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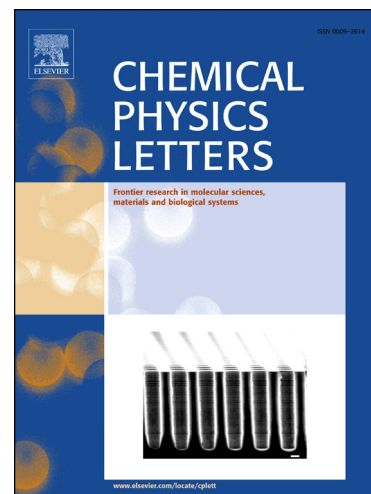
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Unifying Diffusion and Seepage for Nonlinear Gas Transport in Multiscale Porous Media

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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.

Keywords Nanopore; Knudsen Diffusion; Collective Diffusion; Advection; Darcy's Law;

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,

$$v = -\frac{k}{\mu} \nabla p,$$

(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 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. **Figure 1** illustrates the three physical mechanisms of gas transport from a molecular point of view at different Knudsen numbers.

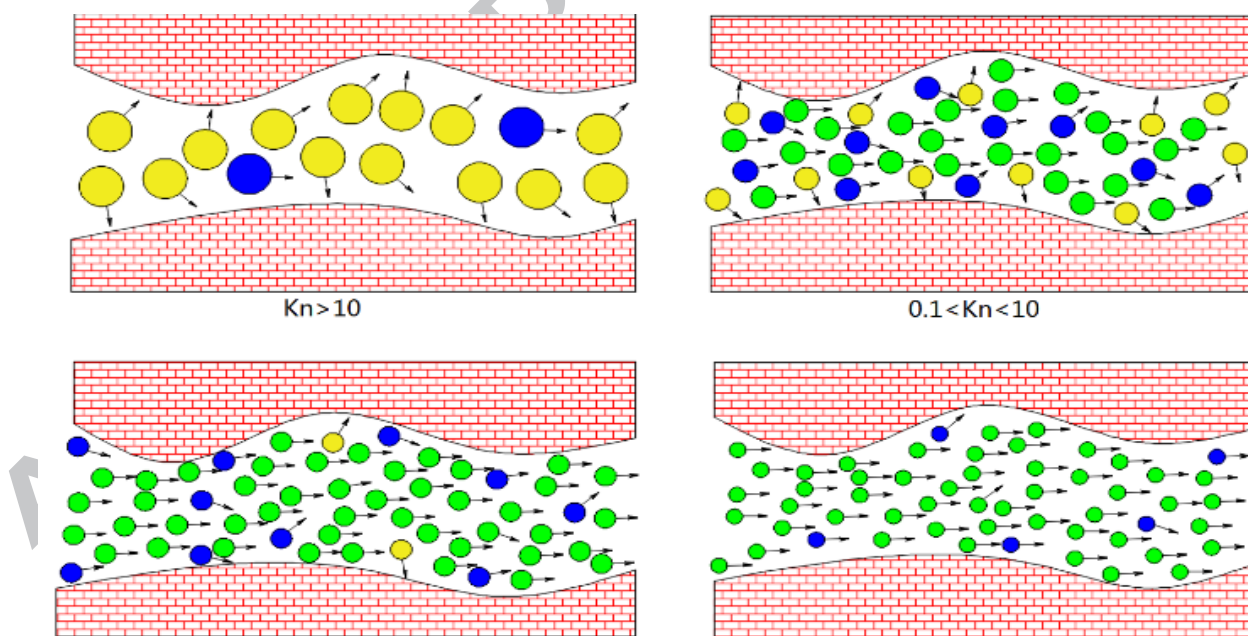


Figure 1. Molecular illustration of gas transport process through a porous throat with different Knudsen numbers. The yellow cycles represent molecules engaged in molecule-wall collisions. The blue cycles represent molecules engaged in molecule-molecule collisions. The green cycles are advective molecular flux driven by pressure gradient. The arrows point to the momentum directions.

