

Micromechanics modeling the solute diffusivity of unsaturated granular materials



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ABSTRACT

This work is devoted to modeling the evolution of the homogenized solute diffusion coefficient within unsaturated granular materials by means of micromechanics approach. On the basis of its distinct role in solute diffusion, the liquid water within unsaturated granular materials is distinguished into four types, namely intergranular layer (interconnected capillary water), isolated capillary water, wetting layer and water film. Application on two sands shows the capability of the model to accurately reproduce the experimental results. When saturation degree is higher than the residual saturation degree S_r^r , the evolution of homogenized solute diffusion coefficient with respect to the saturation degree depends significantly on the connectivity of the capillary water. Below S_r^r , depending on the connectivity of the wetting layer, the homogenized solute diffusion coefficient within unsaturated sands decreases by 2–6 orders of magnitude with respect to that in bulk liquid water. The upper bound of the solute diffusion coefficient contributed by the water films is 4–6 orders of magnitude lower than that in bulk liquid water.

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1. Introduction

Solute diffusion is an essential transport mechanism in either saturated porous media with low permeability or unsaturated porous media at low saturation degree (Hu and Wang, 2003). Solute diffusion in unsaturated granular materials plays a significant role in agriculture engineering, contaminant transport and remediation, and nuclear waste disposal (Romkens and Bruce, 1964; Conca and Wright, 1992; Hu and Wang, 2003). The liquid distribution within unsaturated granular material is believed to greatly influence the transport properties of the unsaturated granular materials. Solute diffusion at different saturation regimes, namely high saturation regime, intermediate saturation regime and low saturation regime, exhibits distinct transport mechanisms (Hu and Wang, 2003). Therefore, characterizing the evolution of liquid distribution at various saturation degrees is of prior importance in transport problems.

Homogenized transport properties are function of liquid content (or saturation degree), which is related to the matric suction (or capillary pressure) according to the water retention curve. In the past decades, extensive empirical models were developed to estimate

the effective solute diffusion coefficient within unsaturated porous media (Romkens and Bruce, 1964; Mehta et al., 1995; Olesen et al., 1999; Hu and Wang, 2003). These models are phenomenological since local physical phenomena are rarely taken into account. For instance, below residual saturation degree, the thick water films (the wetting layer) attached on the solid grain surface are found to be dominant in the transport problems, which is the main physical phenomenon neglected by the Archie's law (Conca and Wright, 1992; Hu and Wang, 2003; Han et al., 2009). Therefore, micromechanics approach, accounting for both microstructure morphology and local phase properties, has been used widely in estimating the homogenized transport properties of porous media (Gruescu et al., 2007; Lemarchand et al., 2009; Nguyen, 2014). The latter often resorts to the classic Eshelby-based schemes such as self-consistent scheme (SC) and Mori-Tanaka scheme (MT) (Lemarchand et al., 2009).

When MT is used in the transport problems, the capillary water is usually considered as matrix, the solid phase inclusions along with gaseous phase inclusions are embedded in the matrix. This morphology ensures the interconnectivity of the capillary water during the overall desaturation (drainage) process, which is in contradiction with several experimental results which exhibit notable percolation threshold at low saturation degree (Romkens and Bruce, 1964; Hamamoto et al., 2010). As an alternative, SC is used to estimate the homogenized solute diffusion since the percolation threshold can

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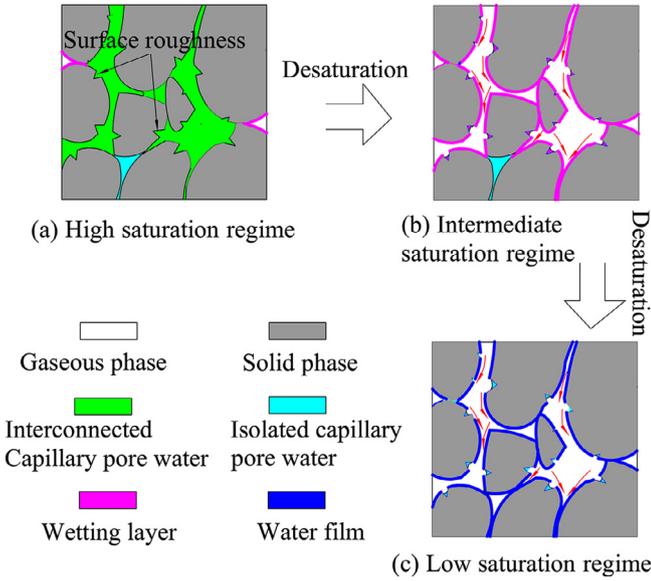


Fig. 1. Schematic illustration of liquid distribution within unsaturated granular material at distinct saturation regimes.

be inherently accounted for in this scheme. Unfortunately, within unsaturated porous media, the percolation threshold saturation degree Sr_p estimated by SC is $\frac{1}{3\phi}$ (ϕ is pore volume fraction and $\phi \geq 1/3$) (Yang, 2013), which is obviously overestimated (Koelman and De Kijper, 1997). The overestimation lies in the fact that the connectivity of the capillary water within unsaturated porous media, as an important topological information, is not physically incorporated into the Eshelby-based micromechanics estimates (Kleinfelder-Domelle and Cushman, 2012).

In this study, four different types of liquid water, namely intergranular layer (interconnected capillary water), isolated capillary water, wetting layer and water film, are distinguished and specified within unsaturated granular materials. Based on the local liquid characterization, a physical-based micromechanics model is developed to predict the solute diffusion coefficient of the unsaturated porous media. The micromechanics model is validated by the experimental results from the literature and compared with the empirical Archie's law. The model provides a link between the local parameters such as connectivity of capillary water and different tendencies of the homogenized solute diffusion coefficient of the two sands with respect to saturation degree.

