



An analytical equation for oil transport in nanopores of oil shale considering viscosity distribution

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Abstract

Huge amount of works was done on modeling of gas transport in nanopores (both organic and inorganic) of shale formation. However, the study on oil transport behaviors is quite limited. Based on the study on water transport in carbon nanotubes, an analytical model is developed for oil transport in nanopores of shale formation. The new model takes the effect of oil–wall interaction on the oil viscosity in the adsorption region into consideration. Results show that: (1) the oil–wall interaction on oil viscosity in the adsorption region plays an important role in oil transport behaviors and cannot be neglected; (2) when the critical thickness is smaller than 1 nm, the volume flux increases slowly with increasing contact angle; (3) when the critical thickness increases to 2 nm, the volume flux increases rapidly to infinity when the contact angle is larger than 140°.

Keywords Shale oil · Nanopore · Oil transport · Viscosity variation · Critical thickness · Analytical model

Introduction

Great pressure is laid on the energy supply with the rapid rising population (Lee 2011; Sun et al. 2017a, b, c, d, e, f, g, h, i, j; Sheikholeslami et al. 2018a, b, c). Besides, the rapid depletion of conventional petroleum and natural gas resources aggravates the seriousness of the problem (Wang et al. 2018; Jia et al. 2018; Sun et al. 2018a, b, c, d, e, f, g, h, i, j, k, l). Fortunately, the geological reserves of unconventional oil and gas are extremely rich throughout the world, and the development of these resources has become a hot spot (Zhu et al. 2016). Shale oil reserves have been explored worldwide, and the US has seen a boom of oil shale development in recent years (Ribas et al. 2017; Soeder 2018; Alfarge

et al. 2018; Pang et al. 2018). However, the extremely poor connectivity of pores in shale and the ultralow permeability are the main characteristics of shale formation (Sheng et al. 2018; Shovkun et al. 2018). At present, the study on the oil transport mechanisms in nanopores of shale is quite limited (Lu et al. 2012; Falk et al. 2015; Bousige et al. 2016). The key physics of oil transport mechanisms in nanopores of shale are still held as mysteries (Cui et al. 2017).

Modern experimental means face challenges in isolating kerogen from the oil shale, and the critical knowledge about the physical and chemical properties of nanopores in shale is quite limited (Ibrahimov and Bissada 2010; Suleimenova et al. 2014). Therefore, molecular dynamics simulation (MDS) is always adopted in the preliminary study (Kondori et al. 2017; Rafati et al. 2018), and the kerogen is always described as graphene in the shale formation (Ambrose et al. 2012; Mosher et al. 2013; Harrison et al. 2014; Wang et al. 2015a, b, 2016a). Wang et al. (2016b) studied the profile of oil transport rate in nanopores of kerogen and the multi-layer sticking phenomenon by adopting the MDS method. They found that there exists slip flow of oil in inorganic nanopores, which is different from that in kerogen, where some layers of oil are stuck to the nanopore wall. It has been pointed out that the flow enhancement, a ratio of oil transport rate in nanopores of shale to the calculated velocity by the Hagen–Poiseuille model, in organic nanopores of shale can be up to three orders of magnitude (Majumder et al.

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2005). The slip flow and apparent viscosity model is then developed to capture the physics of liquid transport in nanopores (Majumder et al. 2005; Chen et al. 2008). However, for the ideal graphite model, some unique characteristics of shale nanopores (e.g. wall roughness, radius change, hydrophilicity and hydrophobicity and tortuosity) cannot be considered (Schmatko et al. 2005; Bahrami et al. 2006; Joseph and Aluru 2008; Falk et al. 2010; Yang et al. 2015a, b; Gu et al. 2016; Guo et al. 2016; Joly et al. 2016). In fact, MDS is time consuming and cannot describe the surface properties well (Secchi et al. 2016). Besides, empirical equations cannot show the internal relationships among the physical parameters (e.g., the function relationship between sticking viscosity and wettability). The present study on the profile of oil velocity in nanopores is still at the early stage: the factors influencing the velocity profile is not well understood and whether the slip flow will always happen is still waited to be explored. What is worth to mention is that the present modeling methods of slip length and apparent viscosity are not the most appropriate for characterizing (Cui et al. 2017).

It has been pointed out that the viscosity of bulk oil in the center of the nanopores is equal to that of oil in macropores (Cui et al. 2017). As a result, adopting the predicted value of apparent viscosity may not reflect the actual profile of velocity. Besides, the present equations for characterizing the slip length may be physically unrealistic under some conditions (Myers 2011; Cui et al. 2017).