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,

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

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

Based on Eq. (1), (3), (4), and (6)-(9) as well as unit system unification, we have

$$\begin{aligned} v &= v_K + v_C + v_A \\ &= - \left(\frac{VD_K}{nRT} \alpha + \frac{VD_C}{nRT} (1 - \alpha) + \frac{k}{\mu} \right) \nabla p \\ &= - \left(\frac{D_K}{P_{wall}} \alpha + \frac{D_C}{P_{molecule}} (1 - \alpha) + \frac{k}{\mu} \right) \nabla p \end{aligned} \quad (10)$$

Under the assumption that total system pressure has the same contribution split of weighting factor α , we simply have $p_{wall} = \alpha p$ and $p_{molecule} = (1 - \alpha) p$. As a result, Eq. (10) is further reduced to,

$$\begin{aligned}
v &= -\left(\frac{D_K}{p} + \frac{D_C}{p} + \frac{k}{\mu}\right)\nabla p \\
&= -\left(\frac{D}{p} + \frac{k}{\mu}\right)\nabla p
\end{aligned} \tag{11}$$

Apparently, from the coherent theoretical derivation above, we obtain the new general equation for gas transport through multiscale porous media. The new formulation now is supported with theoretical ground to model the four gas flow regimes with different K_n , i.e. multiscale porous media. Under this new framework, Darcy's law and Klinkenberg equation can be considered as special cases of Eq. (2). It is also clear that the previously cryptical D now actually is the summation of Knudsen and collective diffusion coefficients as $D = D_K + D_C$. We call this D as nominal diffusion coefficient. Knudsen diffusion characterizes the microscopic motion of the molecules and collective diffusion, on the other hand, quantifies the collective motion of particles which is more dominant for meso- and micro- scale porous media. In this sense, Eq. (2) is a general law of gas transport in multiscale porous media. We can use this formulation to describe single component gas flow through porous media spanning from nanoscale to macroscale, using different nominal diffusion coefficients.

In addition, Knudsen diffusion coefficient D_K is given by $D_K = \frac{4kc}{r_{pore}} \sqrt{\frac{\pi RT}{2M}}$ or $\frac{4cpk}{\mu} K_n$

¹¹ and collective diffusion coefficient D_C can be evaluated using $D_C = \frac{435.7T^{3/2}}{p(2V^{1/3})^2} \sqrt{\frac{1}{2M}}$, where c

is a dimensionless constant, r_{pore} is the pore radius, M is the molecular mass, and V is the molar volume. Clearly, D_K can dominate $D_K/(D_K + D_C)$ when K_n is very large (>10) or the pore size is very small (<100 nm). The weight of $D_C/(D_K + D_C)$ increases as K_n decreases or pore size

increases. In addition, when $\frac{D/p}{(D/p+k/\mu)}$ can be safely ignored, Eq. (1) is recovered from Eq. (2).

In coherent with the theoretical analysis, we conducted a series of physical and numerical experiments to investigate the gas transport characteristics through porous media, in order to provide credible support to the new formulation. As a critical component, we also provide an approach to characterize the gas seepage and diffusion fluxes simultaneously and quantitatively, i.e. to obtain D and k of the porous media at the same time.

A few naturally occurring porous media samples were cored from the Sichuan Basin shale gas play in Southern China. The samples are approximately 2.5×10^{-2} m and 5.0×10^{-2} m in diameter and length, respectively. The experimental apparatus is mainly composed of porous core, core holder, plungers, pump, pressure sensor, gas flow meter, gas supply device, and data acquisition system. A special design was in place to distribute the gas evenly and reduce the end effects. The volumetric velocities were measured at different pressure drops across the cores. Those data were collected to estimate their nominal diffusion coefficients and permeabilities. All the experiments were conducted at room ambient temperature. Three different cores were measured and sets of measurements were collected for each sample. It should be noted that it is difficult to achieve a completely stable volumetric velocity due to the extremely tight nanoporous cores. We ensured long enough gas flow stabilization time to reduce this source of experimental errors.

