BRIDGING BETWEEN MICRO- AND MACROSCALES IN REACTIVE CONTAMINANT TRASNPORT IN EXPANSIVE POROUS MEDIA

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Summary In this article we provide a precise correlation between the macroscopic partition coefficient governing absorption of charged solutes in swelling clays and the microscopic distribution of the electric potential satisfying the Poisson-Boltzmann problem in the micro pore domain occupied by the electrolyte solution. This is accomplished within the framework of a dual porosity model which is derived within a formal homogenization asymptotic analysis applied to two levels of averaging to include the micro and macro-pores of the swelling medium. In addition to the adsorption of the ions, which is accurately captured by the proposed three-scale formulation, a notable consequence of the approach are the other microscopic problems posed in a unit cell for the electro-chemo-mechanical coefficients. By solving the local closure problems we are capable of developing accurate constitutive laws for the effective coefficients.

INTRODUCTION

Swelling porous media such as 2-1 lattice clays, hydrophilic polymers, shales, corneal endothelium and connective biological tissues are ubiquitous in almost all aspects of life. For example swelling clay soils are widely distributed in the earth's crust. In agriculture, water adsorption by the clay determines the ability of soils to transport and supply water, nutrients and pesticides. Clay swelling is of widespread relevance in geotechnical and geoenvironmental fields. Due to the low hydraulic conductivity, plasticity, swelling and adsorptive capacity for contaminants, compacted clays (bentonites) have been suggested appropriate engineered barriers to minimize the leakage and dissolution of radioactive materials from high-level waste repositories into the groundwater supply. The buffer material must have swelling properties because cracks in the surrounding rock may appear and need to be filled up. Compacted soil liners have been used as earthen barriers to minimize the leakage from landfills to the subsurface environment. In the context of oil and gas production swelling plays a crucial role in the stability of wells drilled through shales in particular using water-based in particular drilling muds. Swelling is also a concern to the civil enginner because of the often severe structural damage caused by expansive soils. Expansive materials have in common a structure that can be loosely identified as a mixture of macromolecules or colloidal particles (polymers, clay particles, proteglycanns) and solvent (water, hydrocarbons). The solvent is either adsorbed to the macromolecules in the form of an electrolyte solution with dissociated ionic species, or in bulk (i.e. free of any adsorptive force). As such, it is imperative the correct understanding of the constitutive behavior of these systems to account for their swelling nature.

At the microscale these physico-chemical effects are governed by the electro-hydrodynamics coupled with Nernst-Planck equations and the Poisson-Boltzmann problem which govern the fluid movement, transport of mobile charges and electric potential distribution in the electrolyte solution Moyne and Murad [1]). The development of ionic atmospheres around the clay minerals is also responsible for the appearance of physico-chemical stresses induced in the solid phase causing the expansion/shrinking of the clay lattice. When water comes in contact with a mass of clay crystals, it penetrates between the layers and forces them apart. When hydration progresses the crystals may expand several times their original thickness. For long-range interactions swelling is dominated by electrostatic forces and the adsorbed fluid is viewedan aqueous electrolyte solution consisting of water and an entirely dissociated salt with strong electrolytes. Derjaguin and coworkers [2] described the lyophilic interaction between fluid and substrate in terms of a disjoining pressure,.

At the mesoscale (the homogenized microscale) the highly heterogeneous microstructural solid-fluid interactions are represented in an averaged fashion. At this scale, fluid and solid are viewed as overlaying continua forming the clay peds (clusters or aggregates) with averaged properties established at every point in the mixture. Under near-equilibrium conditions, the averaged fluid flow, flux of ions and electric current are linearly coupled with the gradients of head, concentration and streaming potentials through Onsager's reciprocity relations (Lai et al. [3], Huyghe and Janssen [4]) .

