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Unsaturated Hydraulic Conductivity of Compacted Bentonite: Revisit of Microstructure Effects

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Abstract. Developing a sound theoretical description for the evolution of unsaturated hydraulic conductivity of compacted bentonite during re-saturation has remained a challenge because of the complex microstructure effects influenced by physico-chemical phenomena. This paper proposes a new relationship for the unsaturated hydraulic conductivity of compacted bentonite based on Kozeny-Carman relationship. A new relationship is also proposed for the relative permeability of compacted bentonite that incorporates the effects of microstructure swelling. The application of the new relationship for the case of compacted GMZ bentonite provides well correlated results with the experimental data reported in the literature.

Keywords: Compacted bentonite · Clay microstructure · Unsaturated Permeability

1 Introduction

In the context of geological disposal concepts for high level nuclear waste in crystalline rock, partially saturated compacted bentonite is to be emplaced in the depositional holes as buffer material. The clay buffer will undergo a re-saturation phase due to moisture intrusion from the host rock. Experimental observations have shown that the moisture flow and chemical transport in smectite rich clays are strongly influenced by swelling/shrinkage of the clay microstructure (Pusch and Yong 2006). The application of conventional unsaturated hydraulic conductivity relationships in predicting the moisture flow and chemical transport processes in compacted bentonite might overestimate the rate of re-saturation or chloride transport against the experimental observations (e.g. Sedighi et al. 2012; 2018). Limited experimental and theoretical approaches exist that can provide a sound description of the evolution of unsaturated hydraulic conductivity of compacted bentonite.

In this paper, various phenomenological aspects of microstructure of compacted bentonite and its evolution during wetting process are re-visited to develop a theoretical

description of pore system. The established Kozeny-Carman relationship (Carman 1939; Kozeny 1927) for permeability of porous media is adopted and modified to describe the intrinsic permeability of compacted bentonite under confined conditions. A relationship for the relative permeability of compacted bentonite is proposed by incorporating the effects of swelling/shrinkage of the clay microstructure. The relationship proposed for the hydraulic conductivity of compacted bentonite is tested against a series of experimental results from the literature.

2 Microstructure Evolution in Compacted Bentonite

The pore system of compacted bentonite can be described by two scales of porosity: (i) micro porosity that includes the spaces between the unit layers of smectite (microstructure) and (ii) macro porosity that represents the pore spaces between the particles and aggregates of particles (Fig. 1). Macro/meso pores are the only major water conduits. Water molecules are exchanged between the micro and macro/meso pores. The water in micro pores has different properties compared with free water and is widely considered as an immobile phase (Sedighi and Thomas 2014). The interlayer hydration or dehydration process can only alter the distance between the clay microstructure units (interlayer distance) by adding or removing discrete layers of water in micro-porosity. During wetting/re-saturation, the size of micro porosity increases as the results of adsorption of water molecules into the interlayer space. Consequently, under confined conditions (i.e. swelling is constrained), the macro porosity can shrink, thus reducing the space for water to flow. By increasing the water content in smectite, the number of unit layers in a particle consistently decreases and the larger aggregates are divided into smaller particles (Saiyouri et al. 2004) that affects the tortuosity of the porous system. Figure 1 presents a schematic diagram of the evolution of pore system in compacted bentonite during re-saturation at constant total volume.

The volume of interlayer water can be described as:

$$V_{IL} = d_{IL}NA_s \quad (1)$$

where, d_{IL} is the thickness of interlayer water (i.e. interlayer distance), N stands for the total number of unit layer of smectite (TOT) in the sample and A_s is the surface area of a TOT.

The thickness of interlayer water varies with water content due to adsorption of one to three layers of water in the interlayer space. The variations of interlayer water with the total volumetric water content follow a hydrodynamic adsorption behaviour that can be simplified by a quasi-dynamic equation (Azizian 2004), given as:

$$d_{IL} = d - d_0 = 9[1 - \exp(b\theta_l)] \quad (2)$$

where, d and d_0 is the basal spacing and thickness of single TOT sheet, respectively. b is a constant that is related to the water adsorption isotherm of smectite ($b = -2.63$

Fig. 1. Schematic diagram of pore system evolution of compacted bentonite during wetting.

provides a good agreement with water/vapour adsorption data of smectite (Holmboe et al. 2012; Villar et al. 2012). θ_l is the total volumetric water content.

The total number of TOT layers in a sample (N) can be given as:

$$N = \frac{m_{\text{smectite}}}{m_s} n_c \quad (3)$$

where, m_{smectite} is the total mass of smectite in the soil, m_s is the mass of single particle containing the number of n_c TOTs and n_c is the number of stacked TOT layers per particle that varies with water content (Saïyouri et al. 2004).

The surface area of a TOT (A_s) can be calculated from the total specific surface area (A_{total}) and mass of smectite (m_{smectite}) in the soil:

$$A_s = \frac{m_{\text{smectite}} A_{\text{total}}}{2N} \quad (4)$$

where, A_{total} is the total specific surface area.

