presented using measured values of the chemico-osmotic efficiency coefficient (ω), the effective diffusion coefficient (D^*), and the hydraulic conductivity (k_h) for a bentonite-based geosynthetic clay liner (GCL) that acts as a semi-permeable membrane. The results highlight the differences between coupled solute transport theory and advective-diffusive theory based on appropriately measured transport parameters.

Results of several laboratory studies have shown that some clay soils have the ability to act as membranes that restrict the passage of charged solutes (i.e., ions) (Kemper and Rollins, 1966; Fritz and Marine, 1983). Such membrane behavior also results in chemico-osmosis, or the movement of liquid in response to a solute concentration gradient (Olsen, 1969; Barbour and Fredlund, 1989). Both of these effects result in reduced solute transport through a soil barrier for waste containment. For example, in the absence of an electrical current, the total contaminant (solute) mass flux, J , through a clay membrane barrier (CMB) can be written as follows (Yeung and Mitchell, 1993):

$$J = (1 - \omega)q_h C + q_\pi C - nD^* \frac{\partial C}{\partial x} \quad (1)$$

where ω is the soil chemico-osmotic efficiency coefficient, C is the contaminant (solute) concentration, $q_h [= k_h i_h]$, where k_h = the soil hydraulic conductivity and i_h = the hydraulic gradient] is the hydraulic liquid flux in accordance with Darcy's law, $q_\pi [= \omega k_h i_\pi]$, where i_π = the gradient in chemico-osmotic pressure head] is the chemico-osmotic liquid flux across a soil membrane from a lower solute concentration to a higher solute concentration (i.e., opposite to the direction of solute diffusion), and D^* is the effective diffusion coefficient. The chemico-osmotic efficiency coefficient, ω , ranges from zero ($\omega = 0$) for non-membranes to unity ($\omega = 1$) for "ideal" membranes that completely restrict the passage of solutes (Kemper and Rollins,

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1966). However, CMBs typically only partially restrict the passage of solutes (i.e., $0 < \omega < 1$) and, therefore, are considered “non-ideal” membranes.

The first term in Eq. 1 is the traditional advective transport term that is reduced by a factor of $(1 - \omega)$ due to the process of hyperfiltration whereby solutes are filtered out of solution during passage through the CMB under an applied hydraulic gradient. The second term represents the counter advective transport of solutes due to chemico-osmotic liquid flux, q_π . The third term represents solute diffusion through soil in the form of Fick's first law. In the limit as $\omega \rightarrow 0$ and, thus, $q_\pi \rightarrow 0$, Eq. 1 reduces to the traditional advective-diffusive solute mass flux expression, or

$$J|_{\omega=0} = q_h C - nD^* \frac{\partial C}{\partial x} \quad (2)$$

However, $J \rightarrow 0$ as $\omega \rightarrow 1$. Thus, the effect of a CMB is to reduce the contaminant mass flux through the barrier relative to the mass flux that would occur in the absence of membrane behavior. This reduction in the contaminant mass flux results explicitly from the coupling processes of hyperfiltration and chemico-osmotic counter advection in Eq. 1. In addition, an “implicit” solute restriction occurs in the diffusive term due to the requirement that $D^* \rightarrow 0$ as $\omega \rightarrow 1$ because, by definition, no contaminant can enter an ideal membrane.

This “implicit” solute restriction is illustrated in Fig. 1 based on laboratory test results using the test apparatus and procedures described by Malusis et al. (2001) in which 10-mm-thick specimens of a needle-punched geosynthetic clay liner (GCL) containing sodium bentonite (i.e., Bentomat[®]) were subjected to potassium chloride (KCl) solutions. The test results indicate that ω decreases from 0.63 to 0.14 and D^* increases from $0.783 \times 10^{-10} \text{ m}^2/\text{s}$ to $2.38 \times 10^{-10} \text{ m}^2/\text{s}$ as the source KCl concentration C_o increases from 0.0039 M to 0.047 M (Malusis, 2001). Extrapolation of the test results indicates that the GCL would behave as a non-membrane (i.e., $\omega = 0$) at a source KCl concentration of approximately 0.1 M or higher. The corresponding value of D^* for the GCL as $C_o \rightarrow 0.1 \text{ M}$ and $\omega \rightarrow 0$ approaches a maximum value of approximately $2.8 \times 10^{-10} \text{ m}^2/\text{s}$. This maximum D^* value is assumed to represent a non-membrane condition for the GCL in which the diffuse double layers are sufficiently compressed that the tortuosity of the solute migration pathway is due solely to the geometry of the interconnected pores (i.e., the matrix tortuosity).

