



Research paper

Electric–hydraulic–chemical coupled modeling of solute transport through landfill clay liners



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ABSTRACT

Pollutant migration in dense clay barriers appears to be strongly influenced by the electric double layer of colloidal surfaces. Osmosis that resulted from chemical potential or electric potential difference across the clay membrane has been successively described in a number of theoretical works. Streaming potential (SP) which is present in charged porous medium under hydraulic gradient has been recognized as a significant factor governing the mass migration in compacted clays. However, few studies have been carried out in geo-environmental area with regard to this physical phenomenon. A coupled model was proposed to account for the effects of electrical, chemical and fluidic fields on solute transport in porous medium in this study. The electrical field deals with both the streaming potential and the externally applied electrical potential. The coupled nonlinear partial differential equations are numerically simulated by finite element method. Both the steady state solution and the time-dependent solution were investigated with the consideration of a series of influential factors. The streaming potential coefficient and the electro-osmotic coefficient were found to control the solute transport process. The potential application of the materials with tendency of producing SP was discussed. With appropriate selection of materials and parameters, optimum barring effect could be obtained for soil barriers in waste containment applications.

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1. Introduction

Solute transport in porous media takes place in many aspects, e.g. the engineering facilities, industrial plants and the environmental processes. In geo-environmental areas, pollutants from contaminated soils or landfills always spread towards the surroundings in terms of diffusion or advection and dispersion (Lake and Rowe, 2000; Leij et al., 1991; Rowe et al., 2000). A well designed and carefully installed barrier system for landfills usually consists of a certain number of layers of geosynthetic clay liners and compacted clay liners. The performance of these barriers is largely dependent on the capacity to resist the breakthrough of the contained pollutants. Many efforts have been made to investigate the solute transportation in dense clay liners, i.e. experimental works on parameterizations (Gorenflo et al., 2002; Manassero and Dominijanni, 2003; Rowe and Badv, 1996a; Rowe et al., 2000; Shackelford and Daniel, 1991a; Shackelford and Redmond, 1995) and theoretical works on risk analysis (Lake and Rowe, 2000; Rowe and Badv, 1996a; Rowe et al., 2000). For GCLs and compacted clays, diffusion was reported to be the dominant pattern of transport that involves a variety of solute types (Rowe and Badv, 1996a,b;

Shackelford and Daniel, 1991a,b; Shackelford and Redmond, 1995). Recent studies have successfully incorporated osmosis in some solute transport models (Malusis and Shackelford, 2002; Manassero and Dominijanni, 2003; Neuzil, 2000; Olsen, 1969; Yeung and Mitchell, 1993). Yeung and Mitchell developed a coupled model to account for the chemical, hydraulic and electrical factors (Yeung and Mitchell, 1993). Manassero and Dominijanni proposed a solid theoretical framework for chemo-osmosis (Manassero and Dominijanni, 2003). The chemo-osmotic coefficient was modified by the latter study, however, irrespective of the electrical field, to predict the total barring of solute by clay membrane in extreme conditions for a perfect membrane with reflection coefficient $\omega = 1.0$ (Manassero and Dominijanni, 2003). Solute transport in charged porous medium depends on various factors such as the electrical potential derived from the concentration gradient, the electric double layer of colloids and the hydration of dissolved solutes (Chatterji, 2004). Further downward scaling will see the rising significance of the coupling effects of these factors due to the close relationship with electric double layers of colloidal surfaces.

Clay minerals, e.g. smectite and illite, have abundant permanent surface charges, high cation exchange capacity, large interlayer space, thick double electric layer and thus appear to be active in both reactivity and hydrophilicity. The flow of charged pore fluid due to a pressure gradient at zero electric current can produce potential difference, namely

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the streaming potential (SP), through such type of porous media enriched with charged capillary (Peeters et al., 1999). Streaming potential is a typical characteristic of selective membranes which inexclusively contain the compacted clays. The direction of the resultant streaming potential relates to the positive or negative charges of a diffuse layer on colloidal surfaces. The measurement of streaming potential has been extensively used in the characterization of surface electric properties, of which the zeta potential is one of the most commonly referred and widely demanded information (Childress and Elimelech, 1996; Deshiikan et al., 1998; Deshmukh and Childress, 2001; Elimelech et al., 1994; Huisman and Trägårdh, 1999; Werner et al., 2001). Despite these investigations, streaming potential still remains to be regarded as a tool or indicator for colloidal characterizations instead of an influencing factor for solute transport through capillary porous media.

