CHAPTER 1

Mesoscopic Simulation of Rarefied Gas Flow in Porous Media

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Abstract: The accurate description of flow in nano-scale pores or channels is very important for the reliable design of materials and processes in the areas of MEMS, mesoporous media, and vacuum technologies. Use of classical flow equations fails in this regime since the continuum assumption is not valid. This is due to the fact that the mean free path is comparable to the characteristic dimensions of the system, and rarefaction effects dominate the process. Such a difficulty arises notably in the intermediate Knudsen number regime (Kn=0.1 to 10), commonly referred to as the "transition" flow regime. To remedy this, slip flow conditions have been adopted in the literature, following the simple first-order approach of the velocity near the walls given by Maxwell, and extended to higher-order treatments. Alternatively, direct deterministic or stochastic atomistic and mesoscopic techniques have been employed for the flow description, which solve the Boltzmann or the Burnett equations and use kinetic theory approaches pertinent to this flow regime. A description of recent advances in simulation techniques, namely, the "continuum" slip approaches, and some direct mesoscopic techniques are presented in this chapter. Illustrative simulation results of permeability and viscosity coefficients in mesoporous media using the DSMC and LB methods are also given, followed by comparisons with classical continuum formulations.

Keywords: Rarefied flow; porous media; transition regime; Direct Simulation Monte Carlo; Lattice-Boltzmann; reconstruction; fractional Brownian motion; slip flow; nanoscale pores; Knudsen number; mesoscopic methods.

INTRODUCTION

Recent advances in microtechnology and, in particular, in microelectromechanical systems (MEMS) and nano porous media have necessitated the elucidation of flow and transport processes in small dimensions. This is also the case with several other industrial applications, which rely on low-pressure conditions, or

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the molecular time and length scales are not sufficiently small compared to the characteristic macroscopic flow scales.

Therefore, it is essential to define additional parameters in order to classify the flow regimes, as well as the fluid state. Gases are considered dilute when the mean molecular spacing, $\tilde{\delta}$, is at least 7 times greater than the mean molecular diameter, $\tilde{\sigma}_0$, [8] that is, $\frac{\tilde{\delta}}{\tilde{\sigma}_0} > \sim 7$ [6]. On the contrary, if the aforementioned ratio is smaller than ~7, the gas is considered as dense. Alternatively, the ratio of a macroscopic characteristic flow dimension scale, $\tilde{L}$, to the gas molecule distance, $\tilde{\delta}$, that is, $\frac{\tilde{L}}{\tilde{\delta}}$, can be used to bound the continuum approach. More specifically, in order for the molecular chaos to be ensured and the molecular fluctuations to be statistically insignificant, this ratio has to be greater than 100, which means that at least 10^6 particles reside within the elementary fluid volume. The various flow regimes and the corresponding governing equations are summarized in Fig. 1 (see also [1]).

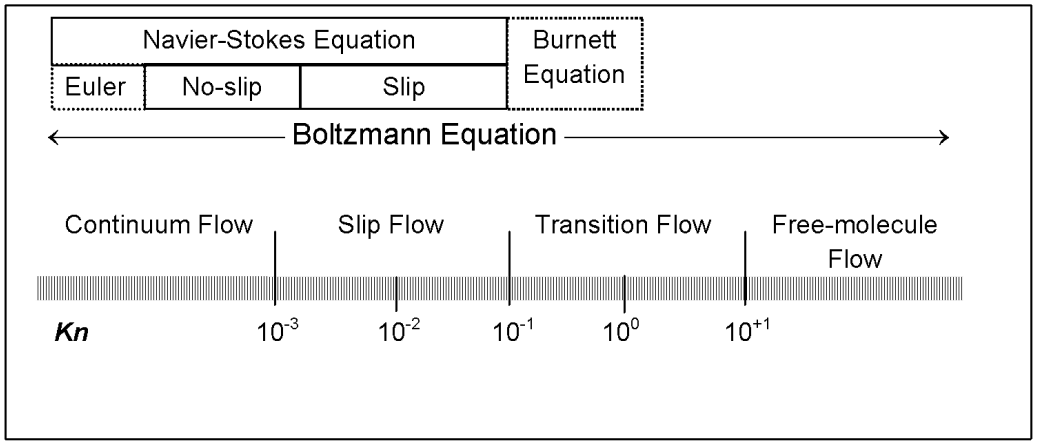

Figure 1: Flow regimes and governing equations.