2. Physical characterization of unsaturated granular materials

2.1. Saturation regimes

On the premise of the two critical saturation degrees Sr^r and 1%, three saturation regimes in unsaturated porous media, namely high saturation regime, intermediate saturation regime and low saturation regime, are used to distinguish the liquid configuration at different saturation degrees. At high saturation regime ($Sr \geq Sr^r$), the capillary water can be classified as percolating (interconnected) capillary water and isolated capillary water (Raouf and Hassanizadeh, 2013). The latter is attributed to either the segmentation by the percolating non-wetting gaseous phase (Raouf and Hassanizadeh, 2013) or the existence of the dead-end pore space (Nakashima, 1995). Therefore, at high saturation regime, the capillary water is classified as isolated capillary water and interconnected capillary water. The latter is believed to play a vital role in the solute diffusion and is idealized as an intergranular layer attached on the surface of solid grain in our model (see “High saturation regime” in Fig. 1).

Intermediate saturation regime corresponds to a regime where the saturation degree is below the residual saturation degree Sr^r ($Sr < Sr^r$) (Romkens and Bruce, 1964; Fredlund et al., 1997). At intermediate saturation degree, the intergranular layers disappear since the connectivity of the capillary water vanishes. As shown in Fig. 1, the capillary water trapped in pendular rings and surface roughness forms a continuous transport pathway, which will play an important role in the solute diffusion (Or and Tuller, 2000; Blunt et al., 2002). This continuous transport pathway (the thick water film) is defined as “wetting layer” by Blunt et al. (2002), which keeps interconnected even when the saturation degree Sr reaches as low as 1% (Dullien et al., 1989). The thickness of the wetting layer is closely associated with the morphological characteristics of surface roughness and the capillary pressure. Owing to the complicated morphological characteristics of the surface roughness, it is a difficult task to precisely characterize the relationship between the capillary pressure and thickness of the wetting layer (Or and Tuller, 2000; Blunt et al., 2002; Kibbey, 2013). However, it is generally agreed that the magnitude of the thickness of the wetting layer is the order of the characteristics size of surface roughness, ranging between microns to sub-microns (Dullien et al., 1989; Han et al., 2009; Kibbey, 2013). In addition, there still exists certain amount of isolated capillary water (Kibbey, 2013), either in small capillary pores or in pendular rings (or surface pits and concaves), which is bridged by the wetting layers (see “intermediate saturation regime” in Fig. 1).

As illustrated in “Low saturation regime” in Fig. 1, with the progressive drying (e.g. $Sr < 1\%$ (Dullien et al., 1989; Blunt et al., 2002)), the low saturation degree regime is reached: capillary water trapped in small capillary pores (the isolated capillary pore water) is totally drained out. Correspondingly, the interconnected wetting layers disappear. Thereafter, the capillary water trapped in surface roughness and pendular rings is connected by the water films. The latter predominate the solute diffusion at low saturation regime.

In the following context, the superscripts/subscripts s , g , il , ip , wl and f denote solid phase, gaseous phase, intergranular layer, isolated capillary water, wetting layer and water film, respectively. The bold-faced letters (e.g. \mathbf{A}) stand for second order tensor while the underlined letters (e.g. \underline{H}) denote vectors.

2.2. Thickness of water film

At nano-scale, the stability and thickness of water film are determined by the so-called disjoining pressure $\Pi(h)$ (Derjaguin and Churaev, 1978; Israelachvili, 1991). The latter is linear sum of three different components: the relative long range repulsive electrostatic force $\Pi_e(h)$, the Van der Waals component $\Pi_v(h)$, the structural component of disjoining pressure $\Pi_s(h)$ (Churaev and Derjaguin, 1985; Majumdar and Mezic, 1999; Gonçalvès et al., 2010):

$$\Pi(h) = \Pi_e(h) + \Pi_v(h) + \Pi_s(h) \quad (1)$$

with (Churaev and Derjaguin, 1985; Israelachvili, 1991):

$$\begin{cases} \Pi_e(h) = 64nkT\zeta_1\zeta_2\exp(-\kappa h) \\ \Pi_v(h) = -\frac{A}{6\pi h^3} \\ \Pi_s(h) = K_{sr}\exp(-h/\lambda_{sr}) + K_{lr}\exp(-h/\lambda_{lr}) \end{cases} \quad (2)$$

where Debye length $\frac{1}{\kappa} = (\epsilon\epsilon_0kT)^{1/2}(2z^2e^2n)^{-1/2}$; ϵ ($=80$) is the dielectric permittivity of liquid water, ϵ_0 ($=8.854 \times 10^{-12}\text{C}^2\text{J}^{-1}\text{m}^{-1}$) is the permittivity of the free space; k ($=1.381 \times 10^{-23}\text{J} \cdot \text{K}^{-1}$) is the Boltzmann constant; T is the temperature (in Kelvin); n is the number density of the ions, $n = N_a \times \rho$, ρ (in $\text{mol}\cdot\text{L}^{-1}$) is the concentration of the bulk electrolyte solution, N_a ($=6.022 \times 10^{23}$) is Avogadro's constant; e ($=1.602 \times 10^{-19}\text{C}$) is the elementary charge; z is the electron charge; $\zeta_i = \frac{\exp(z\psi_i/(2kT)) - 1}{\exp(z\psi_i/(2kT)) + 1}$ ($i \in \{1, 2\}$), ψ_i are the surface potentials of i^{th} interface ($i \in \{1, 2\}$), herein, subscript 1 denotes solid-water film interface, subscript 2 denotes gas-water film interface; A is the

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