Streaming potential has widely been observed in a number of porous media, e.g. clays, rocks, mud and oil shale, in both laboratory and field studies (Chatterji, 2004; Demir, 1988; Gairon and Swartzendruber, 1975; Heister et al., 2005, 2006; Lorne et al., 1999; Tenchov, 1992). It is noted that the conduction current that resulted from the streaming potential through a porous media is non-negligible (Szymczyk et al., 2007). The migration of charged ions is likely to be influenced as long as electrical current exists in a porous medium. Heister et al. (2005) reported an obvious effect of SP on hydraulic conductivity of bentonite. Demir (1988) found a significant reduction in the flow rate and solute concentration of chloride brine through a smectite layer, which were related to the salt infiltration and streaming potential. More and more evidences clearly indicated the non-negligible coupling effects of streaming potential on permeation and solute transport (Revil et al., 2007). In spite of well-known recognition of its existence, as indicated in Yeung's paper (Yeung, 1990), few attempts have been done to take SP into the account of modeling works.

The aim of this study is to propose an electric–hydraulic–chemical coupled solute transport model to assess the complicated interaction of the multi-fields and its impact on pollutant leakage through landfill liners. Based on previous relevant theories and experimental observations, permeation-induced SP was incorporated into the governing equations with a reasonable simplification with respect to the final equilibrium state (steady state solution). The model was first validated by fitting several sets of test data about the effect of salt concentration on hydraulic conductivity of bentonite, and was further investigated numerically by varying different sets of parameters in order to understand the transport process with a bit more depth. Critical factors dominating the solute transport were discovered and evaluated numerically, revealing potential engineering application significance. The key objective of this study is not only to provide a simulational analysis of the landfill barrier system, but also to identify the governing factors that have long been ignored.

2. Theoretical framework

Various physical field variables could result in complex coupled effects on solute transport in porous medium, as shown in Table 1.

Table 1
Field variables and the coupled effects on solute transport in porous medium (after Mitchell and Soga, 2005).

Variables	Coupled effect		
	Solvent flow	Electrical current	Solute transport
Pressure head (P)	Darcy's Law	Streaming potential/ current	Advective dispersion
Electrical potential gradient (U)	Electro-osmosis	Ohm's Law	Ionic mobility
Concentration gradient (C)	Chemo-osmosis	Diffusion potential	Fick's Law

Yeung and Mitchell (1993) have reported the detailed deduction process under non-equilibrium thermodynamic principles. Manassero and Dominijanni (2003) carried out a similar study on phenomenological modeling for chemo-osmosis. The equations proposed here are consistent with these previously established models which in most parts are founded on classical assumption, however, we focus more on the development of functions and discussions relevant to streaming potential and its effect on both solute and permeate fluxes with specific assumptions.

2.1. Governing equations

2.1.1. Solvent flux

The electro-osmotic flow could be written as (Li et al., 2011; Shang, 1997)

$$J_e = k_e \nabla(-U) \quad (1)$$

where J_e is electro-osmotic flux, k_e is electro-osmotic coefficient, and U is electrical potential. The parameter k_e (m^2/sV), which controls the water flow rate under a unit voltage gradient, is expressed in this theoretical relationship (Mitchell and Soga, 2005): $k_e = \frac{\zeta \epsilon n}{\eta}$, where ζ (V) is the zeta potential of solids, ϵ (F/m) is the permittivity of the pore fluid, n is the porosity of the porous medium and η (Ns/m²) is the viscosity of the pore fluid. It is clear that k_e is mainly governed by the zeta potential (ζ) and porosity (n) given that the permittivity and viscosity of the pore fluid remain constant.

The gradient of chemical concentration induces osmotic pressure in the form of

$$\nabla \pi = \omega RT \nabla(-C) \quad (2)$$

where π is osmotic pressure (Pa), ω is chemo-osmotic coefficient, T is temperature (K) and C is chemical concentration (M).

The fluidic flux driven by hydraulic gradient takes the form as

$$J_p = \frac{k}{\gamma V_w} [\nabla(-P) - \omega RT \nabla(-C)] \quad (3)$$

where J_p is pressure-driven flux, k is hydraulic conductivity (m/s), P is hydraulic pressure, γ is unit weight and V_w is volume of pore fluid in the representative elementary volume. The equation for J_p is consistent in form with that of Manassero and Dominijanni (2003).

The overall flux (J_v) consists of two parts,

$$J_v = J_e + J_p \quad (4)$$

Then

$$J_v = k_e \nabla(-U) + \frac{k}{\gamma V_w} [\nabla(-P) - \omega RT \nabla(-C)] \quad (5)$$

2.1.2. Solute flux

Solute migration in liquid phase has three forms, i.e. advective–diffusion, electro-osmosis and hydraulic dispersion according to the classification in Table 1. The overall solute flux is

$$J_s = J_{ds} + J_{es} + J_{ps} \quad (6)$$

where J_s is solute flux, J_{ds} is diffusive flux, J_{es} is electro-osmotic flux and J_{ps} is dispersive flux.

Following the deduction of Yeung (1990) this equation becomes

$$J_s = nD_e \nabla(-C) + \frac{nD_e |z| FC}{RT} \nabla(-U) + (1-\omega) C J_v \quad (7)$$

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