It has been pointed out that the wettability is the dominant factor influencing the liquid transport in nanopores under certain conditions (Neto et al. 2005; Thomas et al. 2009; Li et al. 2010; Ho et al. 2011; Botan et al. 2011; Majumder et al. 2011; Lee et al. 2012; Gruener et al. 2016; Wu et al. 2017; Cui et al. 2017). Mattia and Calabro (2012) presented a model for estimating the slip velocity of water flow in carbon nanotubes. Then, based on Mattia et al.'s work, a series of works were done on modeling of the flow enhancement at nanoscale (Park and Aluru 2007, 2010; Wei et al. 2011; Mattia et al. 2015). At present, in the theoretical study of water transport in nanotubes, the effect of surface diffusion, representing the water–wall interaction, adhesion work, wall roughness and the water–wall wettability were all taken into consideration, and some novel physics were discovered. However, present model shows a relatively poor fitness compared with experimental data (Zhang et al. 2002; Mashl et al. 2003; Kou et al. 2014, 2015; Cui et al. 2017). Ritos et al. (2014) conducted a verification test of the Mattia's model by simulating water transport in nanotubes of various wall materials. However, these previous works were almost focused on water transport in nanotube (mostly carbon nanotubes) (Thomas et al. 2008, 2010; Kannam et al. 2013; Muscatello et al. 2016; Wu et al. 2017). At present, the study on oil transport in nanopores of shale is quite limited.

It has been pointed out that the polar components in crude oil can be adsorbed on the surface of the nanopores and this part of oil is not easy to flow. Therefore, these polar components can be regarded as a part of the shale formation (Schwark et al. 1997; Qin et al. 2000; Pan et al. 2005; Cui et al. 2017). Cui et al. (2017) presented an analytical model for oil transport estimation in nanopores of oil shale with consideration of van der Waals adsorption (McGonigal et al. 1990), and compared the contributions of slip factor and adsorption factor on the flow enhancement. Besides, Cui et al. (2017) found the contribution of physical adsorption is negligible compared with slip flow, which is in agreement with water transport behaviors in carbon nanotubes (Kondratyuk and Yates 2007). For liquid flow through nanopores, the adsorption phenomenon is extremely important, which has an obvious influence on the flow enhancement (Wu et al. 2017). The adsorption region is the area where some layers of fluid are stuck to the nanopore walls and its physical parameters are significantly different from that of bulk liquid (Do and Do 2005; Severson and Snurr 2007; Sha et al. 2008). Riewchotisakul and Akkutlu (2016) presented a model for gas transport in nanopores of shale with consideration of physical adsorption. However, the physical parameters of gas are significantly different from that of liquid. Despite this, the studies on gas transport revealed the importance of physical adsorption on fluid transport at nanoscale (Akkutlu et al. 2012; Deng et al. 2014; Yang et al. 2015a, b; Wu et al. 2015). Zhang et al. (2017a) presented a model for predicting the volume flux of oil in nanopores of shale. However, their model failed to take the effect of oil–wall interaction on the viscosity of oil in the adsorption region into consideration.

In this paper, a modified model is developed for oil transport estimation in nanopores of shale considering spatial variation of oil viscosity. Then, sensitivity analysis is conducted based on the new equation.

Model description

The shale formation is rich in nanopores and nanoslits (Li et al. 2017; Feng et al. 2018a, b, c; Zhang et al. 2017a, b). A cross-section of a SEM image of a shale sample is shown in Fig. 1 below. It is observed that nanopores and nanoslits are distributed in both organic and inorganic materials of the shale formation.

A further study showed the nanopores of kerogen (the organic material) in the shale formation, as shown in Fig. 2 below. It is observed that nanopores (most of them are in cylindrical shape) are rich in abundance in the kerogen (Mohammed et al. 2017; Feng et al. 2018a, b). Therefore, in this paper, a cylindrical model is developed for oil transport in nanopores of shale formation.

Fig. 1 A cross-section of a SEM image of a shale sample: **a** unsegmented and **b** segmented. Reproduced with permission from (Walls and Sinclair 2011; Song et al. 2018)

