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#### **Chemical Engineering Science**

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## An apparent liquid permeability model of dual-wettability nanoporous media: A case study of shale



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#### HIGHLIGHTS

- Water transport behavior in dual-wettability shale matrix system is revealed.
- Wettability and pore size related liquid slippage effect is considered.
- Liquid slip in nanopores improves permeability of organic-rich shale significantly.
- Sensitivity of structural parameters on apparent liquid permeability is examined.

#### ARTICLE INFO

# Article history: Received 1 January 2018 Received in revised form 18 April 2018 Accepted 8 May 2018 Available online 9 May 2018

Keywords:
Nanoporous media
Dual-wettability
Shale
Apparent liquid permeability
Fractal
Stochastic method

#### ABSTRACT

The hydraulic fracturing fluid can easily infiltrate the ultra-tight shale matrix due to the remarkable slip feature of the liquid flow in nanoscale pores, showing a higher-than-expected fluid-loss in shale gas development. In this paper, a stochastic apparent liquid permeability (ALP) model is developed to reveal water transport mechanisms in dual-wettability nanoporous shale based on the transport behavior in a single nanotube. The present model considers the wettability and pore size related liquid slip effect, total organic carbon (TOC) content, and the structural parameters (maximum and minimum pore size of inorganic or inorganic matter, porosity) of shale matrix. The results show that the multilayer sticking effect (structural water molecules in the pore surface) constricts the flow capacity and slightly decreases the ALP for the inorganic hydrophilic matter, while, a large slip length for the water flow can be observed and the ALP is dramatically improved if the nanopores in the organic matter are strong hydrophobic, especially in organic-rich shale reservoir. The ALP can be reduced or enhanced with the increase of TOC content, which is determined by the relative importance of pore size difference (between organic matter and inorganic matter) and wettability of organic matter. Additionally, the sensitivity analysis of structural parameters on the ALP are examined.

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#### 1. Introduction

Liquid flow in porous media with nanoscale pores is of special importance to energy storage and conversion (Aricò et al., 2005), water purification (Shannon et al., 2008; Mattia et al., 2015), environmental engineering (Warner et al., 2012), fibrous materials (Woudberg, 2017) and petroleum industries (Zhang et al., 2017a). Recently, thanks to the technological advancement in multi-stage slip water hydraulic fracturing, the commercial gas production from ultra-tight shale reservoirs have emerged as major sources of natural gas supply in the North America, and possibly an critical

role in Europe and Asia (EIA, 2016). As a result, the hydraulic fracturing fluid (mainly composed by water) transport through the nanoporous shale becomes an important process involved in shale gas development.

During the hydraulic fracturing operation process, thousands of cubic meters of fracturing fluid are forcibly injected into the subsurface reservoir. However, field data indicate that only a small fraction of the injected fluid (some gas wells are even less than 5%) can be recovered during clean-up period (Zhang et al., 2017b). The huge unrecovered fluid has attracted lots of attention related to the concerns in economy (lowering fracturing fluid recycling rate) (Rassenfoss, 2011), environment (threatening groundwater) (Vengosh et al., 2014), and technology (diminishing stimulation efficiency) (Javadpour et al., 2015). Scientists and

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scholars have proposed the following mechanisms that should be responsible for the large amount of fluid-loss: imbibition, electro-chemical forces, fracture fluid entrapment, gravity segregation, clay hydration, and vapor-diffusion (Dehghanpour et al., 2012, Singh, 2016). Besides those factors, the liquid slip in the nanoscale pores of shale formation, which makes the fluid easily infiltrate the ultra-tight shale matrix, is another possible reason to explain the high fluid-loss during injection period (Javadpour et al., 2015). Therefore, modeling and predicting water transport in shale formations with nanopores is the fundamental task not only for understanding whereabouts of the unrecovered fracturing fluid, but also for the possible application in other nanoporous media (Wu et al., 2016a, 2016b).

The pores in shale can be segregated into two types: pores that exist in the inorganic matter (hydrophilic), and pores that exist in the organic matter (hydrophobic) (Li et al., 2016; Li et al., 2018). Consequently, shale is ta typical dual-wettability/mixed-wet porous media (Lan et al., 2015; Yassin et al., 2016; Zolfaghari et al., 2017). Further, the pore networks are mostly at the nanoscale ranging from a few nanometers to several hundred nanometers (Curtis et al., 2012). Since the diameters of these nanopores are comparable to that of liquid molecules, water transport behavior in those pores considerably deviates from that in conventional reservoirs (Afsharpoor and Javadpour, 2016; Afsharpoor et al., 2017). A considerable body of experiments and molecular simulations have shown that the classical no-slip assumption in Poiseuille law, zero fluid velocity on the wall surface, is not well obeyed in the nanoscale channels whether the channel is hydrophobic or hydrophilic, and the liquid structure is strongly affected by the liquid-wall interaction and thus changing the wall boundary condition (Ortizyoung et al., 2013; Lei et al., 2016). In most of previous research, the flow of water are mainly confined in a single nanoscale tube instead of a nanoporous media like the shale. For instance, Holt et al. (2006) measured the flow rates of water transport through carbon nanotubes (diameter of 2 nm) and found the enhancements of up to 8400 over the no-slip Poiseuille flow, giving the slip length between 380 nm and 1.4 um. The slip length value is defined by the extrapolated length where the tangential velocity component disappears. Qin et al. (2011) measured the flow rates of a known smallest carbon nanotube with the diameter in the range of 0.81-1.59 nm and obtained the diameter dependent slip length of 8-53 nm. In addition, the measured slip length by Gruener et al. (2009) for water flow through hydrophilic nanoporous silica with mean pore radius of 3 nm is a negative one. In this paper, we incorporate the fluid flow model in a single nanotube into the complicated heterogeneity and dual-wettability shale matrix system, to obtain the apparent permeability of the shale matrix.