In the context of this work a description of recent advances in simulation techniques, namely, the "continuum" slip approaches [1, 9], and direct mesoscopic techniques, such as the Direct Simulation Monte Carlo (DSMC) method [8, 10-12], the Information Preservation (IP) method [13-16], and the Lattice Bolzmann (LB) method [17-18], are presented. Illustrative simulation results of permeability and viscosity coefficients in mesoporous media using the

$$\lambda = \frac{3\mu}{n}\sqrt{\frac{\pi}{8mkT}} \qquad (1)$$

Following eq. (1), the Knudsen number can be expressed as

$$Kn = \frac{\lambda}{L} = \frac{3\mu}{nL}\sqrt{\frac{\pi}{8mkT}} \qquad (2)$$

where L is a characteristic macroscopic flow dimension.

Several methods to simulate rarefied flows have been proposed, which fall into two main categories, namely macroscopic and microscopic approaches. The former consist of the direct solution of the Navier-Stokes equation or semi-empirical equations with appropriate boundary condition to induce the slip effect, whereas the latter consist of either direct or stochastic particle monitoring methods using kinetic theory descriptions, such as Molecular Dynamics (MD) and Monte Carlo (MC) methods. Even though the microscopic approaches are more accurate inherently, most processes involving porous media and micro-sized devices (*i.e.* MEMS) would require enormous CPU resources to be simulated effectively. Therefore, in order for the direct methods to be applicable in systems of micro-size, mesoscopic approaches have been introduced in this area, such as the Direct Monte Carlo Simulation (DSMC), the Information Preservation (IP), and, more recently, the Lattice Boltzmann (LB) methods. In the context of this

chapter the discussion will elucidate some aspects of the macroscopic approaches and will be focused on the DSMC and the LB methods.

MACROSCOPIC APPROACH

Strictly, the continuum condition (Fig. **1**) prohibits the use of the Navier-Stokes equation for non-zero Knudsen numbers. Nevertheless, by introducing slip boundary conditions, Arkilic and coworkers [9] proposed a two-dimensional scheme for rarefied flows involving the Navier-Stokes equation and a first order boundary condition, proposed by Maxwell [2] (Fig. **2**), which reads

$$\breve{u}_s - \tilde{u}_w = \frac{2-\sigma_v}{\sigma_v}\frac{\tilde{\lambda}}{\tilde{\mu}}\tau_s\Big|_{wall} + \frac{3}{4}\frac{\gamma-1}{\gamma}\frac{\mathrm{Pr}}{\tilde{P}}\left(-\tilde{q}_s\right)\Big|_{wall} \qquad (0)$$

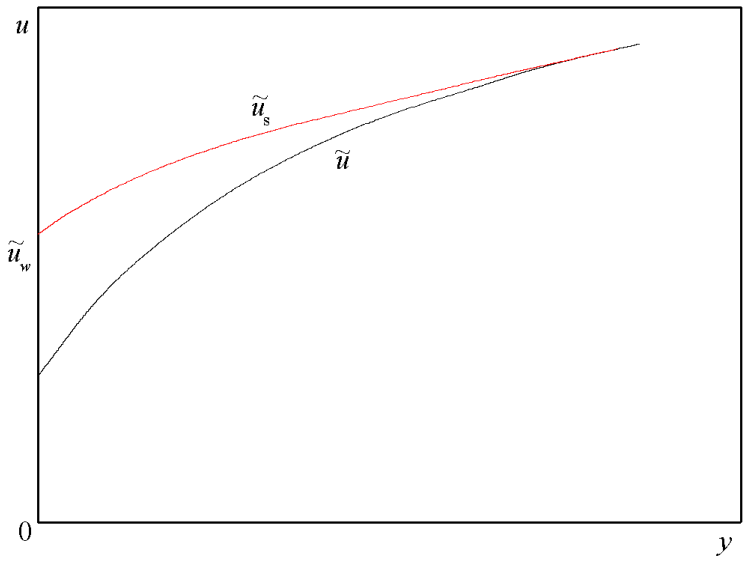

Figure 2: Slip velocity at the wall.

$$\breve{u}_s = \frac{2-\sigma_v}{\sigma_v}\left[Kn\frac{\tilde{\partial}u}{\partial\hat{n}}\bigg|_{wall} + \frac{Kn^2}{2}\frac{\partial^{\tilde{2}}u}{\partial\hat{n}^2}\bigg|_{wall} + \ldots \right]. \qquad ()$$

The authors [12] further simplified the boundary condition in order to avoid the calculation of the 2nd order velocity derivatives that may introduce computational difficulties, to the following slip boundary condition

$$\breve{u}_s = \frac{2-\sigma_v}{\sigma_v}\frac{Kn}{1-bKn}\frac{\tilde{\partial}u}{\partial\hat{n}}\bigg|_{wall}, \qquad ()$$

where b is a parameter that "incorporates" the 2nd order derivative term and takes the value b =-1 for fully developed flow in a channel.