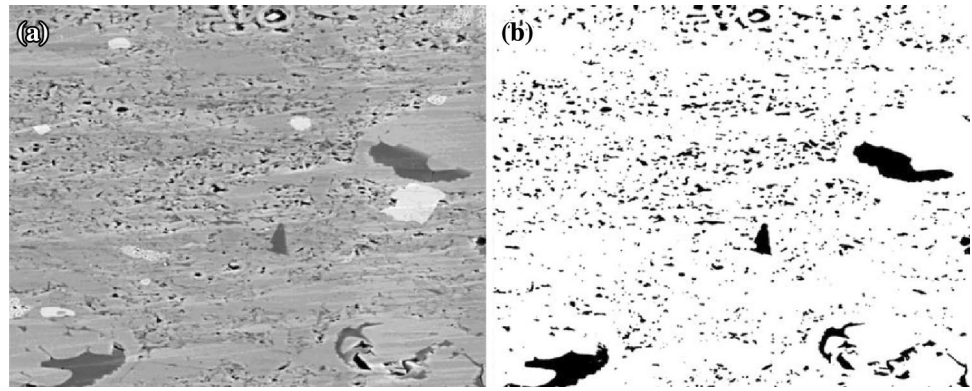
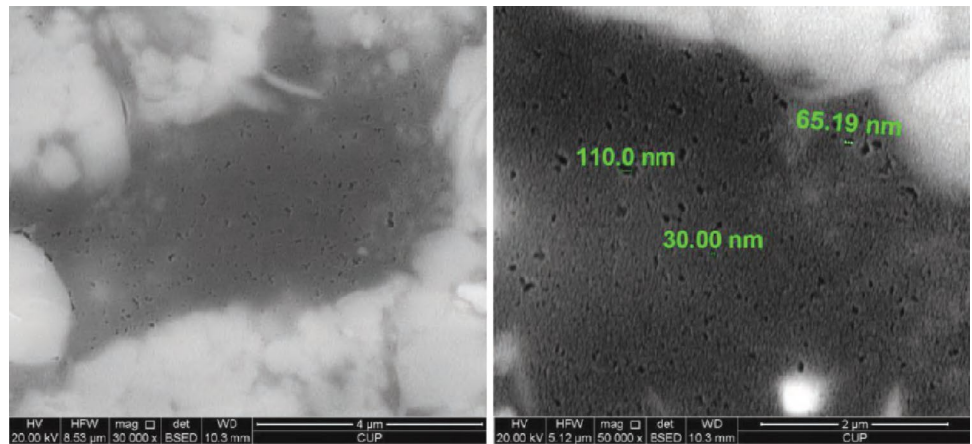


Fig. 2 Structure of nanopores in organic material of shale formation. Reproduced with permission from (Zeng et al. 2017)



Based on both theoretical and MDS research, Zhang et al. (2017a) presented an analytical equation considering multiple mechanisms (e.g. physical adsorption, slip flow and physical and chemical properties of nanopore wall, etc.) of oil transport in nanopores of oil shale. The volume flux in their model is given below (Zhang et al. 2017a; Cui et al. 2017):

$$q_o = \xi_C \left[\frac{\rho_{\text{ads}}}{\rho_{\text{bulk}}} (1 - \xi_{\text{ads}}) + \xi_{\text{ads}} \right] \frac{\pi R^4 \Delta P}{8 \mu L} \quad (1)$$

$$\xi_C = 1 + \frac{8 \mu C_1}{R^2} \quad (2)$$

$$C_1 = \frac{D_S L}{W_A} \quad (3)$$

$$\xi_{\text{ads}} = \left(1 - \frac{h}{R} \right)^2 \quad (4)$$

where q_o denotes the volume flux of oil in nanopores of shale, nm^3/s ; ξ_C denotes the slip factor (Cui et al. 2017), dimensionless; ρ_{ads} denotes the oil density in the adsorption region, g/nm^3 ; ρ_{bulk} denotes the density of bulk oil, g/nm^3 ; ξ_{ads} denotes the factor (Cui et al. 2017), dimensionless; R

denotes the radius of the nanopore, nm ; P denotes the oil pressure, mPa ; μ denotes the oil viscosity, $\text{mPa}\cdot\text{s}$; L denotes the length in the flow direction, nm ; C_1 denotes the coefficient of slip velocity (Cui et al. 2017), dimensionless; h denotes the thickness of the adsorption layer, nm .

As mentioned above, the oil–wall interaction plays an important role in oil properties, especially in the adsorption region. The oil viscosity in the adsorption region will increase or decrease according to the wettability of the nanopore wall. However, the oil viscosity in Eq. (1) is a constant, which leads to deviation to the calculated results. Thomas et al. (2008) proposed an analytical model for estimating effective water viscosity, which is a weighted average of water viscosity in the adsorption region and the bulk water, in nanotubes. In this paper, the model proposed by Thomas et al. is extended to describe the oil viscosity distribution in the nanopores of shale formation. According to Thomas et al.'s model, the effective viscosity of oil confined in the nanopores can be expressed as (Thomas et al. 2008):

$$\mu(r) = \mu_i \frac{A_i(r)}{A_r(r)} + \mu_\infty \left[1 - \frac{A_i(r)}{A_r(r)} \right] \quad (5)$$

where $\mu(r)$ denotes the effective viscosity of oil confined in the nanopores of shale formation, $\text{mPa}\cdot\text{s}$; μ_i denotes the oil

viscosity in the adsorption region, mP s; μ_∞ denotes the viscosity of bulk oil, mP s; $A_i(r)$ denotes the cross-sectional area of the adsorption region, nm^2 ; $A_r(r)$ denotes the cross section area of nanopore, nm^2 .

