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Research paper

Fluid flow and effective conductivity calculations on numerical images of bentonite microstructure

F. Bouchelaghem[a,*], R. Pusch[b]

[a] Sorbonne Universites, UPMC Univ Paris 06, CNRS, UMR 7190, Institut Jean Le Rond d'Alembert, F-75005 Paris, France
[b] Lulea University of Technology, Geotechnical Division, Porson, Lulea, Sweden

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ABSTRACT

Hydraulic conductivities of compacted water-saturated bentonite were computed based on the real microstructure. The Homogenization of Periodic Media approach employed fully acknowledges the heterogeneous and multiscale microstructure of clay, as well as locally varying physical flow properties. Consequently, three levels of description were considered : the microscopic level of clay particles, the mesoscopic level of clay aggregates, mineral grains and inter-aggregate porosity, and the macroscopic level of the sample subjected to fluid pressure gradients in the laboratory. Starting from the local description of fluid flow, the expression of the effective hydraulic conductivity tensor was derived. The soft and dense gels and the open voids may form a connected flow path or remain occluded. The local problems were solved on the microstructure obtained from a digitalized micrograph by image analysis. The contribution to macroscopic flow by the soft and dense gels was investigated in various configurations, and comparisons were made with hydraulic conductivity data for MX-80 bentonite.

1. Introduction

1.1. Clay microstructure

Smectites are characterized by a heterogeneous arrangement of nanometer-scale lamellae stacked together to form clay particles, micrometer-scale grains, water and air which saturate the various pore spaces that appear on different scales (Jozja, 2003; Holzer et al., 2010; Keller et al., 2014; Pusch and Yong, 2006; Tessier et al., 1992). As a result, clay microstructure controls most of their physical properties such as ion diffusion, swelling pressure and hydraulic conductivity. In water-saturated compacted bentonite, mesopores that contain free pore water may be distinguished from nanoscale pores that contain water molecules bound by surface forces. The multi-scale pore structure continuously changes during hydration, swelling and compaction processes, which complicates its characterization (Holzer et al., 2010; Tomioka et al., 2010). This explains the scarcity of microstructural data available to quantify the pore size distribution and pore connectivity in compacted bentonite. As pointed out in Holzer et al. (2010) and Keller et al. (2014), the proportions of free and bound porewater in compacted bentonite are the subject of ongoing discussions and investigations, and tomography techniques are still under development in order to quantify precisely the mesopore distribution in compacted swelling clays.

In Pusch and Schomburg (1999) and Pusch (2001) Transmission Electron Microscopy (TEM) micrographs of compacted and hydrated MX-80 bentonite displayed the formation of clay gels of variable density by linking of aggregates exfoliated from the expanding clay grains. Consequently, four main phases were distinguished: open voids which present no resistance to flow, clay gel-filled voids (designated as soft gels) which are characterized by a finite hydraulic conductivity, clay aggregates (designated as dense gels) and non-smectite grains which are assumed to be impermeable. Impervious zones and denser and softer gels were schematized by a grid of elementary cells with distinct hydraulic conductivities. The effective conductivity was then approximated by an analytical formula combining the individual cells in parallel or in series depending on their orientation with respect to the macroscopic flow direction. Tomioka et al. (2010) used X-ray microtomography for examining the morphological evolution of compacted Na-montmorillonite before and after saturation. They concluded that the outer montmorillonite sheets are likely to swell and form a gel that occupies the intergranular voids, whereas the inner sheets are not affected by the water saturation process. Holzer et al. (2010) and Kelleret al. (2014) investigated the intergranular pore space (mesopores) of compacted and hydrated MX-80 bentonite using high resolution 3D imaging with Focused Ion Beam nanotomography. They observed that clay particles' hydration in non-compacted bentonite (dry density $\rho_{dry} = 0.39$ g/cm^3) is associated with extensive exfoliation and dispersion of thin clay layers. At $\rho_{dry} = 1.24$–1.46 g/cm^3, clay

* Corresponding author.
E-mail address: fatiha.bouchelaghem@upmc.fr (F. Bouchelaghem).

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aggregates located in the intergranular pore space displayed a honeycomb structure of stacked clay platelets filled with a colloidal clay gel of lower density. At higher densities, the small remaining fraction of mesopores were elongated in shape and no longer inter-connected, which led the authors to conclude that fluid flow and transport cannot take place through the mesopores alone. In compacted hydrated bentonites such as those intended for radioactive waste disposal, the isolated mesopores are connected only through the nanopores present within the clay matrix. As we will see in Section 3.1 the macroscopic fluid flow in the case of unconnected mesopores is then controlled by the conductivity of the clay matrix.

1.2. Multiscale modelling of fluid flow in geomaterials

In recent studies (Adler et al., 1990; Al-Omari and Masad, 2004; Blunt et al., 2013; Masad et al., 2000) the effective permeability of various porous materials (natural sandstones, carbonate reservoirs, asphalt pavements, sands, glass beads) was determined by solving the local equations of fluid flow. The microstructure was obtained directly from the sample by image processing, or idealized by utilizing statistical characteristics. The computed flow fields displayed preferential paths depending on the size and connectivity of the pores, with whole areas of the pores relatively isolated from the flow. In the study by Blair et al. (1996), a two-point spatial correlation function obtained from image processing was employed to estimate several microstructural parameters (porosity, specific surface area, mean grain size, effective pore size), and combined with a Kozeny-Carman relation to estimate the intrinsic permeability of porous glasses and natural sandstones. Blunt et al. (2013) applied network modelling at larger scales to extract a topologically representative network consisting of pores and throats from the image, and the flow and transport properties were described semi-analytically. In Adler et al. (1990), Al-Omari and Masad (2004), and Masad et al. (2000), the computed velocity field was driven by the pressure difference prescribed between the inlet and the outlet of the microstructure. Periodic boundary conditions were imposed for the velocity components, Darcy's law was written at the macroscopic scale and the effective permeability tensor was simply defined as the ratio between the average fluid velocity vector and the known pressure gradient. The previous studies concerned materials of intrinsic permeability higher than 10^{-12} m^2, and did not investigate very low permeability materials such as compacted clays. Moreover, the pore space comprised only intergranular porosity with no nanoporosity such as encountered in clays.