The proposed unified model predicts mass flow rates, pressure distributions and the well-known Knudsen minimum effect, for flows in channels and tubes with reasonable accuracy compared with DSMC calculations, linearized Boltzmann equation [21] solutions and experimental data [1].

$$\tilde{P} = \tilde{\rho}\tilde{k}T / \tilde{m} \qquad \textbf{(19)}$$

Temperature at location $\breve{y}$:

$$\breve{T}(\breve{y}) = \frac{2\tilde{m}}{3\tilde{k}\tilde{\rho}}\left(\frac{1}{2}\left\langle \tilde{m}\tilde{v}^2 \right\rangle_y - \frac{1}{2}\tilde{\rho}\tilde{u}^2 \right) \qquad \textbf{(20)}$$

where $\tilde{m}$ is the molecular mass, and $\breve{k}$ is the Boltzmann constant.

In the DSMC treatment, boundary interactions are treated as collisions of either purely specular reflection type, purely diffuse reflection type, or a combination of specular and diffuse reflections, with specific description of the energy and

momentum transfer using accommodation coefficients of the reflection behavior along the tangential and the normal directions to the walls [32].

Recently, Michalis and co-workers [12] proposed an effective viscosity in order to account the rarefaction effects present at high Knudsen numbers. Using the Direct Simulation Monte Carlo method in a wide range of Knudsen number flows along a channel, they concluded that a Bosanquet-type of approximation describes very satisfactorily the Knudsen number dependence of the viscosity over the entire transition regime, that is, from the slip-flow to the free-molecular flow limit assuming that the system is not far from thermodynamic equilibrium conditions. The effective viscosity is obtained by assuming the following Bosanquet-type expression

$$\frac{1}{\tilde{\mu}_e} = \frac{1}{\tilde{\mu}_{Bulk}} + \frac{1}{\tilde{\mu}_{Kn}} \tag{21}$$

where μ_{Bulk} is the bulk viscosity ($Kn \to 0$) and μ_{Kn} corresponds to the free molecular flow ($Kn \to \infty$). A simple expression that relates the effective viscosity to the Kn is given by the expression [12]

$$\tilde{\mu}_e = \tilde{\mu}_{Bulk} \frac{1}{1 + aKn} \tag{22}$$

which was also suggested by Beskok and Karniadakis [1]. The parameter a is a numerical factor, which is not constant but depends on the Kn value. However, this dependence is rather weak over the majority of the transition regime, suggesting an "effective" value close to 2, as shown in [12].

Mesoscopic Methods: The LB Approach

The lattice Boltzmann method (LBM) statistically monitors the evolution of discrete particle populations on a given lattice. The evolution of the system is

described by the discrete Boltzmann equation, whose simplest form looks as follows [33, 34],

$$f_i(\mathbf{x}+\mathbf{e}_i, t+1) = f_i(\mathbf{x}, t) + \Omega_i \quad ; \quad i = 0,...,8 \tag{23}$$

where f_i is the single particle distribution function, Ω_i is the collision operator, $\boldsymbol{x}$ and t the spatial and time variables, respectively, and $\{\boldsymbol{e_i}\}$ is the discrete velocity set on the lattice.

For a two dimensional 9 speed LB model the nine unit-velocity vectors (D_2Q_9) are depicted in Fig. **3**, and are defined below [33, 34]:

$$\mathbf{e}_0 = (0,0) \tag{24}$$

$$\mathbf{e}_i = \left(\cos\frac{i-1}{2}\pi, \sin\frac{i-1}{2}\pi \right) \quad ; \quad i = 1,...,4 \tag{25}$$

$$\mathbf{e}_i = \sqrt{2}\left(\cos\frac{i-5}{2}\pi + \frac{\pi}{4}, \sin\frac{i-5}{2}\pi + \frac{\pi}{4} \right) \quad ; \quad i = 5,...,8 \tag{26}$$

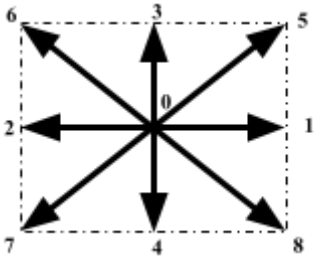

Figure 3: Velocity vectors on a lattice cell.