Eq. (12) provides the relationship to connect the volumetric velocity and pressure drop. It is actually the one-dimension analytical solution of Eq. (2),

$$Q_{sc} = \frac{T_{sc} Z_{sc}}{TZ P_{sc}} \frac{\pi d^2}{4l} \left[D(p_{in} - p_{out}) + \frac{k}{\mu} (p_{in}^2 - p_{out}^2) \right] \quad (12)$$

where Q_{sc} is the volumetric velocity, T_{sc} is the standard condition temperature, T is the ambient temperature, Z_{sc} is the standard condition gas compressibility factor, Z is gas compressibility factor, p_{sc} is the standard condition pressure, d is the sample diameter, l is the sample length, p_{in} is the inlet pressure and p_{out} is the outlet pressure.

3. Results and discussion

The collected sets of measurements (Q_{sc} and p_{in} pairs) are regressed using Eq. (12) by a least square method to obtain the three D and k pairs for the three samples, respectively. The constant parameters are: $T_{sc} = 293.15$ K, $T = 283.15$ K, $P_{sc} = 0.101$ MPa, $Z_{sc} = 1$, $Z = 0.89$, $\mu = 18.3 \times 10^{-6}$ Pa s. **Figure 2** provides the experimental measurements and regressed curves for the three core samples. Since it plots volume velocity vs. pressure square difference, when the difference is low the trend is not linear as shown in its upper right plot according to Eq. (12). In **Table 1** are the estimated values of D and k pairs.

Table 1. Estimated nominal diffusion coefficient D and permeability k of the three core samples.

Core code	Nominal diffusion coefficient D (m^2/s)	Permeability k (m^2)
LS1-3-7	4.01×10^{-9}	1.35×10^{-19}
LS1-11-5	7.19×10^{-9}	5.56×10^{-19}
LS1-5-1	7.64×10^{-9}	1.80×10^{-19}

