



Research paper

Unifying diffusion and seepage for nonlinear gas transport in multiscale porous media

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ABSTRACT

We unify the diffusion and seepage process for nonlinear gas transport in multiscale porous media via a proposed new general transport equation. A coherent theoretical derivation indicates the wall-molecule and molecule-molecule collisions drive the Knudsen and collective diffusive fluxes, and constitute the system pressure across the porous media. A new terminology, nominal diffusion coefficient can summarize Knudsen and collective diffusion coefficients. Physical and numerical experiments show the support of the new formulation and provide approaches to obtain the diffusion coefficient and permeability simultaneously. This work has important implication for natural gas extraction and greenhouse gases sequestration in geological formations.

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1. Introduction

Gas transport in porous media is a topic of much scientific interest and practical importance in several fields, including natural gas flow through subsurface formation, catalysis and mass separation [1–5]. As a particular interest, the gas-bearing subsurface formation exhibits multiscale porous characteristics with pore size ranging from macroscale (>1 mm) to nanoscale (<100 nm) [6,7]. In this context, gas travels by way of seepage and diffusion processes [8]. Darcy's law is the standard approach to study the gas flow in porous media [1–8]. It simply assumes a global permeability k that relates the average macroscopic gas velocity v with the pressure gradient ∇p across the media,

$$v = -\frac{k}{\mu} \nabla p, \quad (1)$$

where μ is the gas viscosity. The permeability k is a macroscopic representation to reflect between the complex pore structure and gas flow. Darcy's law was initially proposed experimentally by Darcy [9] and can be theoretically derived from Navier-Stokes equation [10–12]. Eq. (1) governs the linear gas seepage process and its alone is sufficient for gas transport in porous media as long as the flow is continuous. However, this assumption may not hold anymore for nanoporous media, suggesting merely Darcy's law may not be sufficient for gas transport process therein. Along with the

Darcy flux, the diffusive flux may have increasing contribution to the total gas flux when the porous media changes from macroscale to nanoscale. As a measure of the degree of rarefaction of gas encountered in gas flow through small porous structure, the Knudsen number (K_n) is introduced to characterize the gas flow regime into four categories: Knudsen flow ($K_n > 10$), transition flow ($0.1 < K_n < 10$), slip flow ($0.01 < K_n < 0.1$) and continuum flow ($K_n < 0.01$) [13,14]. Darcy's law is sufficient for continuous flow when $K_n < 0.01$. Thanks to the addition of diffusive flux, the gas transport process through porous media becomes nonlinear as K_n increases from 0.01.

In previous studies, Klinkenberg approach is used to modify permeability to consider the gas slippage effect [10,11,15]. Based on the Dusty-Gas model, Song et al. introduced a new general equation (Eq. (2)) to incorporate the four gas transport regimes by modifying the gas slip factor [11,16–19]

$$v = -\left(\frac{D}{p} + \frac{k}{\mu}\right) \nabla p, \quad (2)$$

where v is the total gas flux and D is the diffusion coefficient. However, the physical meaning of D is rather cryptical and there is no coherent theoretical framework to support Eq. (2) nor to simultaneously and quantitatively characterize the gas seepage and diffusion fluxes separately using Eq. (2).

In this Letter, we report our investigation of the nonlinear gas transport through multiscale porous media (from macroscale to nanoscale) theoretically via the kinetic molecular theory and experimentally by physical measurement and direct numerical simulation. We not only provide a coherent theoretical derivation

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and explanation of Eq. (2), but also an approach to characterize the gas seepage and diffusion fluxes simultaneously and quantitatively. The physical and numerical experiments also complement as a support to the credibility of the new formulation.

2. Methodology

Without losing generality, we consider the single component gas flow scenario, such as methane. The gas molecules transport through the porous media via three physical mechanisms: Knudsen diffusion, collective diffusion and advection. Knudsen and collective diffusions are similar in that they both are induced by molecular density gradient, while the difference is in the dominant type of molecular collision mechanisms of each [20,21]. Knudsen diffusion is in the free-molecular region with minimal molecule-molecule collisions; it is the statistical representation of molecule-wall collisions due to the scale of porous path is comparable to or smaller than the mean free path of the molecules therein. On the other hand, collective diffusion is initiated by molecule-molecule collisions when the porous pathway is large enough to allow frequent inter-molecule interactions. Clearly, each diffusion has its own collision mechanism and diffusion coefficient. Gas advection, i.e. the viscous gas flow, is simply driven by pressure gradient; it is the linear gas seepage process governed by Eq. (1). The three processes in fact co-exist and the observed gas flow is their combined representation. Fig. 1 illustrates the three physical mechanisms of gas transport from a molecular point of view at different Knudsen numbers.