In this paper, it is assumed that Eq. (5), developed for water transport in carbon nanotubes, can also be used to calculate the oil viscosity in the nanopores of shale formation. This is because the lipophilicity or oleophobicity properties of an oil–wall system can lead to increase or decrease of oil viscosity in the surface region, which is similar to a water–wall system (Cui et al. 2017).

Wu et al. (2017) proposed an analytical equation for estimating water viscosity in the adsorption region based on MDS and experimental results. In this paper, we also assume that the equation developed for water viscosity calculation is effective in oil viscosity calculation. The empirical equation can be expressed as (Wu et al. 2017):

$$\frac{\mu_i}{\mu_\infty} = -0.018\theta_w + 3.25 \tag{6}$$

where θ_w denotes the contact angle of a water droplet on the surface of a certain material, $^\circ$.

Given the fact that the components of crude oil and the mineral compositions of shale sample are much more complicated than that of pure water and carbon, Eq. (6) is rewritten with two fitting parameters. The expression for oil transport in nanopores of shale or tight formation can be expressed as:

$$\frac{\mu_i}{\mu_\infty} = -C_2\theta_o + C_3 \tag{7}$$

where θ_o denotes the contact angle of an oil–wall system, $^\circ$.

Combining Eqs. (7) and (5), we can obtain:

$$\mu(r) = (-C_2\theta_o + C_3)\mu_\infty \frac{A_i(r)}{A_r(r)} + \mu_\infty \left[1 - \frac{A_i(r)}{A_r(r)}\right] \tag{8}$$

Then, combining Eqs. (8) and (1), we can obtain:

$$q_o = \xi_C \left[\frac{\rho_{ads}}{\rho_{bulk}} (1 - \xi_{ads}) + \xi_{ads} \right] \frac{\pi R^4 \Delta P}{8L} \frac{1}{(-C_2\theta_o + C_3)\mu_\infty \frac{A_i(r)}{A_r(r)} + \mu_\infty \left[1 - \frac{A_i(r)}{A_r(r)}\right]} \tag{9}$$

Equation (9) is the analytical equation for estimating oil transport behaviors in nanopores of shale formation.

Discussion

In this section, the key findings of this work are discussed in detail. The relationship between the volume flux and contact angle is shown in Fig. 3 below. Given the fact

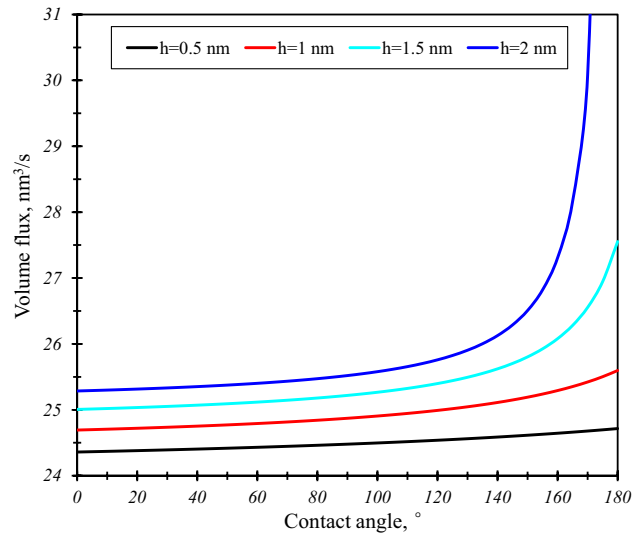


Fig. 3 The relationship between volume flux and contact angle under different critical thickness conditions

that the composition of crude oil varies significantly to each other, the curves under various critical thickness are shown for comparison.

It is observed from Fig. 3 that when the critical thickness is smaller than 1 nm, the volume flux increases slowly with increasing contact angle. However, when the critical thickness increases to 2 nm, the volume flux increases rapidly to infinity when the contact angle is larger than 140°. This means the effect of wettability on oil transport only make a contribution when the critical thickness is larger than 1.5 nm.

Conclusions

In this paper, an improved model is developed with consideration of the effect of oil–wall interaction on the oil

viscosity in the adsorption region. Then, the effects of physical adsorption and wettability on the oil transport in nanopores are discussed in detail. Some meaningful conclusions are listed below:

- (1) The oil–wall interaction on oil viscosity in the adsorption region plays an important role in oil transport behaviors and cannot be neglected.

- (2) When the critical thickness is smaller than 1 nm, the volume flux increases slowly with increasing contact angle.
- (3) When the critical thickness increases to 2 nm, the volume flux increases rapidly to infinity when the contact angle is larger than 140°.

Following researchers are suggested to perform more studies on the physics in the surface region to obtain a satisfactory oil recovery efficiency of the oil shale reserves.

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