Introducing the single relaxation parameter τ [35] the collision operator can be approximated by the following expression [33, 34]:

$$\Omega_i = -\frac{f_i - f_i^{eq}}{\tau} \quad ; \quad i = 0,...,8 \tag{27}$$

where f_i^{eq} is the equilibrium distribution function (EDF). The EDF is expanded around the local velocity [35-38] as follows:

$$f_i^{eq} = w_i \rho \left(1 + \frac{1}{c_s^2} (\mathbf{e}_i \cdot \ ) + \frac{1}{2c_s^4} (\mathbf{e}_i \cdot \ )^2 - \frac{1}{2c_s^2} \upsilon^2 \right);$$

$$w_0 = \frac{4}{9} \quad ; \quad w_i = \frac{1}{9}, i = 1,..,4 \quad ; \quad w_i = \frac{1}{36}, \qquad i = 5,..,8 \tag{28}$$

In the above equilibrium distribution function the speed of sound is

$$c_s = \frac{1}{\sqrt{3}} \tag{29}$$

The local density and velocity of the LB particles are given by the following summations:

$$\rho(\mathbf{x},t) = \sum_i f_i(\mathbf{x},t) = \sum_i f_i^{eq}(\mathbf{x},t) \tag{30}$$

$$\rho \mathbf{\upsilon}(\mathbf{x},t) = \sum_i f_i(\mathbf{x},t)\mathbf{e}_i = \sum_i f_i^{eq}(x,t)\mathbf{e}_i \tag{31}$$

The Navier-Stokes equation can be recovered from the LB equations using the Chapman-Enskog expansion [30, 39, 40]. The mass and momentum balance equations derived from equations (23)-(31) are:

$$\frac{\partial \rho}{\partial t} + \nabla \cdot \left(\rho \mathbf{\upsilon} \right) = 0 \tag{32}$$

$$\frac{\partial(\rho\mathbf{v})}{\partial t}+\nabla\cdot(\rho\mathbf{v}\mathbf{v})=-\nabla P+\mu\nabla^2\mathbf{v}+O(\upsilon^3) \tag{33}$$

In the LB method the dimensionless mean free path and relaxation parameter are related to their dimensional quantities through

$$\tilde{\lambda}=\lambda\,\tilde{\delta}x\ ,\ \tilde{t}_{rel}=\tau\,\tilde{\delta}t \tag{35}$$

where $\tilde{\delta}x$ is a reference (if more than one) lattice spacing and $\tilde{\delta}t$ is the time that is required for a fluid particle to travel this distance.

During a time interval equal to the relaxation time, $\tilde{t}_{rel}$, the particles with mean speed $\tilde{\bar{c}}$ travel a distance equal to the mean free path $\tilde{\lambda}$. Therefore, a way to express the mean free path is [48]:

$$\tilde{\lambda}=\tilde{\bar{c}}\tilde{t}_{rel}=\bar{c}\,\tau\,\tilde{\delta}x \tag{36}$$

The speed of sound ($\tilde{c}_s$) through the fluid is given from

$$\tilde{c}_s^2=\left(\frac{\tilde{\partial}P}{\tilde{\partial}\rho}\right)_S \tag{37}$$

that is,

$$\tilde{c}_s=\sqrt{\frac{\tilde{k}\tilde{T}}{\tilde{m}}} \tag{38}$$

Combination of equations (37) and (38) gives for the mean thermal speed:

$$\tilde{c} = \sqrt{\frac{8}{\pi}} c_s \tag{39}$$

Using the node-to-node particle speed as reference speed, we get

$$\tilde{c}_s = c_s \frac{\tilde{\delta} x}{\tilde{\delta} t} \tag{40}$$

The dimensionless speed of sound is a known quantity that depends on the prescribed spatial discretization and the specific lattice that is used. For instance,

boundary condition can be altered according to the value of ξ ($\xi=1$ corresponds to no-slip boundary condition and $\xi=0$ to free slip and, eventually, to plug flow). In what follows the 1st order value presented in the previous paragraph (ξ=0.4658) is used.