In order to obtain a unified governing equation for the whole process, it is advantageous to properly decouple the diffusions and advection. We properly define the diffusion processes and realize a simple additive characteristic of the total gas fluxes consisting of Knudsen diffusive flux, collective diffusive flux and advection flux as in Eq. (3)

$$v = v_K + v_C + v_A, \quad (3)$$

where v , v_K , v_C , v_A are the total gas velocity, Knudsen diffusive velocity, collective diffusive velocity and advection velocity, respectively. The advection process must be independent to the

diffusion to maintain a simple additive characteristic. From the molecule kinetic perspective and considering the total pressure gradient in this single gas system, in the molecule-wall collision region, the partial pressure gradient $(\nabla p)_{wall}$ because of molecules bounce back and forth between the walls is the driving force for the Knudsen diffusive flux. Similarly, for the region of molecule-molecule collisions, the collective diffusion is driven by another partial pressure gradient $(\nabla p)_{molecule}$. The summation of these two partial pressure gradient equals to the total pressure gradient as shown in Eq. (4).

$$\nabla p = (\nabla p)_{wall} + (\nabla p)_{molecule}, \quad (4)$$

Knudsen diffusion shares the similar governing law with Fickian diffusion as follows,

$$u_K = -D_K \nabla n_c, \quad (5)$$

where D_K is the Knudsen diffusion coefficient and ∇n_c is the molecular density gradient. Thanks to the ideal gas law of $pV = nRT$, Eq. (5) can be expressed in the form of

$$-(\nabla p)_{wall} = \frac{RT}{D_K} u_K, \quad (6)$$

where R is the universal gas constant and T is the temperature. In the same fashion, collective diffusion can be expressed in,

$$-(\nabla p)_{molecule} = \frac{RT}{D_C} u_C, \quad (7)$$

where D_C is the collective diffusion coefficient.

The total pressure gradient ∇p is the driving force for gas advection. Eq. (4) provides a relationship to connect among the Knudsen diffusion, collective diffusion and advection. However, it is rather difficult and obscure to distinguishingly calculate $(\nabla p)_{wall}$ and $(\nabla p)_{molecule}$ because of their molecular kinetic origins. Nevertheless, we can obscurely define a weighting factor α ($0 < \alpha < 1$) to characterize the contribution of molecule-wall or molecule-molecule collision to the total pressure gradient as follows,

$$(\nabla p)_{wall} = \alpha \nabla p, \quad (8)$$

$$(\nabla p)_{molecule} = (1 - \alpha) \nabla p \quad (9)$$

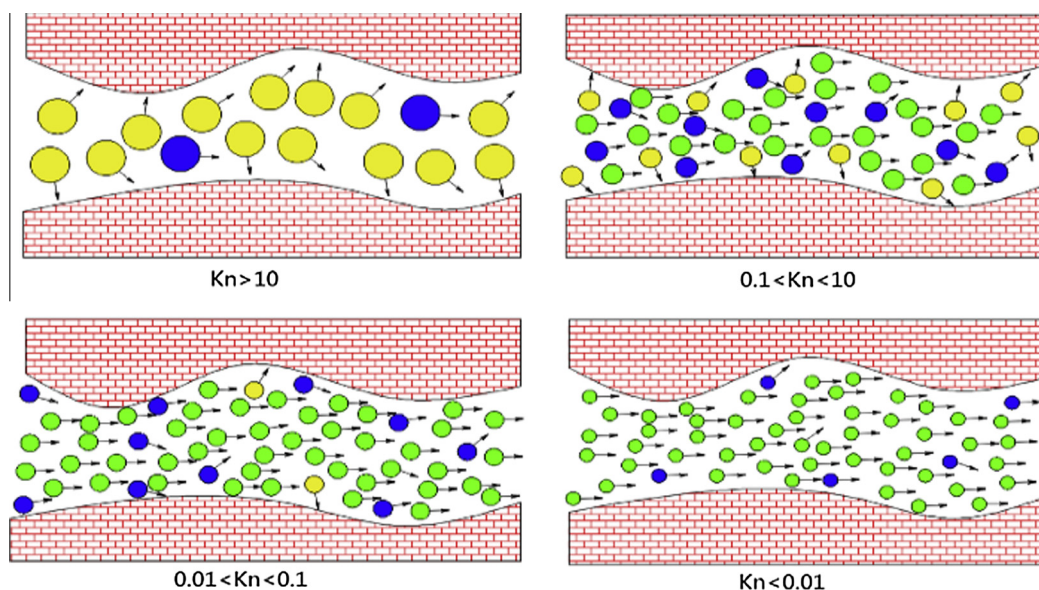


Fig. 1. Molecular illustration of gas transport process through a porous throat with different Knudsen numbers. The yellow cycles represent molecules engaged in molecule-wall collisions. The blue cycles represent molecules engaged in molecule-molecule collisions. The green cycles are advective molecular flux driven by pressure gradient. The arrows point to the momentum directions. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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