Recently, the reconstructed digital core combined with the Lattice Boltzmann method has been successfully applied to model gas transport in shale gas reservoir considering nanoscale effect but failed to extend to the liquid transport due to the complexity of boundary conditions and unacceptable computational resources (Zhang, 2011; Wang et al., 2016a). The analytical model, based on some approximations and assumptions, not only yields instantaneous calculation results and identifies the impact of each physical variable but also provides general observations and predictions, which is a promising and meaningful method to predict the apparent permeability of the shale reservoirs (Wu et al., 2016a, 2016b; Sun et al., 2017a, 2017b, 2017c; Sun et al., 2018). One of the classical and most widely used analytical method to upscale the flow in conventional reservoir is idealized capillary bundle model benefitting from its simplicity and relative rationalisation (Iii et al., 1981). However, due to the dual-wettability, high tortuosity, disordered and complicated heterogeneity of shale matrix, directly applying the classical capillary bundle model in shale reservoir is difficult and even impossible. Luckily, numerous studies show that the interspaces of the natural porous media including shale gas formation follow statistically fractal scaling laws with the assistance of Scanning Electron Microscopy (SEM) (Krohn, 1988; Yang et al., 2014; Yuan et al., 2016), and fractal theory has been proven to be a powerful tool and successfully adopted to describe the self-similar pore structures of these fractal porous media (Katz and Thompson, 1985; Anderson et al., 1996; Yu and Liu, 2004; Daigle et al., 2015; Geng et al., 2017).

In the present study, based on the water flow behavior in circular nanotube, the fractal scaling theory, and the stochastic upscaling method, the model of apparent water permeability in dual-wettability shale matrix system was obtained in Section 2. The proposed model considers the wettability and pore size related liquid slip effect, TOC content, and the structural parameters of shale matrix. Thirdly, wettability of inorganic and organic matter, organic matter content, and structural parameters on the liquid apparent permeability of shale matrix are discussed, whilst the sensitivity of each structural parameter is analyzed in Section 3. Finally, conclusions are summarized in Section 4.

#### 2. Mathematical modeling

In this section, the water transport behavior in a nanotube is elaborated, and combined with fractal scaling theory (characterize heterogeneity of pore structures), the liquid flow through an elementary volume (organic or inorganic matter) in shale matrix sample is properly described. Furthermore, the apparent liquid permeability (ALP) of shale matrix in sample scale with dual-wettability is obtained by upscaling the flow in each elementary volume using the stochastic method. The model distinguishes the flow mechanisms in inorganic and organic matter and accounts for the effect of the wettability, pore size distribution (PSD), and tortuosity of the each of that.

#### 2.1. Slip-corrected liquid flow in nanotube

The pores in shale matrix system including inorganic and organic pores are mostly at nanoscale (Curtis et al., 2012). Hence, in order to model the flow in shale matrix, the flow behaviors in nanopores need to be characterized first.

Owing to the surface-dominated characteristics of fluid flow in nanoscale pore, the structural and dynamical properties (eg. viscosity) of confined water dramatically deviates from the bulk water. As a result, the conventional continuous flow with no-slip boundary is no longer applicable in both hydrophilic and hydrophobic nanopore surface (Levinger, 2002).

When subjected to an axial pressure driven flow of an incompressible liquid creeping (Reynolds number much less than one) steadily through a circular tube is (White, 2006):

$$q_s = \frac{\pi}{8\mu_{\infty}} [r^4 + 4r^3l_s] \frac{\partial P}{\partial z} \eqno(1)$$

where  $\mu_{\infty}$  is the liquid viscosity; r is the reference radius;  $\partial p/\partial z$  is the pressure gradient;  $l_s$  is the slip length at the liquid/solid boundary. The slip length  $l_s$  describes the velocity discontinuity between the liquid and the solid, is typically defined as the extrapolated length where the tangential velocity component disappears (Joseph and Tabeling, 2005):

$$l_s = \frac{v}{\partial v / \partial r}|_{r=r_0} \tag{2}$$

where v is the axial velocity;  $r_0$  is the radius of the tube.

Predictions from molecular dynamic (MD) simulation indicate that the Poiseuille parabolic form Eq. (1) (continuum approximation) is valid when the characteristic flow diameter is 5–10 times

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