A joint lattice Boltzmann and molecular dynamics investigation for thermohydraulic simulation of nano flows through porous media

A.H. Meghdadi Isfahani\textsuperscript{a}, Iman Tasdighi\textsuperscript{b}, Arash Karimipour\textsuperscript{a,\ast}, E. Shirani\textsuperscript{b}, Masoud Afrand\textsuperscript{a}

\textsuperscript{a} Department of Mechanical Engineering, Najafabad branch, Islamic Azad University, Najafabad, Iran
\textsuperscript{b} Foolad Institute of Technology, FooladShahr, Isfahan, 8491663763, Iran

\textbf{ARTICLE INFO}

\textbf{Article history:}
Received 7 February 2015
Received in revised form 6 July 2015
Accepted 6 August 2015
Available online 13 August 2015

\textbf{Keywords:}
Molecular dynamics
Nano channel
Nano porous
Poiseuille flow

\textbf{ABSTRACT}

The performance of standard Lattice Boltzmann, LB, is confined to the micro scale flows with a Knudsen number, $Kn$, less than 0.1. In the previous works we proposed a modified LB which is able to extend the ability of LBM to simulate wide range of Knudsen flow regimes. To study the ability of this model for porous media, in the present work we investigate nanoscale gaseous flows through porous media confined between parallel plates by means of LB, and molecular dynamics, MD, simulations. The comparison between modified LB and MD shows a very good agreement at sufficiently low solid fractions. In addition, flow characteristics through various porous structures are studied via the MD simulation and the effect of various parameters such as driving force, porosity and Knudsen number on the velocity and temperature distributions is investigated. Our results demonstrate the good ability of MDS for investigating different flow phenomena such as permittivity and Darcian flow in porous media.

© 2015 Elsevier Masson SAS. All rights reserved.

1. Introduction

Flow and heat transfer through the porous medium have become one of the most popular research subjects in fluid flow problems due to their wide applications [1]. With recent advances in micro and nano technology mechanical devices such as micro/nano channels, nozzles and pumps can be fabricated. In micro/nano fluids, porous media can be used for micro filtration, fractionation, catalysis and microbiology related applications [2].

Molecular based methods such as Molecular Dynamics Simulation, MDS, is one of the most certain methods, which can be used for simulation of flow at nanoscales. MDS is a computational method based on Newton’s second law, i.e. $F = ma$, which calculates the time dependent behavior of a molecular system. In this method, for tracking system trajectories, the equations of motion for molecules are integrated. MD produces detailed information on position and velocity of molecules at nanoscale. Using statistical mechanics, one can determine the macroscopic properties of system such as pressure, energy, heat capacity, surface tension, temperature, and density. However, the computational cost of these molecular based methods is prohibitively high such that they cannot be used for practical fluid flow simulations in micro and nano scales except for free molecular regimes. A midway approach for the flow simulation in small scales is using the Boltzmann equation [3] or its approximate forms such as BGK kinetic model [4,5] and Lattice Boltzmann Model, LBM. Because the LBM is a mesoscopic method which can be considered as a particle based method and at the same time is independent of actual number of molecules, it require less computational resources for low Kn flows. Recently there have been attempts to use the LBM for gaseous flows in slip flow regime (micro scale) [6–18] but only a few papers can be mentioned for the use of LBM in transition regime (nano scale) [19–26]. In our previous work, we proposed a modified thermal lattice LBM, in which a new effective relaxation time is defined as a function of the Knudsen number, which can simulate wide range of thermal flow regimes. For a brief introduction to this model, please refer to [25,26]. In the present work, one of our goals is to assess the validity of the modified LBM predictions for nanoscale flow and heat transfer in porous media by comparing the MD and modified LBM results in terms of dimensionless quantities.