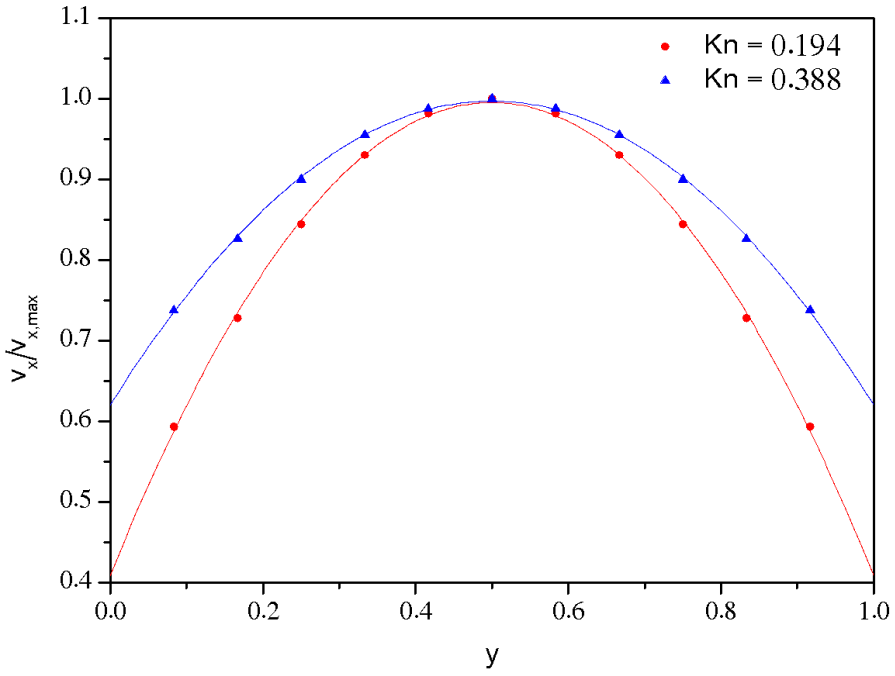

Figure 4: Gas flow velocity profile at a cross section inside a tube LB method for *Kn*=0.194 (bottom line), and *Kn*=0.388 (top line).

In Fig. **4** the cross-section velocity profiles and the slip length are presented for two Knudesn numbers, namely, *Kn*=0.194 and *Kn*=0.388. The slip length appears to increase by 50% upon doubling the *Kn* value. The cross-section velocity profiles in a channel are depicted in Fig. **5** for *Kn*=0.388, using the DSMC method, the LB method with *Kn*-dependent gas viscosity, and a standard numerical solution to the NS equation assuming no-slip at the wall. Fig. **6** presents the dimensionless relative pressure along the tube, *i.e.*, its deviation from linearity, defined as:

$$p^* = \frac{p - p_{linear}}{p_{outlet}} = \frac{p - \left[p_{inlet} - \left(p_{inlet} - p_{outlet} \right)\left(x/L \right) \right]}{p_{outlet}} \qquad \textbf{(46)}$$

Even though the rarefied LB model results compare satisfactorily with the DSMC ones for low Knudsen numbers (*Kn*<0.2), the LBM seems to fail to reproduce the DSMC data at intermediate or higher Knudsen numbers. In order to extend the validity of the rarefied LBM to intermediate and higher *Kn* a modification that relates the fluid viscosity to the Knudsen number has to be employed. To this end the effective viscosity that was recently proposed [12], and presented previously can be adopted in the LBM. Therefore, a modified *Kn* can be obtained that will render the LB more accurate as it takes into account the rarefaction effect over the entire flow regime. The correct prescription of the viscosity in the LB model is of tremendous importance in cases of flow in porous media, where more than one flow regime may be present across different subregions of the medium.