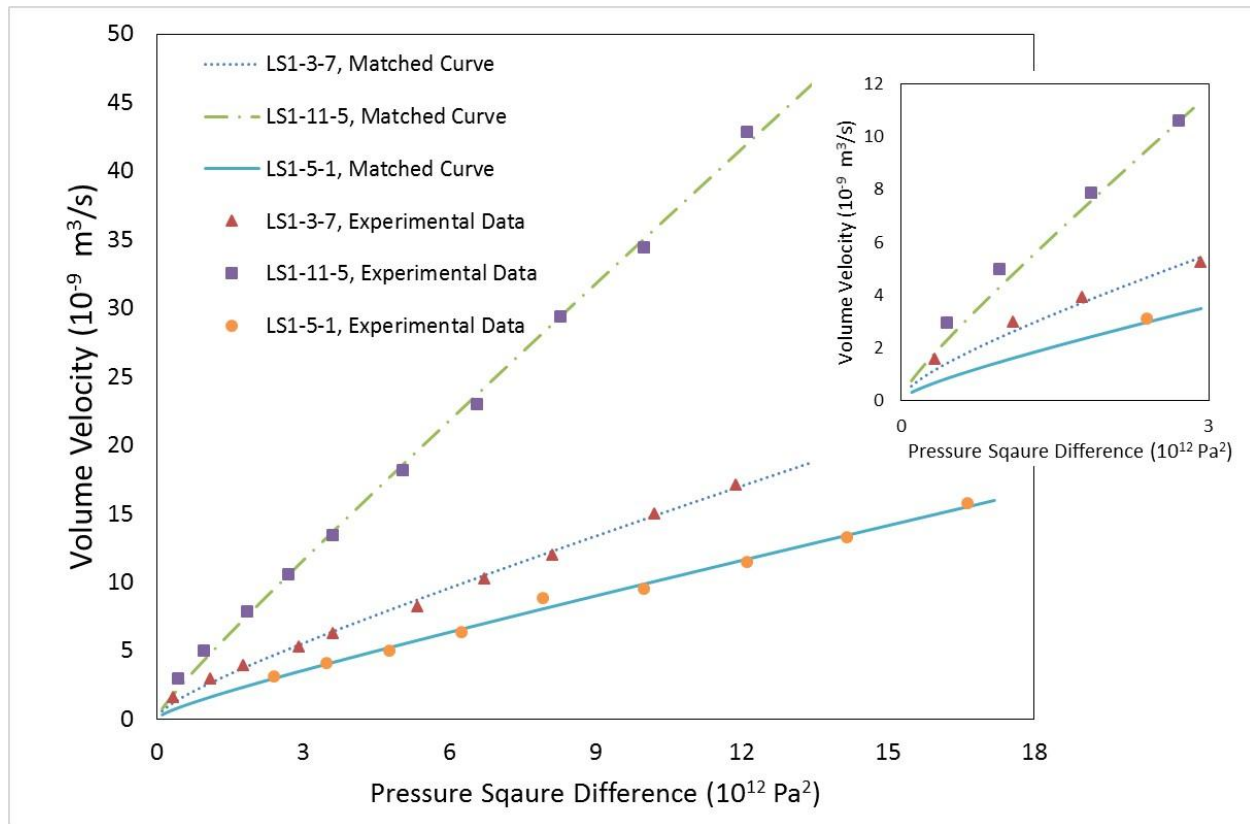


Figure 2. The experimental measurements and regression curves for the three core samples.

Our experimental results are clearly in good agreement with the new formulation and demonstrate its capability in proper description of the gas transport in nanoporous media. The experiment also showcases an approach to determine D and k simultaneously. However, coring samples from subsurface is not a feasible practice and is monetarily expensive. Plus, the experimental measurement is difficult to obtain credible readings due to the extremely long time needed to realize stabilized gas flow, especially for nanoporous samples. In this sense, numerical simulation can offer an efficient and effective alternate. We further employed Lattice Boltzmann Method (LBM) to model the gas transport process in porous media [22-27]. The LBM model is based on three assumptions considering (1) only two molecules collision, (2) independent velocities of two molecules as for the Chaos theory, and (3) no effect of external forces on the micro kinetic actions. The fluid is modeled using a single-particle distribution function. The

lattice Boltzmann equation is expressed by the simplified Boltzmann-BGK equation. The lattice arrangement is represented by the DnQm model. Since the molecular scale roughness may affect the collision frequency significantly [28], in order to consider the pore wall roughness, the boundaries are modeled as ideal diffuse reflection condition mixing the pure bounce-back and pure specular reflections with 50/50 splitting. The general transport equation (Eq. (2)) is used to match the simulation results. The average pore diameter (\bar{d}_{pore}) of the model is assumed to range from 10 nm to 1000 nm. **Figure 3** illustrates the gas velocity section planes sliced at 4 locations along the gas flooding direction. The hotter color stands for locations of higher velocities, while the coldest color represents for the stagnant solid portion of the porous media.

