



A three-scale model for ionic solute transport in swelling clays incorporating ion–ion correlation effects



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ABSTRACT

A new three-scale model is proposed to describe the movement of ionic species of different valences in swelling clays characterized by three separate length scales (nano, micro, and macro) and two levels of porosity (nano- and micropores). At the finest (nano) scale the medium is treated as charged clay particles saturated by aqueous electrolyte solution containing monovalent and divalent ions forming the electrical double layer. A new constitutive law is constructed for the disjoining pressure based on the numerical resolution of non-local problem at the nanoscale which, in contrast to the Poisson–Boltzmann theory for point charge ions, is capable of capturing the short-range interactions between the ions due to their finite size. At the intermediate scale (microscale), the two-phase homogenized particle/electrolyte solution system is represented by swollen clay clusters (or aggregates) with the nanoscale disjoining pressure incorporated in a modified form of Terzaghi's effective principle. At the macroscale, the electro-chemical-mechanical couplings within clay clusters is homogenized with the ion transport in the bulk fluid lying in the micro pores. The resultant macroscopic picture is governed by a three-scale model wherein ion transport takes place in the bulk solution strongly coupled with the mechanics of the clay clusters which play the role of sources/sinks of mass to the bulk fluid associated with ion adsorption/desorption in the electrical double layer at the nanoscale. Within the context of the quasi-steady version of the multiscale model, wherein the electrolyte solution in the nanopores is assumed at instantaneous thermodynamic equilibrium with the bulk fluid in the micropores, we build-up numerically the ion-adsorption isotherms along with the constitutive law of the retardation coefficients of monovalent and divalent ions. In addition, the constitutive law for the macroscopic swelling pressure is reconstructed numerically showing patterns of attractive forces between particles for bivalent ions for particular ranges of bulk concentrations. The three-scale model is applied to numerically simulate ion diffusion in a compacted clay liner underneath a sanitary landfill. Owing to the distinct constitutive behavior of the swelling pressure and partition coefficient for each ionic species, different compaction regimes and diffusion/adsorption patterns, with totally different characteristic time scales, are observed for sodium and calcium migration in the clay liner.

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1. Introduction

The adequate design of engineered barriers for waste containment to fulfill environmental regulations involves optimization of the performance of surface caps and subsurface liners to ensure minimization of pollutant migration to the subsurface. Such issue remains very relevant as society continue to generate large

amounts of hazardous waste requiring efficient long-term storage to the solid waste containment industry. Hydrated swelling clays have been widely adopted as buffer materials in geoenvironmental engineering playing a key rule in engineered liner systems in municipal solid waste landfills, mining waste containment facilities and seals for high-level long lived radioactive waste disposal sites. Owing to the self-sealing capability arising from the expandable lattice along with large specific surface area, high chemical capacity for adsorption of contaminants and low permeability, compacted clays such as bentonite possess effective barrier properties for isolation of wastes from the surrounding environment [28].

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