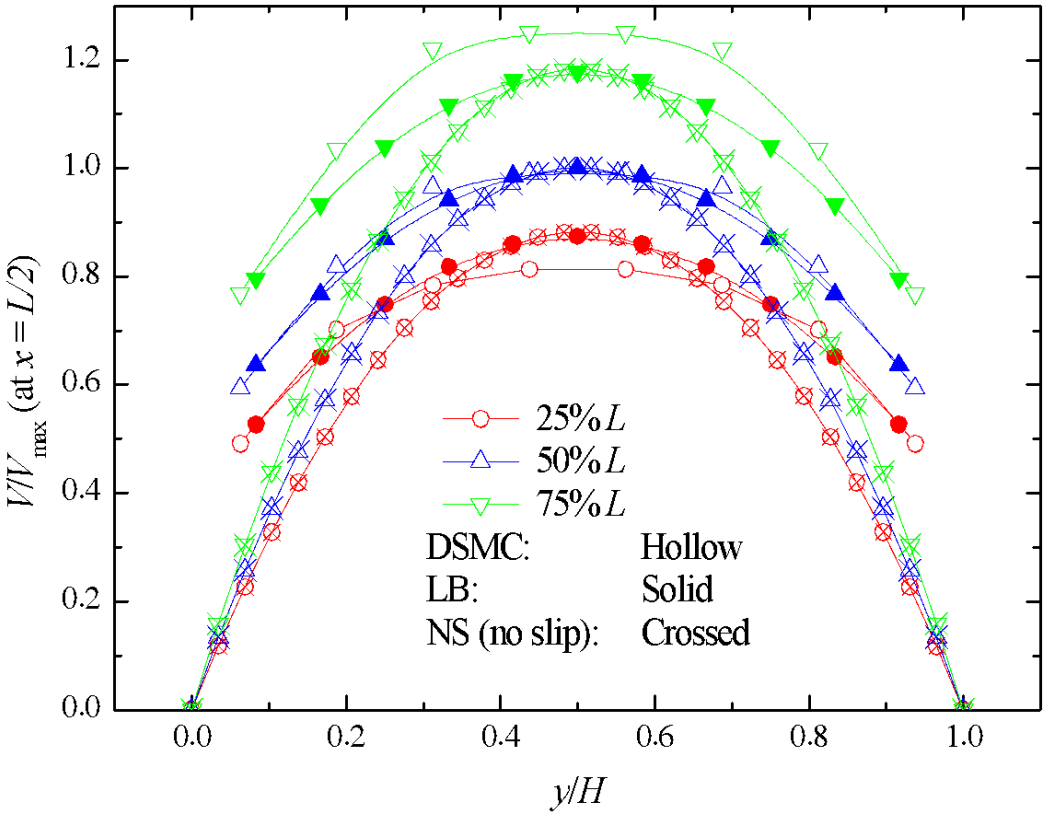

Figure 5: Gas (N_2) flow velocity profiles at characteristic cross-sections inside a tube (at $L/4$, $L/2$, $3L/4$ from the entrance) obtained using the mesoscopic DSMC and LB methods, and the "macroscopic" NS (with no-slip BC) method, for Kn_{out} = 0.388. The magnitude of the velocity at the middle of the tube ($L/2$, $H/2$) was used for reduction.

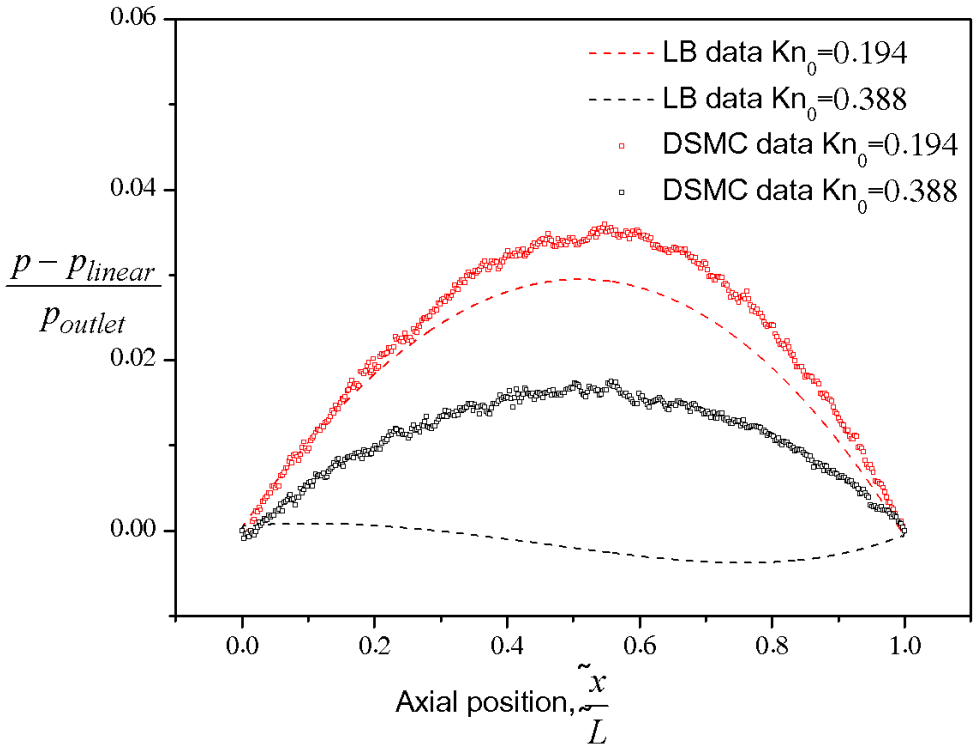

Figure 6: Relative pressure deviation from linearity.