At the macroscale, the mesoscopic two-scale electro-chemo-mechanical model for the clay clusters is homogenized with the equations governing flow and transport of the species in the bulk solution lying in the macro-pore system. Application of this second level of up-scaling leads to a macroscopic model resembling in form the so-called dual porosity models for fissured media . In the macroscopic picture of a dual porosity model, an interconnected network of macro-pores (or fissures) provides most of the global permeability for the macroscopic flow and transport of species whereas most of the storage takes place in the relatively low permeability matrix blocks (Arbogast [5]).

The goal of this contribution is the derivation of a three-scale model of dual porosity type for expansive clays based on a rigorous up-scaling of the microstructure response. To accomplish this task we begin by presenting the two-scale electro-chemo-mechanical model or the clay clusters based on Onsager's reciprocity relations. Further, to evaluate the magnitude of the Onsager's coefficients e consider a particular form of microstructure herein each clay cluster is composed of parallel particles of close face-to-face contact. In this simplified microscopic arrangement we establish precise correlations between the averaged parameters and the microscopic electrokinetics of the electrolyte solution. and then perform a rigorous homogenization procedure to upscale the meso-scale electro-chemo-mechanical model for the clay aggregates coupled with the low and transport of the species in the bulk fluid lying in he macro-pore system. Assuming local periodicity of the aggregates, the homogenization procedure yields a microstructure model of dual porosity type with macroscopic equations coupled via generalized transfer functions to local mesoscale problems posed within each clay cluster. By assuming instantaneous local thermodynamic equilibrium between he clay peds and macro-pores we show that the constitutive law for the mass transfer function of the species is equivalent to the appearance of a partition coefficient which governs the instantaneous sorption/desorption of the the species by the micro-pores. This leads to a quasi-steady approach which is characterized by a time-scale assumption wherein the macroscopic system follows regular evolution process coupled to mesoscopic local equilibrium states of the clay cluster and macro-pore systems. The notable feature of the three-scale approach relies in its capability in providing a double averaging macro/meso/micro representation or the partition coefficient in terms of the microscopic behavior of he electrolyte solution in the micro-pores, whose charge distribution is ruled by the Poisson-Boltzmann roblem.

By discretizing the Poisson-Boltzmann problem we compute the microscale electric potential and adopt two levels of averaging to obtain the behavior of the macroscopic partition coefficient. Fig. 1 depicts the numerical partition coefficient in terms of salinity for two values of the disjoining pressure obtained within this up-scaling procedure.

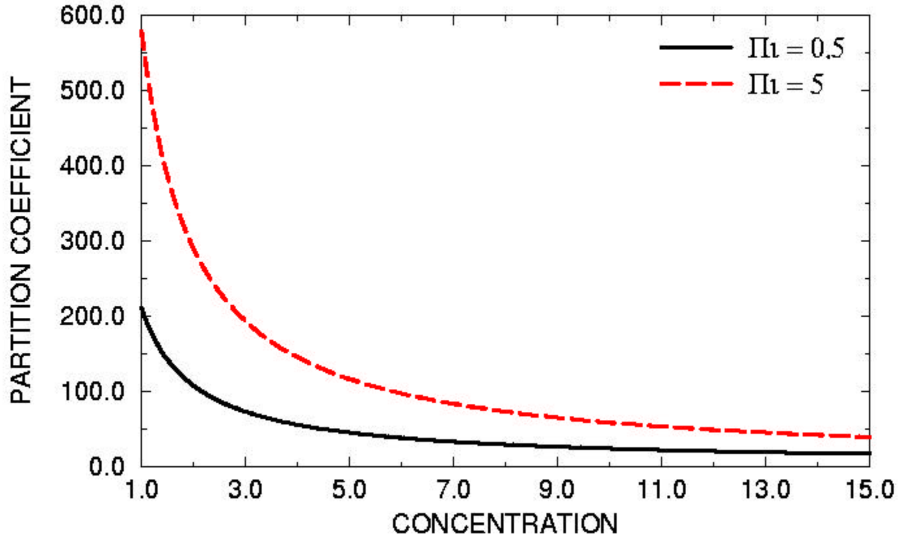

Fig.1 Partition coefficient derived by a double averaging procedure form the microscopic Poisson-Boltzmann problem

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