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Characterization of unsaturated diffusivity of tight sandstones using neutron radiography

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ABSTRACT

Water flow in unsaturated tight sandstones plays a significant role in the area of the secondary and enhanced hydrocarbon recovery as well as the geological storage of carbon dioxide and nuclear wastes. Although a few non-destructive techniques, such as nuclear magnetic resonance, magnetic resonance imaging and X-ray imaging, can be used to capture fluid transport in sub-micron pores, great challenges exist due to the presence of iron or the use of the contrast agents (e.g. cesium chloride and salts), resulting in inaccurate results or alteration of the wetting behavior of the porous media. In addition, models for describing diffusivity and water transport in unsaturated tight sandstones are also limited. In this work, the neutron radiography facility at China Advanced Research Reactor was used to determine water content profiles during the water imbibition in two types of tight sandstones: silty sandstone and coarse grained sandstone. The diffusivity was determined separately by three methods, including Matano's method, Meyer-Warwick method and a fractal method, which was introduced as probably the first attempt to relate the microstructure observed by the high resolution X-ray computed tomography (CT) with the unsaturated diffusivity function for the tested tight sandstones. The air-entry value and the fractal dimension used in the fractal model were calculated based on the results of mercury intrusion porosimetry and CT data, respectively. The results from neutron images illustrate that the fractal model can give a reasonable description of the diffusivity function for the tested sandstones. Meyer-Warwick model produces a little bit higher diffusivity value at low water content range. The fractal model works better for the silty sandstone. Results also show that the value of water diffusivity increases with the increase in volumetric water content for both tested tight sandstones. This work shows that neutron radiography offers a feasible and more reliable way for characterizing fluid flow in other tight geo-materials and the fractal model also provides an easier way to give a quantitative description of the diffusivity than the core-flooding or centrifuge drainage experiment.

1. Introduction

Unsaturated is a state of 'partial saturation' or 'aerated', which means the pore space of geo-materials has not been fully occupied by water [1]. In most case of water flow in unsaturated rocks, the penetration and accumulation of water is driven by capillary force and gravity in a strongly nonlinear way. In recent years, the importance of understanding water transport in unsaturated porous rocks has attracted increasing attention from many fields, including hydrological geology [2], mining engineering [3], petroleum engineering [4–6], carbon dioxide geological sequestration [7,8], nuclear waste storage [9], building and heritage conservation [10,11]. Normally, the sorptivity and unsaturated diffusivity function are used to describe transient water movement in rocks under partially-saturated conditions [10]. The weighting method [10,12,13] and instantaneous profile method [1] were usually selected to determine unsaturated diffusivity of geo-materials. However, the reliability of the traditional weighting method for providing an accurate prediction of dynamic water distribution during water imbibition in low permeability sandstones has been increasingly questioned due to the lack of direct experimental

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observations to verify the calculated results. As an alternative, the non-destructive monitoring method offers the capability to directly measure the evolution of moisture content profiles with water absorption time, allowing the analysis of unsaturated water transport and the determination of the moisture diffusivity of porous materials [14–18]. These non-destructive testing techniques mainly involve X-ray computed tomography [14], neutron imaging [16,19,20], Nuclear Magnetic Resonance (NMR) and Magnetic Resonance Imaging (MRI) [21,22].

Many studies have been carried out to investigate the flow in unsaturated porous media by the aforementioned non-destructive testing techniques. An early investigation on unsaturated water flow in porous materials using NMR was performed by Gummerson et al. [21]. Leech et al. [22] derived the unsaturated diffusivity functions for large cylindrical concrete samples (100 mm in both diameter and length) from magnetic resonance images during a simple one-dimensional wetting process. However, due to the presence of iron, it is extremely challenging for NMR or MRI to obtain accurate measurement of spatial water content. X-ray imaging is more widely used to investigate the microstructure and water distribution of geo-materials such as rock, soil and concrete with different sizes [14,23–25] due to its easy access and relatively lower cost. However, as the contrast agents (e.g. cesium chloride (CsCl) and salts) are helpful for X-ray imaging to detect water movement and content distribution in unsaturated porous media, it may alter the wetting behavior of the porous media, such as the contact angles of water to solids [26]. Compared with X-ray, neutrons interact with the nuclei of the atoms only and this interaction is not enhanced with the increase of atomic number [27]. Thus, neutrons are strongly attenuated by hydrogen in water but much less attenuated by gas phase in pores and the mineral matrix of rocks, which make the neutron imaging an effective method to investigate hydrogen-rich fluids in geo-materials and engineered porous materials [20,28]. Neutron radiography has been commonly applied to capture the variations of moisture content during water imbibition in both natural and engineered porous materials, such as brick, concrete, porous glass, sand, rock and soil [20,26,29–33]. Recently, Kang et al. [19] utilized neutron imaging facility in Oak National Laboratory to investigate moisture diffusion in Berea sandstone, and the extracted data were fitted to obtain the diffusivity as a function of water content of Berea Sandstone.

