



A new formulation of apparent permeability for gas transport in shale



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ABSTRACT

Recently, a few apparent permeability formulations have been proposed for gas transport in tight formations aiming of correcting flow deviation caused by slip flow and Knudsen diffusion. However, the effect of surface diffusion has always been neglected. Surface diffusion happens when gas molecules adsorb on the solid surface. Considering significant proportion of adsorbed gas (20%–80%) in shale, surface diffusion could play an important role during gas transport. In this paper, we presented a new apparent permeability formulation, which takes surface diffusion into account, for gas transport in shale. The parametric study of the presented formulation shows apparent permeability is sensitive to pressure, temperature, pore radius and surface roughness. The permeability caused by surface diffusion can be several times larger than shale intrinsic permeability at low pressure. Neglecting surface diffusion in the formulation would underestimate gas flux by 25%.

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1. Introduction

Due to tremendous amount of gas in place and development of hydraulic fracturing technology, the increasing shale gas production has aroused great interest in the world (Gunningham, 2014; Jaffe, 2010; Jing et al., 2011; Wang et al., 2014), which also makes the study of gas transport mechanisms in such ultra-tight rock a very hot topic.

Shale is considered as fine grained compacted sediments with ultra-low permeability (several to hundreds of nano-Darcy) and lots of nano-pores (Nelson, 2009). Multi-flow mechanisms happen when gas transport in such media, such as slip flow and Knudsen diffusion. Sorption also happens in organic rich shale, which would lead to surface diffusion (Freeman et al., 2011). Therefore, conventional assumption of continuum flow is no longer valid for gas transport in shale.

To account for the multi-flow mechanisms in shale, several investigators have presented a formulation of apparent permeability to substitute the intrinsic permeability in Darcy's law. Javadpour (2009) combined slip flow and Knudsen diffusion into gas flux equation and derived an apparent permeability

formulation by comparing the new flux equation to Darcy's law. Civan (2010), Florence et al. (2007), Sakhaee-Pour and Bryant (2012) derived a general expression for apparent gas permeability in terms of Knudsen number based on the Hagen-Poiseuille-type equation proposed by Beskok and Karniadakis (1999). Freeman et al. (2011) used Dusty-gas model to account for Knudsen diffusion in shale. Clarkson et al. (2012) presented an apparent permeability formulation using the dynamic-slip concept. Niu et al. (2014) presented a second order gas apparent permeability formulation in which methane molecular accumulation effect near the wall of nano-tubes was considered.

However, all above apparent permeability formulations were used to correct the flux deviation caused by slip flow and Knudsen diffusion, the influence of surface diffusion has been neglected.

Many experiments and modeling work have been conducted to investigate surface diffusion in porous media. Surface diffusion can be experimentally observed and probed in porous media by nuclear magnetic resonance methods, and the surface diffusivity is found to be dependent on surface coverage (Medved and Černý, 2011; Valiullin et al., 2005; Dvoyashkin et al., 2009; Do et al., 2001). In earlier studies, gas flux due to surface diffusion was written in the form of Fick's law

$$J_S = -\rho_l D_S \nabla C_S$$

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The diffusivity D_S increases substantially with C_s (Barrer, 1941; Gilliland et al., 1974). Darken (1948) assumed transport of adsorbed species is driven by its gradient in the chemical potential, and derived surface diffusion coefficient as

$$D_S = D_{S0} \frac{\partial \ln p}{\partial \ln q} \quad (1)$$

here, D_{S0} is called corrected diffusivity, and is independent of surface coverage. For systems where adsorbed concentration can be described by Langmuir isotherm, D_S can be reduced as

$$D_S = D_{S0} \frac{1}{1 - \theta}$$

here θ is surface coverage. The same expression has been produced by Higashi et al. (1963) which is known as HIO model, using a random hopping mechanism.

For the complexity of surface diffusion, many other models also have been developed, such as multilayer theories (Okazaki et al., 1981; Chen and Yang, 1993), hydrodynamic model (Gilliland et al., 1958; Petropoulos, 1996), statistical-mechanical approach (Popielawski and Baranowski, 1965; Hwang and Kammermeyer, 1966; Lee and O'Connell, 1975) and so on (Do et al., 2001). These models have their own advantages to describe surface diffusion. However, they are not suitable to be applied in natural gas industry because of the parameters in the model can't be easily obtained in practice.