Flow in a Porous Medium

The porous structures examined here were reconstructed with the Fractional Brownian Motion method [56], in which the Hurst exponent allows the adjustment of the degree of the local correlation of the structure. Figs **7-8** present indicative calculation results in a porous medium with porosity ε=0.7 and Hurst exponent H=4, obtained using the DSMC and LBM mesoscopic techniques, and the conventional NS solution in the same locations. More specifically, indicative streamlines in a typical part of the porous sample are displayed at several Knudsen numbers in Fig. **7**, while comparison of the mean field values is given in Fig. **8**, where the dependence of the superficial velocity on the rarefaction effects is apparent.

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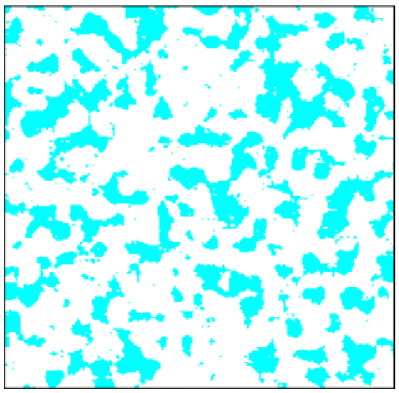
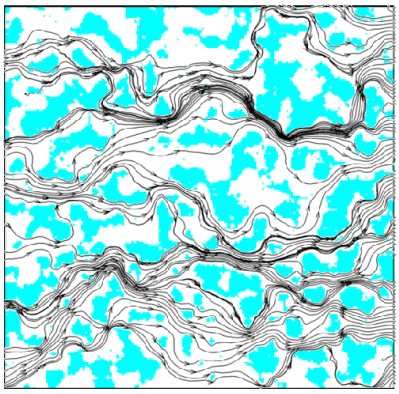
(a)



(b) Kn=5



(c) Kn=10

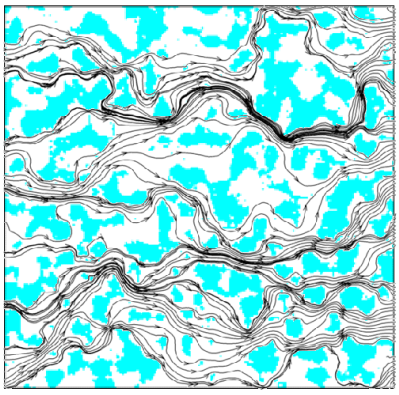


Figure 7: Gas (N_2) flow patterns in a tortuous reconstructed (FBM) porous medium (ε=0.7, H=0.4) for (a) Kn_{out}=0.5, (b) Kn_{out}=5, and (c) Kn_{out}=10, using DSCM methods. Streamlines end at equidistant positions along the exit face.

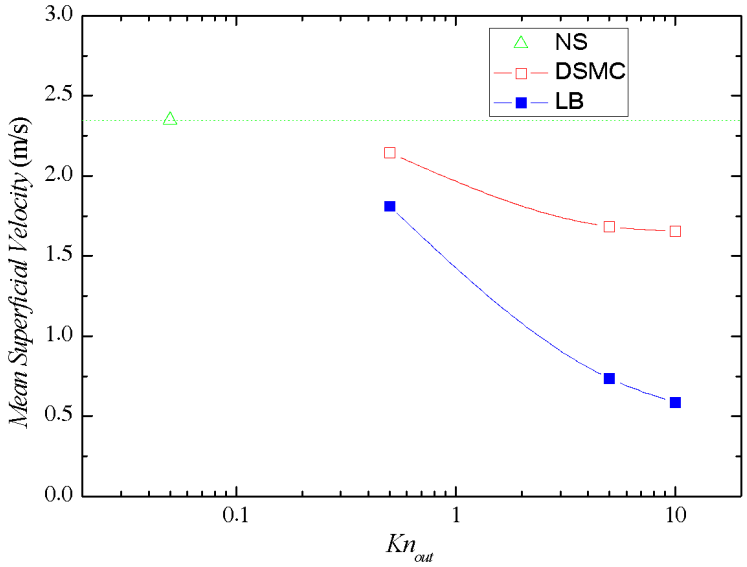


Figure 8: Mean superficial velocity at elevated Kn numbers at the outlet of a porous medium (reconstructed with FBM, ε=0.7, H=0.4).