The micro porosity (ϕ_{micro}) and macro porosity (ϕ_{macro}) can therefore be calculated as:

$$\phi_{\text{micro}} = 0.5 d_{\text{IL}} A_{\text{total}} \rho_d^s \quad (5)$$

$$\phi_{\text{macro}} = \phi_t - \phi_{\text{micro}} \quad (6)$$

where, ρ_d^s is the dry density of smectite in the soil and ϕ_t is the total porosity

3 Saturated Hydraulic Conductivity of Compacted Bentonite

Kozeny-Carman relationship (KC) is an established theoretical approach that provides a relationship between porosity and permeability in porous media (Carman 1939; Kozeny 1927). However, it has been shown that the KC for compacted clays does not provide a good correlation with experimental observations (Chapuis and Aubertin 2003). The hypothesis presented here is that since part of the pore water in smectite is adsorbed in the microstructure and it is regarded as an immobile phase, the system of compacted bentonite should then be conceptualised by a dual porosity system, in which only the macro porosity contributes to the flow. Accordingly, the total porosity in KC relationship should be replaced by the macro porosity to provide a realistic pore system of compacted bentonite.

A modified Kozeny-Carman relationship for saturated hydraulic conductivity of compacted bentonite is therefore proposed that reflects the effects of microstructure on flow behaviour through the use of macro porosity in the relationship:

$$k_{sat} = \left(\frac{\gamma_w}{\mu_w} \right) \left(\frac{C_s}{\rho_d^2 \tau^2 A_{ext}^2} \right) \frac{\phi_{macro}^3}{(1 - \phi_{macro})^2} \quad (7)$$

where, γ_w and μ_w are the unit weight and viscosity of water, respectively. C_s is a shape factor. ρ_d is the dry density of soil. τ is the tortuosity and A_{ext} is the external specific surface area (i.e. $A_{ext} = A_{total}/n_c$).

4 Unsaturated Hydraulic Conductivity of Compacted Bentonite

The unsaturated hydraulic conductivity of compacted bentonite can be presented as a function of its saturated hydraulic conductivity (Mualem 1976), given as:

$$k_{unsat} = k_r k_{sat} \quad (8)$$

where, k_r is the relative permeability that is commonly described as a function of water content (Mualem 1976):

$$k_r = \left(\frac{\theta_l - \theta_r}{\theta_{sat} - \theta_r} \right)^\alpha \quad (9)$$

where, θ_{sat} and θ_r are the saturated and residual volumetric water contents, respectively. α is a constant parameter. θ_r is calculated from the relationships between water content and suction based on experimental water/vapour adsorption data.

The relative permeability relationships are mainly based on observations and measurements in coarse grained soils where the entire porosity is available for water flow. However, in the case of compacted bentonite a considerable proportion of water

is located in micro porosity, which is almost immobile. To account for this, a revised relationship for relative permeability is proposed as follows:

$$k_r = \left(\frac{\theta_{macro} - \theta_{macro,r}}{\phi_{macro} - \theta_{macro,r}} \right)^\alpha \quad (10)$$

where, θ_{macro} is the volumetric water content in macro porosity and $\theta_{macro,r}$ is the residual macro water content. The value of θ_{macro} is calculated from ϕ_{micro} (Eq. 5) and total volumetric water content (θ_l) at a given soil water potential or suction ($\theta_{macro} = \theta_l - \phi_{micro}$).

5 Application to Compacted GMZ Bentonite

The relationship proposed for hydraulic conductivity is applied for the case of GMZ bentonite, compacted at a dry density of 1.7 Mg/m^3 . GMZ bentonite contains 75.4% Na-smectite (Ye et al. 2009). Figure 2 presents the variations of micro porosity and macro porosity with suction for GMZ bentonite calculated from Eqs. 5 and 6. Figure 2 presents a comparison of micro porosity with those for compacted MX-80 bentonite calculated by Sedighi and Thomas (2014) based on a geochemical modelling approach. The results are compatible with those presented for compacted MX-80. Both MX-80 and GMZ are sodium smectite-rich clays although, the smectite content of the GMZ bentonite is lower than that of the MX-80 bentonite. It can be observed that at saturation, ϕ_{micro} is approximately 2.6 times larger than that of ϕ_{macro} that accounts only for 27.9% of the total porosity. Substituting all variables and constants, the saturated hydraulic conductivity of compacted GMZ is found to be $1.01 \times 10^{-13} \text{ (m/s)}$. The value obtained is very close to the value reported based on experimental tests (e.g. 1.18×10^{-13} by Ye et al. (2009)). In this study we use van Genuchten's relationship between water content and suction (Van Genuchten 1980):

$$\theta_l = \frac{\theta_{sat} - \theta_r}{[1 + (\alpha s)^n]^{(1-1/n)}} \quad (11)$$

where, s is suction. α and n are fitting parameters.

Figure 3 presents the variations of unsaturated hydraulic conductivity of compacted GMZ bentonite with total volumetric water content. The relative permeability is calculated using Eqs. 9 and 10 for comparison with experimental data presented by Ye et al. (2009). For both approaches the value of α is assumed to be 3 that is acceptable value across in the literature. The results obtained by using the proposed relationship for the relative permeability correlate well with the experimental data. In comparison with the classic approach that considers total volumetric water content/porosity, the new relationship provides a better correlation with the experimental results.

Fig. 2. Variations of micro and macro porosity of compacted bentonite with suction.

Fig. 3. Variation of unsaturated hydraulic conductivity of compacted GMZ with water content.

6 Conclusions

A new approach for describing the effects of microstructure on hydraulic conductivity of compacted bentonite was presented. The Kozeny-Carman relationship was revisited to include the dual porosity system of compacted bentonite. It was shown that by using the revised relative permeability relationship proposed in this paper, an improved correlation with experimental data for the case of compacted GMZ bentonite is achieved. The new developments have important applications in predicting the flow behaviour of smectite rich clays in which a more accurate description of the pore system evolution is included.

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