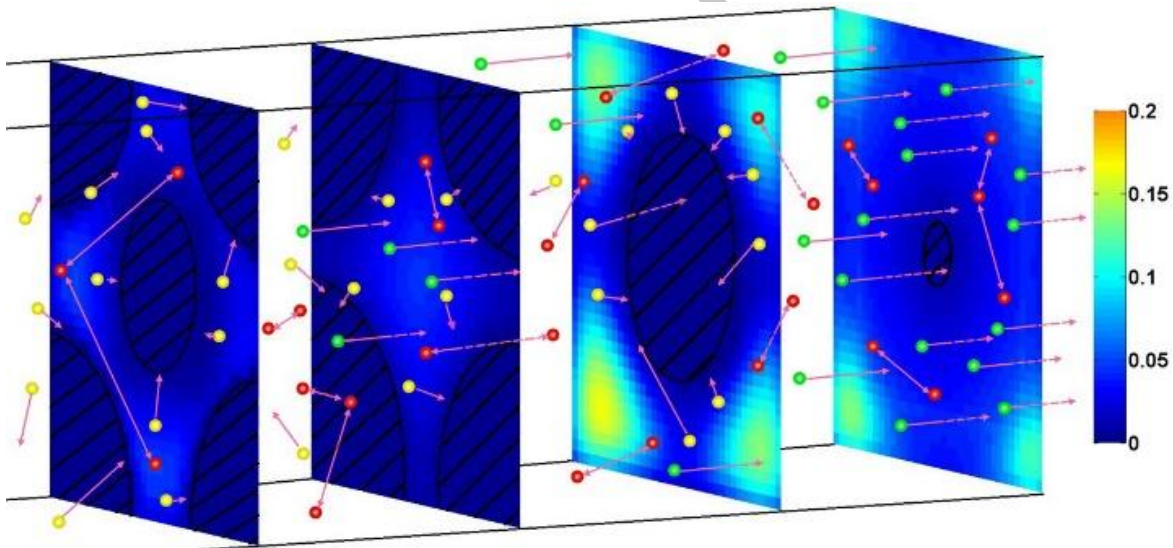


Figure 3. Gas velocity section planes sliced at 4 locations along the gas flooding direction. The solid black circles with diagonal lines outline the solid pore structure. Yellow balls represent molecules engaged in molecule-wall collisions. Red balls represent molecules engaged in molecule-molecule collisions. Green balls are advective molecular flux driven by pressure gradient. The arrows point to the momentum directions.

In **Figure 4 (a)** we plot the average gas velocities from LBM simulation versus pressure differences for \bar{d}_{pore} . As expected, the average velocity increases with the pore size. In **Figure 4 (b)**, together with LBM results of 10 nm porous model, we plot the gas velocities versus pressure

differences curves using the three different kinetic equations, namely, general transport equation, Klinkenberg equation and Darcy's law. Obviously, the general equation yields the best fit while Klinkenberg equation and Darcy's law fail to match the simulation results. Similarly, we calculate the nominal diffusion coefficient D , permeability k of the four different LBM models and list the results in **Table 2**. In a nutshell, all the parameters in the proposed general equation have precise physical meaning. The nominal diffusion coefficient can be obtained by experimental data analysis or LBM simulation as the same as permeability in classic Darcy's law. As shown clearly, Darcy's law now is a special case of this general equation.

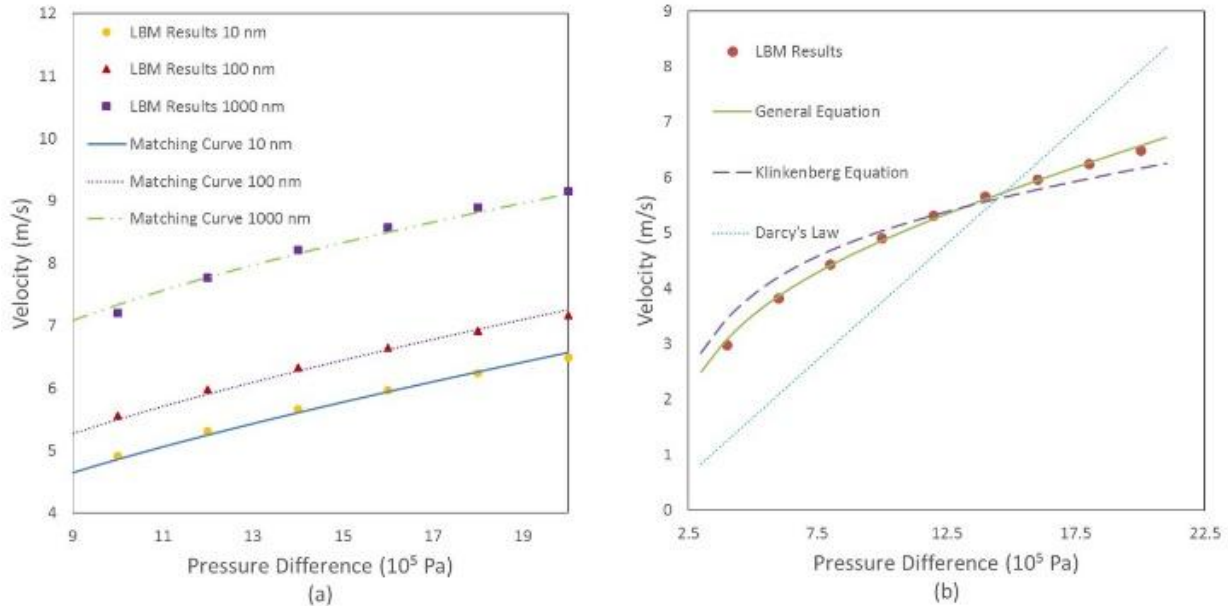


Figure 4. (a) Average gas velocities versus pressure differences of four porous model with different \bar{d}_{pore} . (b) Comparison of the LBM results with Eq. (2), Klinkenberg equation and Darcy's law for $\bar{d}_{pore} = 10$ nm.