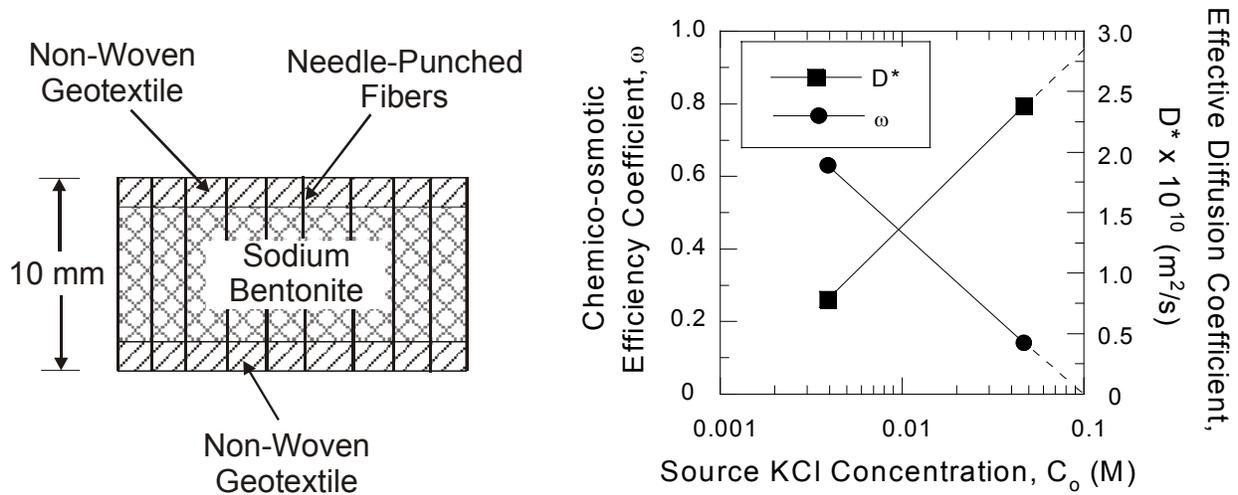


Fig. 1 – Schematic cross-section of Bentomat[®] GCL and chemico-osmotic/diffusion test results.

Theoretical differences between the coupled solute transport model (CTM) based on Eq. 1 and the advective-diffusive model in which $\omega = 0$ by definition (ADM) based on Eq. 2 are evaluated for a 1.0-m thick CMB in Fig. 2 based on the data in Fig. 1. The transient exit flux curves [i.e., $J(x=L)$] for KCl in Fig. 2 are based on a constant source concentration ($C_o = 0.0039$ M or 0.047 M), a perfectly-flushing exit boundary condition [i.e., $C(x=L) = 0$; Rabideau and Khandelwal, 1998], and $i_h = 0$ (Malusis, 2001). The results indicate that the exit fluxes given by the CTM are lower than the exit fluxes given by the ADM in which coupling effects inherently are ignored. This example illustrates that use of the coupled solute transport model may be more appropriate than traditional advective-diffusive theory for modeling contaminant transport through a CMB. However, any correct simulation of the contaminant mass flux through a CMB must be based on values of D^* measured at the correct concentration for the application and, therefore, the appropriate ω value (Shackelford et al., 2001). The use of D^* values measured separately from ω at concentrations greater than the “threshold” concentration at which $\omega = 0$ (e.g., to decrease the testing time) in simulations performed to evaluate contaminant transport through CMBs will neglect the implicit correlation between D^* and ω (as illustrated in Fig. 1) and, therefore, will be fundamentally incorrect.

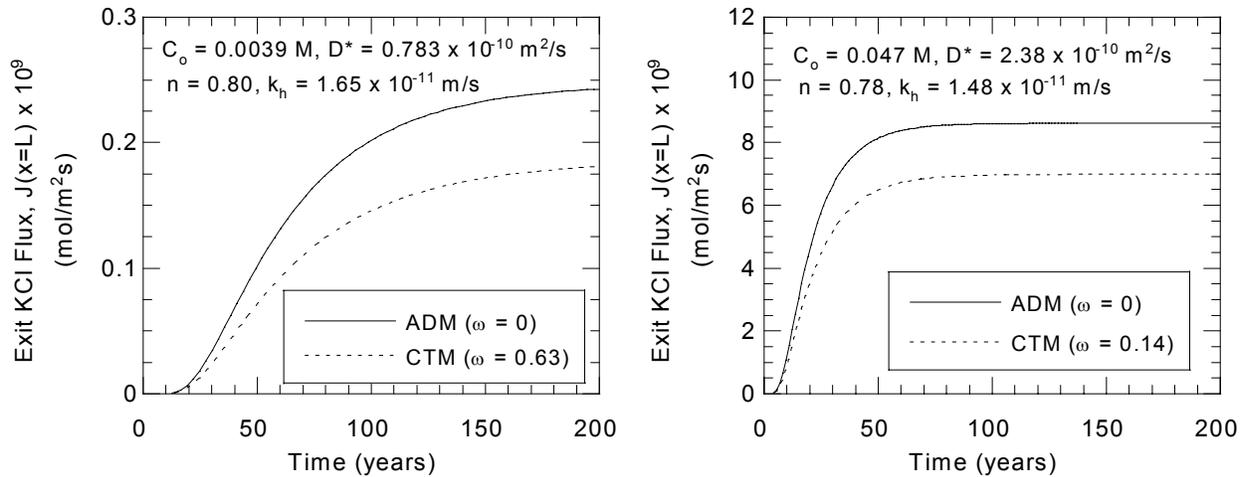


Fig. 2 – Theoretical exit flux of KCl from a 1.0-m-thick CMB ($i_h = 0$).

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