Understanding water transport in tight sandstone with low porosity and permeability is of significance to many geologic processes such as the recovery of gas and oil from unconventional resources [34] and carbon dioxide geological sequestration project [7,8]. In contrast with high porosity and permeability sandstone such as Berea sandstone, the pore structure of tight sandstones is more complex. For water flow in unsaturated tight sandstone, high capillary force is expected due to the abundant micron to submicron scale pore throats. What makes the water transport in tight sandstone even more complex is the tortuous flow path caused by the low porosity, poor connectivity, irregular pore shape associated with the clay minerals filling in the pores [35]. Thus, two contrasting media i.e. homogeneous silty sandstone and heterogeneous coarse grained sandstone were selected in this study to check the effect of pore structure on water flow in unsaturated tight sandstone. Compared with conventional sandstone with high permeability, the detection of fluid flow behavior in tight sandstones is expected to be more challenging because of the smaller pore size and slower diffusion. Moreover, microstructure effects on the unsaturated diffusivity in tight sandstone have not yet to be fully exploited. Although the microstructure of geo-materials has the features of fractal [36,37] and fractal theory has been widely applied in geo-mechanics, only limited reports [38–42] have been found that apply the fractal theories to the prediction of water permeability or sorptivity of rocks.

In this study, the unsaturated diffusivity of tight sandstones with different pore structures was investigated using neutron radiography. The pore structures of tested sandstones were described and characterized by high resolution X-ray CT and mercury intrusion porosimetry (MIP). An empirical correction parameter β was introduced to the *Lambert-Beer* law to reduce the effect of beam hardening and neutron scattering. Calibration experiment was also performed using the neutron radiography facility located at the China Advanced Research Reactor (CARR) with a special aluminum-made calibration cell. The measured water attenuation coefficient and the related correction parameter were reported for the first time, which can provide a valuable reference for subsequent experiments using the CARR neutron radiography facility. Moreover, the unsaturated diffusivity of tight sandstones was calculated based on the Matano's method [43], Meyer Warwick model [44] and a fractal diffusivity model. The air-entry value and the fractal dimension used in the fractal model were calculated based on the results of MIP and CT data, respectively. Finally, the time-lapse water content maps during water imbibition in two types of unsaturated tight sandstones were analyzed.

2. Theories for unsaturated flow

Theories for describing water flow in unsaturated soils have been well established [1,10,45]. The diffusivity defined as the ratio of the hydraulic conductivity to specific moisture capacity [1] is considered as the dominant parameter to characterize its flow. The diffusivity function can be determined directly using horizontal infiltration method [46] or calculated by combining the hydraulic conductivity function and water retention curve with outflow methods [47]. Recently, models to predict soil-water retention curve, unsaturated hydraulic conductivity and unsaturated diffusivity using fractal theories [48–51] have also been established.

In this part, the basic theory of the diffusivity was first introduced and then two methods (i.e. Matano method and Meyer-Warwick method) for the calculation of unsaturated diffusivity with the Boltzmann transformation were presented. Finally, in Section 2.3, a fractal model to calculate the unsaturated diffusivity function was presented.

2.1. Diffusivity function based on Matano's method

The water content dependent diffusivity has been widely used as the theoretical framework for assessing the evolution of the main macroscopic variables during imbibition [44,52,53]. A classical theory of unsaturated water flow was established for the cases where water was absorbed by dominant capillary forces in a column of porous media. In this case, the flow process can be considered as one dimensional because the effect of gravity can be assumed negligible. The governing equation, i.e. Richards equation, of the one-dimensional capillary-driven flow was described as [10,45]:

$$\frac{\partial \theta_{\mathrm{w}}}{\partial t} = \frac{\partial}{\partial x}\left(D(\theta_w)\frac{\partial \theta_{\mathrm{w}}}{\partial x}\right) \tag{1}$$

where θ_w is the water content expressed as the ratio of water volume to bulk volume, t represents the time and x is the space coordinate along water flow direction. $D(\theta_{\mathrm{w}})$ denotes the diffusivity function.

To transform Eq. (1) into an ordinary differential equation from a partial differential equation, the Boltzmann variable $\lambda = x{\cdot}t^{-0.5}$ is applied [46]. If the sample is initial dry and the satiated volumetric water content is denoted as θ_s, with the well known initial and boundary conditions, i.e. $\theta_{\mathrm{w}}(x, 0) = 0$ and $\theta_{\mathrm{w}}(0, t) = \theta_s$, Eq. (1) can