



Apparent permeability for liquid transport in nanopores of shale reservoirs: Coupling flow enhancement and near wall flow



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ABSTRACT

Multiple mechanisms of oil transport in inorganic and organic nanopores of shale oil reservoirs are still unclear and possibly more complex than those of gas transport in nanoporous media, due to differences of molecules free path and fluid-solid molecular interactions. The accurate apparent permeability model considering oil transport mechanisms and different pore types is important for macroscale modeling in shale oil reservoirs development. Based on studies of molecular dynamics simulations (MDS), liquid flow through carbon nanotubes (CNTs) and theoretical analysis, a unified apparent permeability model of liquid hydrocarbon flow in the shale is derived coupling different transport mechanisms in inorganic and organic nanopores. The model of oil-wet organic nanopores considers liquid-solid adsorption, while the model of water-wet inorganic nanopores incorporates near wall flow and velocity slip. We then introduce complicated structural parameters including the tortuosity, porosity and total organic carbon (TOC) to develop models from nanotubes into porous media. After that, the proposed model is validated by MDS and experimental results, and the total apparent liquid permeability (ALP) as well as contributions of different mechanisms are studied. The results indicate that, flow enhancement should be considered in the characterization of oil transport in nanopores, and the velocity of oil in inorganic nanopores much faster than that in organic nanopores in this work. For pore radii under 10 nm, the total ALP is much larger than intrinsic permeability, and adsorption effect as well as velocity slip in organic matter (OM) and inorganic matter (IM) influence the total ALP slightly when the pore radius is larger than 100 nm. In addition, the greater slip length in IM results in greater contributions of oil transport in IM to the total ALP if slip length is less than 10 nm. Moreover, the ratio of the total ALP to intrinsic permeability decreases as TOC increases when TOC is larger than 20%. This work focuses on enriching the theoretical research of oil transport in nanopores and provides a unified ALP model for macroscale modeling study in the shale reservoirs development.

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1. Introduction

Shortage of conventional resources has accelerated the development of unconventional resources, such as shale gas and oil reservoirs. Shale oil with rich reserves, great potential and especially the efficient development in North America and China, has gained an increasing amount of worldwide attention [1]. However, the distinguishing characteristics of shale oil from other unconventional resources are mainly caused by the hydrocarbon accumulation mechanism and reserving space types. Similar to shale gas reservoirs, oil is stored in both organic and inorganic pores with nanoporosity and extreme low permeability, which makes oil

transport mechanisms in shale difficult to clearly understand [2,3]. Meanwhile, Darcy's law cannot be used to describe liquid transport in shale. Currently, limited studies focusing on oil transport in the nanotube are almost based on MDS [4,5]. Therefore, a further understanding of oil transport mechanisms in different nanopore types is urgently needed, and a mathematical model of ALP coupling multiple mechanisms is essential for macroscale numerical simulation, production forecast and optimization [6,7].

Experimental studies, such as Atomic force microscopy (AFM) and scanning electron microscopy (SEM) used to characterize shale pore structures, suggest that nanopores ranging in size from 1 to 200 nm are abundant presence, and most frequent pore radii are generally smaller than 5 nm [8]. Through X-ray diffraction (XRD) mineralogy analysis, typical components of shales can be divided into OM (oil-wet kerogen) and IM (water-wet quartz, clays et al.)

