



Full Length Article

An analytical model for shale gas transport in kerogen nanopores coupled with real gas effect and surface diffusion



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ABSTRACT

Understanding the behavior of shale gas transport in kerogen is a key issue in predicting gas production. The reservoir structure is characterized by widespread micro/nanoscale pores, various occurrence states, and typical high pressure. An analytical model is proposed to effectively reveal the gas transport behavior in kerogen nanopores. The model can fully consider the real gas effect, gas slippage, and surface diffusion derived from adsorbed gas. In particular, a method based on dense gas theory with the Redlich–Kwong equation of state is used to acquire the viscosity of shale gas under high pressure. The second-order slippage boundary condition coupled with surface diffusion is presented to describe the free gas slippage, and Langmuir isotherm theory and Fick's law are adopted to calculate the surface diffusion. The real gas effect has a significant effect on the physical properties of methane, Knudsen number, and the flow behaviors of free gas and adsorbed gas. The surface diffusion velocity can enhance the free gas flow. The mass flow rate of total adsorbed gas increases as pore size increases, and its major influence is obtained from the induced free gas at the increased pore size. The slippage effect is reduced as the pressure increases and the temperature decreases. The adsorbed gas comprises a substantial proportion of the total gas produced when the pore size is less than 2 nm. The combined influences of slippage effect and adsorbed gas cannot be ignored when the pore size is less than 10 nm. This work provides a comprehensive and theoretical guidance for the effective development of shale gas.

1. Introduction

Shale gas is a type of unconventional natural gas that has been extensively produced from fine grain shale rocks in recent years with improvements of horizontal drilling and hydraulic fracturing technology [1]. Shale gas, whose main content is methane, is considered a potential energy in the future because of its abundant reserves, long production life, and environment-friendly qualities. Gases in shale have three states: free gas in fractures and matrix pores, adsorbed gas on the surfaces of kerogen pores and clay particles, and few dissolved gas in kerogen bulk and bitumen [2–4]. Free gas is the major component to determine gas-in-place in shale reservoirs. Adsorbed gas becomes a dominant component during the late period of the gas production process to determine the longevity of shale-gas-producing wells [5].

Unlike conventional hydrocarbon reservoirs, shale strata have considerable microscale and nanoscale pores that exhibit extremely low permeability on the order of nanoDarcies [6]. The pore size of shale is usually less than 2 μm and is smaller in kerogen, ranging from 2 nm to 50 nm, to enable the slippage effect [7,8]. Shale gas is deposited as supercritical fluid at an underground depth of 3000–6000 m with high

pressure (up to 60 MPa) [9]. The volume of gas molecules cannot be overlooked and the intermolecular interaction is remarkable. Therefore, the real gas effect has a significant influence on gas transport. In kerogen nanopores, a large proportion of gas is adsorbed on the pore surface in the form of a monolayer, which can be described by Langmuir adsorption theory [10]. The concentration gradient drives the transport of the monolayer adsorbed gas, which is called surface diffusion [11,12]. The adsorbed gas reduces the flow space of free gas and couples with the free gas in the transport process [13]. During the gas production process, the gas flow properties are nonlinear, and similar phenomena can be found in furrow irrigation, evapotranspiration, rainfall forecasting [14–19], which poses more difficulties in gas yield prediction.

Several studies have been performed to identify the shale gas transport characteristics in micro and nanoscale pores. In the pioneering studies, the ideal gas model combined with rarefied gas theory is one of the popular methods in which the mass flux of total gas is treated as the superposition of slip flow and Knudsen diffusion. The slip flow refers to the viscous flow with a slippage boundary. The Knudsen diffusion is attributed to the interaction between gas molecules and

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Nomenclature*List of Symbols*

| | |
|----------------|--|
| C_1, C_2 | Slippage coefficients |
| C_μ | Adsorbed gas amount, [kg/m ³] |
| d | Molecular diameter, [m] |
| D_s | Surface diffusion coefficient, [m ² /s] |
| dp/dx | Pressure gradient, [Pa/m] |
| H | Pore size, [m] |
| H_a | Thickness of adsorbed layer, [m] |
| H_f | Thickness of free layer, [m] |
| Kn | Knudsen number |
| Kn_f | Effective Knudsen number |
| L | Length of the channel, [m] |
| m | Mass of molecular, [kg] |
| \bar{M} | Molar mass, [kg/mol] |
| \dot{M} | Mass flow rate of total shale gas, [kg/s] |
| \dot{M}_a | Mass flow rate of pure adsorbed gas, [kg/s] |
| \dot{M}_f | Mass flow rate of pure free gas, [kg/s] |
| \dot{M}_{fa} | Mass flow rate of induced free gas, [kg/s] |
| \dot{M}_a^* | Mass flow rate of total adsorbed gas, [kg/s] |
| n | Number density of gas molecules, [1/m ³] |
| N_A | Avogadro's number, $N_A = 6.02 \times 10^{23}$, [1/mol] |
| p | Pressure, [Pa] |
| p_{cr} | Critical pressure, [Pa] |
| p_L | Langmuir pressure, [Pa] |
| q_L | Langmuir volume, [m ³ /kg] |
| R | Universal gas constant, [J/(mol·K)] |
| R_{ig} | Ideal gas constant, $R_{ig} = 8.314$, [J/(mol·K)] |
| T | Temperature, [K] |