CONCLUDING REMARKS

The need to describe rarefied flows in a variety of applications that involve micro, and nanoscale pores or channels has stimulated the development of a multitude of theoretical and numerical methodologies. A comparison of the predictions of the mesoscopic approaches with those of the Navier-Stokes formulation complemented by a non-slip flow condition revealed that the latter can be safely employed for very small Knudsen number values only. The DSMC method was used to determine the flow field in different types of porous media and processes.

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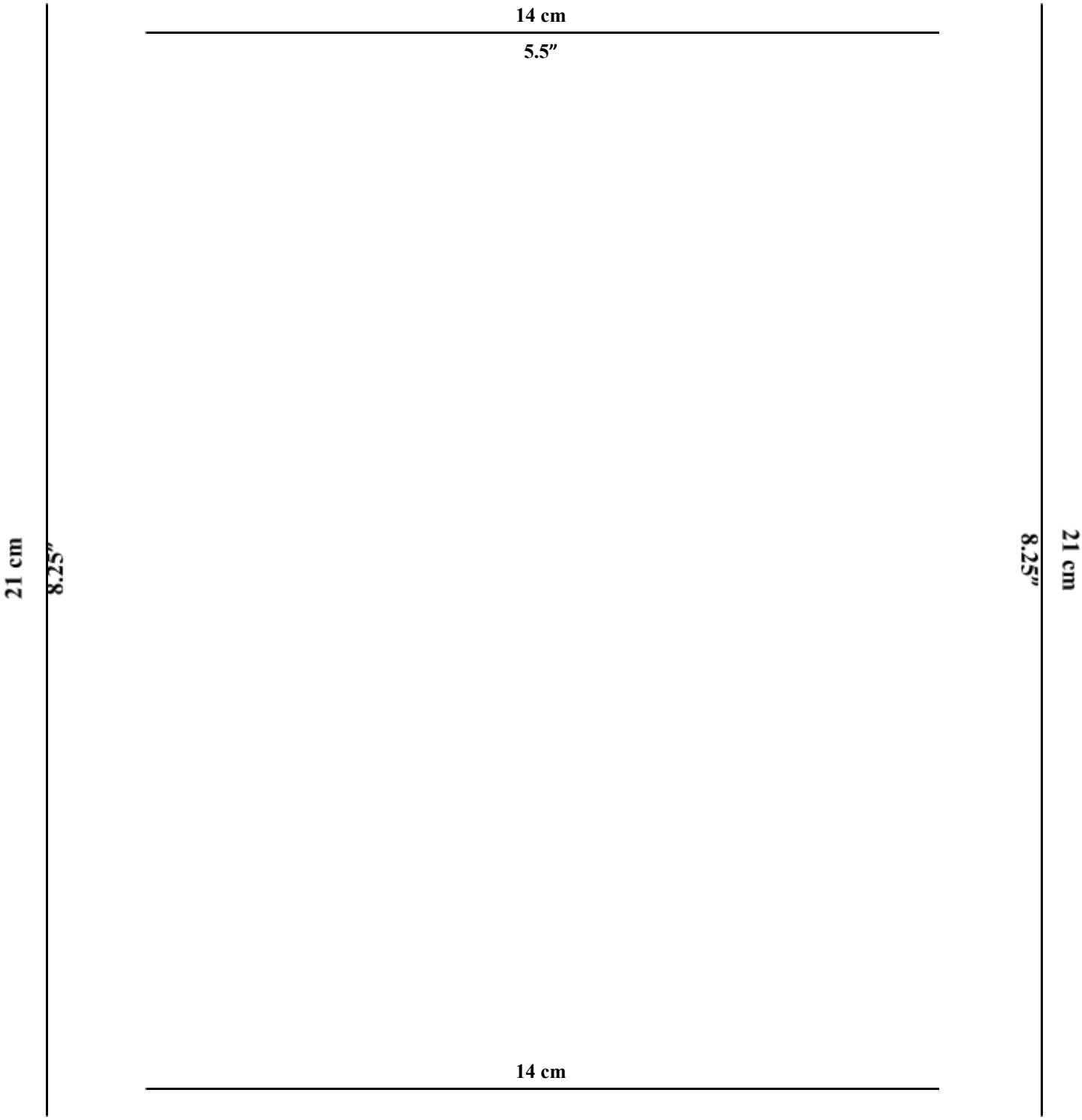
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