Surface diffusion must play an important role during gas transport in shale, because of the significant amount of adsorbed gas in shale (20%–80% of gas in place, as reported by Hill and Nelson, 2000). In this paper, we propose a new formulation of apparent permeability for gas transport in shale, which is able to account for surface diffusion. For the sake of ease of use in reservoir simulation, Darken's model is used to describe surface diffusion and is extended to be suitable for porous media. The new formulation is verified by rigorously analyzed experimental data. The importance of surface diffusion and parametric study of the new apparent permeability formulation are investigated after.

2. Derivation of apparent permeability formulation

In this section, the gas flux due to various flow mechanisms are introduced, and extended to make them better suit for describing gas flow in porous media. Then the flux are added to account for different flow mechanisms. The apparent permeability is derived by comparing the final expression of flux with traditional Darcy's law.

2.1. Gas flow due to pressure force and Knudsen diffusion

Gas flow in porous media is traditionally described by Darcy's law:

$$J_D = -\frac{k_0 \rho_g}{\mu} \nabla p \quad (2)$$

which is capable of describing gas flow due to pressure force in conventional reservoirs. While in unconventional shale reservoirs, multi-flow mechanisms happen during gas transport, therefore Darcy's law is no longer suitable.

To account for multi-flow mechanisms, Javadpour (2009) presented a new formulation to describe gas transport in a single straight cylindrical nanotube:

$$J = - \left[D_K + F \frac{r^2 \rho_g}{8\mu} \right] \frac{\Delta p}{L} \quad (3)$$

The first term in the square brackets of Equation (3) represents Knudsen diffusion coefficient which can be expressed as:

$$D_K = \frac{2r}{3} \left(\frac{8RT}{\pi M} \right)^{0.5} \quad (4)$$

The second term represents gas flow due to pressure forces, and F is correction coefficient due to slip boundary of the nanotube

$$F = 1 + \sqrt{\frac{8\pi RT}{M}} \frac{\mu}{pr} \left(\frac{2}{\alpha} - 1 \right) \quad (5)$$

Javadpour (2009) successfully matched this model with the experimental flux data, which is measured from Argon flow in a commercial Alumina membrane with the average pore size of 200 nm (Roy et al., 2003), with an average error of 4.5%.

The Javadpour's expression is derived from gas flow in a single straight cylindrical nanotube. To extend this expression to be suitable for gas flow in natural porous media, the effect of properties, such as porosity (ϕ), tortuosity (τ), etc, should be considered. Amann-Hildenbrand et al. (2012) considered the effect of porosity/tortuosity factor (ϕ/τ) and pore-surface roughness on the Knudsen diffusion and derived a new Knudsen diffusion coefficient:

$$D'_K = \frac{\phi}{\tau} (\delta')^{D_f - 2} D_K \quad (6)$$

Therefore, the flux due to Knudsen diffusion can be written as:

$$J_K = -\frac{1}{RT} D'_K \frac{\Delta p}{L} \quad (7)$$

Similarly, introducing porosity/tortuosity factor to gas flux due to pressure force, yields:

$$J_p = -F \frac{r^2 \rho_g \phi}{8\mu\tau} \frac{\Delta p}{L} \quad (8)$$

Gas flux in natural porous media due to pressure force and Knudsen diffusion can be expressed as the summation of Equations (7) and (8):

$$J = - \left[\frac{2r\phi}{3RT\tau} (\delta')^{D_f - 2} \left(\frac{8RT}{\pi M} \right)^{0.5} + F \frac{r^2 \rho_g \phi}{8\mu\tau} \right] \frac{\Delta p}{L} \quad (9)$$

2.2. Gas flow due to surface diffusion

Surface diffusion is defined as the process that particles jumping between adjacent adsorption sites on a surface. In gas–solid systems, the surface diffusion contribution is often more profound than liquid–solid systems (Miyabe and Guiochon, 2010). Gas flux due to surface diffusion in porous media for a pure gas can be written as (Keizer et al., 1988; Uhlhorn et al., 1989):

$$J_S = -\rho_l \frac{1 - \phi}{\tau} D_S \nabla q \quad (10)$$

Assume temperature keeps constant and q increases monotonically with pressure in porous media, Darken's surface diffusivity D_S can be rewritten as:

$$D_S = D_{S0} \frac{q}{p} \frac{\partial p}{\partial q} \quad (11)$$

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