| | |
|------------|--|
| T_{cr} | Critical temperature, [K] |
| u_f | Free gas velocity without adsorbed gas, [m/s] |
| u_f^* | Free gas velocity with adsorbed gas, [m/s] |
| u_s | Slip velocity without adsorbed gas, [m/s] |
| u_a | Adsorbed gas velocity in the x direction, [m/s] |
| V_m | Molar volume, [m ³ /mol] |
| $V_{m,cr}$ | Critical molar volume, [m ³ /mol] |
| V_{sid} | Gas volume per mole at standard condition, [m ³ /mol] |
| Z | Compressibility factor |
| Z_{cr} | Critical compressibility factor |

Greek symbols

| | |
|-------------------|---|
| η | Molecular volume ratio, $\eta = 2\pi n d^3/3$ |
| θ | Gas coverage of real gas |
| $\bar{\lambda}$ | Mean free path, [m] |
| $\bar{\lambda}_f$ | Mean free path of real gas, [m] |
| μ | Dynamic viscosity of real gas, [Pa·s] |
| μ_{ig} | Dynamic viscosity of ideal gas, [Pa·s] |
| μ_0 | Dynamic viscosity at low pressure, [Pa·s] |
| ρ_a | Average density of adsorbed gas, [kg/m ³] |
| ρ_f | Free gas density, [kg/m ³] |
| ρ_s | Organic solid density, [kg/m ³] |
| χ | Correction factor |

Subscripts

| | |
|----|----------------|
| a | Adsorbed gas |
| cr | Critical state |
| f | Free gas |

constraint surface. It occurs when the pore size is very small and the pressure is low [20]. According to the magnitude of Knudsen number (Kn , the ratio of mean free path to characteristic length), the four different flow regions are determined. They include the continuous flow ($Kn < 0.001$), slip flow ($0.001 < Kn < 0.1$), transition flow ($0.1 < Kn < 10$), and free molecular flow ($Kn > 10$). Javadpour [21] combined the slip and Knudsen flows to analyze the effects of pressure, temperature, and pore size on the apparent permeability of shale and Knudsen diffusion in mudrocks. He found that the apparent permeability of shale increased sharply when the pore size was less than 100 nm and the influence of Knudsen diffusion was high with small pore size and low pressure. Subsequently, Sun et al. [22] used the similar method and found that the effect of Knudsen diffusion can be ignored when the equivalent hydraulic radius of the pore was greater than 100 nm. When shale gas flows in micro and nanoscale pores under the typical reservoir condition, the flow regions usually fall into slip and weak transition regions ($0.001 < Kn < 1$), where the slippage effect dominates and the Knudsen diffusion can be ignored [23,24]. The aforementioned studies overestimated the mass flow rate because they included the Knudsen diffusion. The real gas effect was also neglected.

The real gas effect actually has two functions. First, it changes the physical properties of gases and the Knudsen number, thereby affecting free gas flow, and then determines gas coverage, which affects the diffusion characteristics of adsorbed gas [9]. Several studies have been performed to realize the significance of the real gas effect in micro and nanoscale flows. On the basis of gas kinetic theory, Wang et al. [25] modified the molecular mean free path by using the collision correction factor to show the effect of real gas on Knudsen number. Ma et al. [26] applied the Van der Waals equation to correct the physical properties of shale gas, but they ignored the effect of real gas on the Knudsen number. The two studies only considered one part of the real gas effect. Afterward, Wang et al. [12] and Wu et al. [27] employed the gas

compressibility factor derived from complex empirical correlations to incorporate the real gas effect on Knudsen number and the physical properties of gases. However, different empirical formulas obtained different results, and these complicated approaches lack universality. Despite the vital importance of adsorbed gas, few scholars incorporate it or deal with its effect properly in their studies. Kazemi and Takbiri-Borujeni [28] reported that the adsorbed layer reduced the cross-sectional area of the channel, but the interaction of free gas with adsorbed molecules was not included. Ren et al. [29] simulated the effect of the adsorbed gas on the mass flux of the total gas mass, but they only focused on a special condition.

The shale gas transport mechanisms in kerogen nanopores are extremely complex due to widespread micro and nanoscale pores, various occurrence states, and typical high pressure. To intensively study these typical characteristics, the influences of real gas effect, gas slippage, and surface diffusion should be entirely considered. The aforementioned studies majorly partially involved in these influences. A comprehensive and systematic model considering real gas effect, gas slippage, and surface diffusion is needed. In the current study, a new gas transport model is proposed to comprehensively investigate the gas slippage, real gas effect, and surface diffusion. The effects of basic parameters, such as temperature, pressure, and pore size, on dynamic viscosity, Knudsen number, velocity profiles of free gas, adsorbed gas, and apparent permeability are discussed. Moreover, the differences of the real gas model and the conventional ideal model, which is based on the ideal gas equation, are compared.

2. Physical problem description

When shale gas is confined in kerogen nanopores, the free gas exists in the middle of the pore space and the adsorbed gas is adsorbed on the surface. The thickness of the adsorbed layer is comparable to the pore

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