Ichikawa et al. (1999, 2001, 2004a) and Wang et al. (2003) employed the Homogenization of Periodic Media to model fluid flow through clays. The microscopic and the macroscopic fluid flow equations were obtained from the Navier-Stokes equations using double scale asymptotic expansions, the two levels of description being related by the macroscopic permeability. Clay material was composed of lamellae of hydrated montmorillonite on the nanometric scale, while the mesoscopic domain consisted of clay particles and non-smectite grains. Water was distributed within the interlamellar and interparticle spaces, disregarding the mesopores. The computed effective permeability changed significantly with the void ratio, and comparison with experimental data (Pusch, 1994) confirmed the difficulties of assessing the permeability of very low permeability clays.

1.3. Modelling approach and outline of the paper

An experimental investigation on saturated bentonite (Pusch and Schomburg, 1999) has been employed in a numerical homogenization model in order to relate the macroscopic hydraulic conductivity with structural and micro-textural properties. With respect to existing works on compacted bentonites (Bouchelaghem and Jozja, 2009a,b; Ichikawa et al., 1999, 2001, 2004a; Wang et al., 2003) which are all based on simplified microstructures (Poiseuille-like flow for lamellae of hydrated montmorillonite, symmetrical arrangements of spherical or cylindrical grains on the mesoscopic scale), the macroscopic permeability tensor is computed on real microstructures, while both mesopores and non-smectite grains are taken into account, leading to new effective permeability formulations. According to Baltean (1999) and Wodie (1992), the effective permeability will be obtained on the macroscopic scale by upscaling from the mesoscopic scale alone, consisting here of the arrangement of clay aggregates, inter-aggregate voids and non-smectite grains. As mentioned in Section 1.1, microstructural data regarding the intergranular space (Holzer et al., 2010; Keller et al., 2014; Pusch, 2001; Pusch and Yong, 2006) allow us to distinguish between dense and soft parts of the heterogeneous clay matrix, implying regions of low permeability and highly permeable channels. Accordingly, we assume that the soft gels always contribute to the macroscopic flow, while the dense gels may be impervious to fluid flow, completely open to flow, or characterized by an effective conductivity that can be determined from a separate upscaling approach.

The mesoscopic clay microstructure is described in Section 2.2, followed by the construction of a Finite Element mesh for numerical treatment in Section 2.3. Based on the description of effective fluid flow behaviour on the mesoscopic scale within clay aggregates (Section 2.4) and within inter-aggregate pores (Section 2.5), the method of asymptotic expansions is employed to derive the effective hydraulic conductivity tensor for unconnected mesopores (Section 3.1), for connected mesopores (Section 3.2), and when no mesopores are present (Section 3.3). Numerical simulations results and a comparison with experimental data on MX-80 bentonite are presented in Section 4. Finally, conclusions are drawn and future work is outlined in Section 5.

2. Formulation of the problem on the mesoscopic scale

2.1. Materials and sample preparation

MX-80 bentonite is a commercial smectite-rich clay, composed of numerous montomorillonite lamellae arranged in basic units or stacks (Pusch, 1999, 2001). The chemical constitution is as follows: 65–75 % montmorillonite, 10–14 % quartz, 5–9 % feldspars, 2–4 % mica and chlorite, 3–5 % carbonates and chlorite, 1–3 % heavy minerals. The stacks are estimated to consist of 3–5 lamellae with equally oriented crystal axes in Na-saturated smectite, and about 10 lamellae if Ca is the dominant exchangeable cation (Pusch, 2001). Buffer clay blocks are prepared by compacting air-dry MX-80 clay powder at a compaction pressure of 100 MPa. The dry density of the grains is ca 1980 kg m^{-3} at 10% by weight and the dry density of the powder mass poured in the form and slightly compacted is ca 1200 kg m^{-3}. Transmission Electron Micrographs of very thin sections (about 3 10^{-2} μ m) are obtained by acrylate impregnation of the clay under confinement, the sample preparation is detailed in Pusch (2001).

2.2. Microstructure

The general description is based on observations of clay fabric that distinguish between two levels of arrangement: the particles and aggregation of particles (Holzer et al., 2010; Jozja, 2003; Pusch and Yong, 2006; Tessier et al., 1992).

The microscopic domain of clay particles, which is characterized by the length l_c, consists of stacked lamellae of montmorillonite, inter-layer water and lenticular pores (Bouchelaghem and Jozja, 2009a,b). Clay aggregates are represented by a periodical repetition of clay particles and inter-particle porosity. The mesoscopic domain is illustrated in Fig. 1a showing a TEM of MX-80 clay taken from Pusch (1999). The periodic microstructure Ω, of characteristic length l, is composed of the porous matrix Ω_c of clay aggregates, water saturated inter-aggregate voids Ω_f which may be connected or not, and non-smectite grains Ω_s (in black in Fig. 1a). We will alternatively assume that the dense clay gels (in pink in Fig. 1a) are open to fluid flow and form a part of Ω_f, or they