Table 2. Calculated nominal diffusion coefficient D and permeability k of the four LBM models.

\bar{d}_{pore} (nm)	K_n	Nominal diffusion coefficient D (m^2/s)	Permeability k (m^2)
10	3.2	2.41×10^{-7}	1.55×10^{-18}
100	0.32	2.84×10^{-6}	1.53×10^{-17}
1000	0.032	4.16×10^{-5}	1.29×10^{-16}

4. Conclusion

Summarizing, we have provided a coherent theoretical derivation of the new general gas transport equation from the molecular kinetics prospective. It is now clear that the new formulation is capable of modeling single component gas transport through multiscale porous media from macroscale to nanoscale. The wall-molecule and molecule-molecule collisions drive the Knudsen and collective diffusive fluxes, and constitute the system pressure across the porous media, respectively. The resulting pressure gradient, if nonzero, is the driving force for gas seepage through the underlying porous media. The so-called nominal diffusion coefficient in the new formulation is the summation of Knudsen and collective diffusion coefficients. We have also reported physical and numerical experiments to support the model and approaches to obtain the nominal diffusion coefficient and permeability of the porous media simultaneously.

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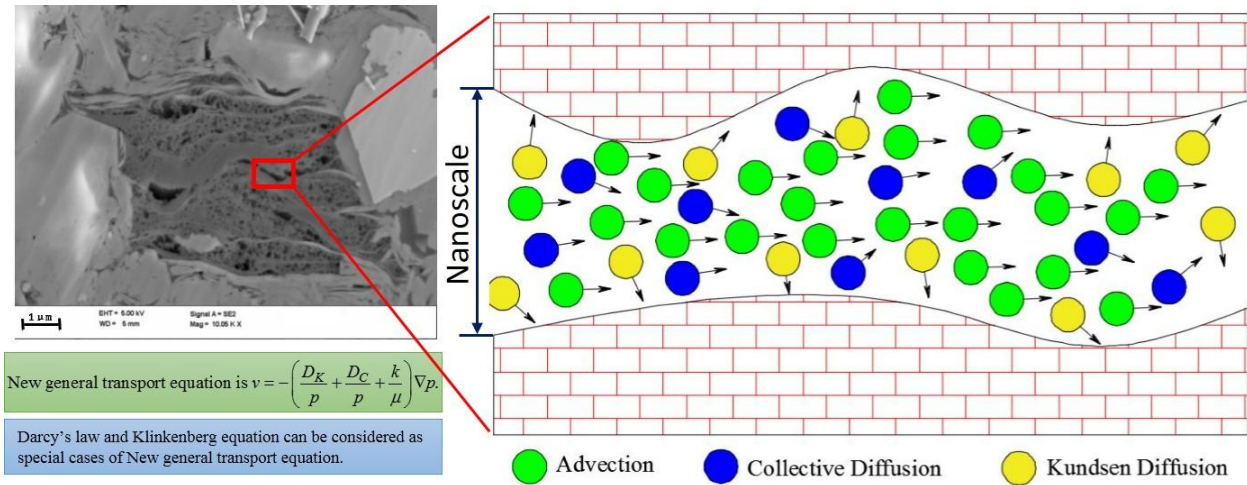
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Graphical abstract



Highlights

1. A new terminology, nominal diffusion coefficient is defined to summarize Knudsen and collective diffusion coefficients.
2. 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.
3. A new general transport equation is proposed to unify the diffusion and seepage process for nonlinear gas transport in multiscale porous media.
4. Physical and numerical experiments show the support of the new formulation and provide approaches to obtain the diffusion coefficient and permeability simultaneously.