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Nomenclature

CNT (s)	carbon nanotube (s)	q_{in}	mass flux in inorganic pores (kg/s)
MDS	molecular dynamics simulation (s)	q_b	mass flux of bulk oil (kg/s)
AFM	atomic force microscopy	q_w	mass flux of near wall oil (kg/s)
SEM	scanning electron microscopy	q_{or}	mass flux in organic pores (kg/s)
XRD	X-ray diffraction	R	average pore radius (m)
OM	organic matter	r	radial distance from the pore center (m)
IM	inorganic matter	T	Temperature (K)
LBM	lattice Boltzmann method	v	velocity profile at r (m/s)
HP	Hagen-Poiseuille	v_b	bulk oil velocity at r (m)
ALP	apparent liquid permeability	v_w	near wall oil velocity at r (m/s)
TOC	total organic carbon	W_A	adhesion work (J/m ²)
C	slip velocity coefficient in inorganic pores (dimensionless)	α	total organic carbon (dimensionless)
C_{or}	slip velocity coefficient in organic pores (dimensionless)	δ	near wall oil thickness (m)
c_1	unknown related to slip velocity (m/s)	δ'	adsorbed oil thickness (m)
c_2	unknown related to slip velocity (m/s)	κ_B	Boltzmann's constant (J/K)
D_s	surface diffusion coefficient (m ² /s)	λ	slip length (m)
J_{or}	total mass flux in OM (kg/(m ² s))	λ_b	bulk factor (dimensionless)
J_{in}	total mass flux in IM (kg/(m ² s))	λ_c	slip factor (dimensionless)
J	total mass flux in the porous medium (kg/(m ² s))	μ	oil viscosity (mPa·s)
J_v	mass flux in the porous medium with no-slip (kg/(m ² s))	μ_{ads}	adsorbed oil viscosity (mPa·s)
k_{ins}	intrinsic permeability of the porous medium (m ²)	μ_{bulk}	bulk oil viscosity (mPa·s)
k_{or}	apparent permeability of organic pores (m ²)	μ_o	average oil viscosity in porous medium (mPa·s)
k_{or_b}	apparent permeability of bulk oil on in organic pores (m ²)	μ_w	average oil viscosity in near wall region (mPa·s)
$k_{or_{ads}}$	apparent permeability of adsorbed oil in organic pores (m ²)	ξ_{ads}'	correction factor for adsorbed oil (dimensionless)
k_{app}	total ALP of shale reservoirs (m ²)	ξ_b	correction factor for bulk oil in IM (dimensionless)
k_{in}	apparent permeability of inorganic pores (m ²)	ξ_b'	correction factor for bulk oil in OM (dimensionless)
k_{in_b}	apparent permeability of bulk oil in inorganic pores (m ²)	ξ_w	correction factor for near wall oil (dimensionless)
k_{in_w}	apparent permeability of near wall oil in inorganic pores (m ²)	ρ_w	average density of near wall oil (kg/m ³)
L	pore length (m)	ρ_b	average density of bulk oil (kg/m ³)
n_L	number of molecules per unit adsorbed volume (dimensionless)	ρ_o	average oil density in the porous medium (kg/m ³)
Δp	differential pressure (MPa)	ρ_{ads}	average adsorbed oil density (kg/m ³)
Q_b	volume flux of bulk oil (m/s)	τ	tortuosity of the porous medium (dimensionless)
Q_w	volume flux of near wall oil (m/s)	ϕ	porosity of the porous medium (dimensionless)
		ϕ_{ads}'	effective porosity for adsorbed oil (dimensionless)
		ϕ_b	effective porosity for bulk oil in IM (dimensionless)
		ϕ_b'	effective porosity for bulk oil in OM (dimensionless)
		ϕ_w	effective porosity for near wall oil (dimensionless)

as shown in Fig. 1 [9], in which black spots in the dark gray area represent nanopores within OM, while the light dark area is IM composed of quartz, clays et al. Up to now, the gaseous methane transport behavior through porous shale media has been mentioned by researchers using experimental [10] and numerical methods, like MDS [11] and the lattice Boltzmann method (LBM) [12]. Several types of apparent gas permeability models based on Knudsen number and microscale pores have been presented for the macroscale flow simulation [2,13–16]. Different from the gaseous methane, mean free path of liquid molecule is greatly smaller than that of gas molecule, in addition to stronger liquid-solid molecular interactions [17], which might lead to more complex transport behaviors. However, the studies of oil transport mechanisms in nanopores are still limited, especially oil transport behavior in OM [4,18].

Like gas slippage, liquid slippage has also been confirmed to exist between liquid molecules and pore inner wall by experimental methods and MDS [19]. Studies about liquid flow in nanotubes suggest that the slip boundary condition is related to wettability and significantly different from the no-slip boundary condition [4,19–27]. Whitby and Quirke [20] reviewed some experimental results with an emphasis on fluid flow through CNTs, and investigated that water flow rates through CNTs were 560–8400 times

higher than those predicted from the macroscale no-slip Hagen-Poiseuille (HP) equation. Hence, the enhancement factor is defined as the ratio of flow rate in a CNT to no-slip HP flow [21]. Majumder et al. [22] observed that the enhancement flow rates of water, hexane, ethanol and alkanes through the multiwalled carbon nanotubes (MWNT) composite membranes were 4–5 orders of magnitude greater than those calculated by HP equation. Their results also show that slip length from 3 to 70 μm is much longer than the tube diameter (7 nm), and decreases as the liquids become more hydrophobic. Holt et al. [23] measured water transport velocity through microfabricated membranes composed of CNTs with <2 nm diameter, and the enhancement factor was calculated to be 3 orders of magnitude. Recently, experimental results have involved in a wide range of CNT diameter from 0.8 to 44 nm, CNT length from 2 to 280 μm , and pressure condition from 0.1–100 MPa [18,22–25]. However, owing to the difficulties of experiments, engineers and scientists focus on the application of MDS to study hydrodynamic properties of fluid transport in nanopores and the near wall region. Barrat and Bocquet [26] used extensive MDS to study hydrodynamic properties of fluid in microscale pores with slip and no-slip boundary conditions, and found slip length exceeding 30 molecular diameters in mercury on glass system. Falk et al. [27] applied MDS to calculate the friction

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