The other aim of the present work is to develop a molecular dynamic model for nanoscale porous applications and investigate the effect of various parameters such as driving force, porosity and Knudsen number, $Kn$, on the velocity and temperature distributions. There are some MD studies that investigate the effect of various parameters on nano-channel flows.

Sofos et al. [27] investigated the effect of temperature and hydrophilic forces on fluid velocity. They found that increasing temperature leads to increase of velocity but the slip length does not
Navier–Stocks and Darcy–Brinkman equations may not lead to proper results. Non-continuous effects increased by decreasing the characteristic length of channel. This issue was investigated by Travis and Todd [31]. They investigated the flow of spherical and uniaxial particles in a channel with width of 1.5 and 10.2 times molecular diameter via MDS and compared the obtained results with predictions of Navier–Stocks equations. Their results show that as the channel width decreases, the differences between the results obtained from MDS and those of the Navier–Stocks equations, increases.

For the porous structure, MDS is usually used to investigate the flow in membranes. In general, several simulations are done in this field via MDS and the related effects are investigated in the non-continuous medium flow. Chang et al. [1], studied Helium, Nitrogen and carbon dioxide gas flows in a silicon porous membrane using hybrid MDS and Direct Simulation Monte Carlo, DSMC, methods. They illustrated the effect of pressure and temperature on flow characteristics.

Also, Pohl et al. [32], modeled Helium, Hydrogen and Argon in a Poiseuille flows in a silicon porous membrane using MDS. They compared properties of flow in zeolite micro-porous membranes with amorphous silicon membranes. In another work, Chang et al. [2] studied adsorption and surface diffusion phenomena in a silicon membrane using MDS and taking into account the number of adsorbed atoms. They investigated permeability of carbon dioxide gas.

Zhang et al. [33] used MDS for computing liquid Argon shear viscosity in a porous media at different temperatures, densities and porosities. They also presented correlations for predicting simple liquids viscosity as a function of temperature, density and porosity. Thus, it is quite necessary to develop a molecular dynamic model for nanoscale porous applications. By using the LAMMPS code, this paper MDS for simulating gas flow in porous media and investigates the effect of various parameters such as driving force, porosity and Kn on the velocity distribution. The current study demonstrates the good ability of MDS for investigating different flow phenomena in porous media such as Darcian and Forchheimer flow regimes.

2. Molecular dynamic simulation

In nano-scale flows, molecular structure of the fluid and surfaces, and their interactions potentials between them at the atomistic length scales play a key role, and hence the Molecular Dynamics Simulation (MDS) emerges as a viable approach for investigation of the flow physics in such scales [34–36]. Recent developments in MDS of the nanoscale fluid flow are summarized in [37,38]. As described in these two review articles, one of the primary interests in the nanoscale fluid flow is the investigation of fluid/surface interactions and their implication on the boundary conditions applicable for continuum level formulations. The flow behavior at the submicron scale strongly depends on the boundary conditions at the solid–liquid interface.

The MDS starts from the integration of Newton’s second law. Applying appropriate integral methods, such as Verlet method algorithm, the basic dynamics parameters such as position, velocity, and interaction force can be determined. The macroscopic physical properties such as pressure, velocity, temperature, and particle number density can subsequently be determined via statistical mechanics.

In MDS, first initial velocities and positions of molecules are assumed. Then the forces between the molecules are calculated and Newton’s second law is solved for each molecule to update their positions and velocities. The solution is continued until the equilibrium state is reached.

On the basis of the MD method, the Newtonian equation (Eq. (1)) for each fluid molecule is written as:

$$m_i \frac{d^2 \mathbf{r}_{ij}}{dt^2} = \sum_{j' \neq j}^{N} \mathbf{F}_{ij} + \mathbf{F}_{\text{external}}$$

where $m_i$ is the mass of the $i$-th molecule, $\mathbf{r}_{ij}$ is the position vector of the $i$-th molecule relative to the $j$-th molecule, $\mathbf{F}_{ij}$ is the interaction force between the $i$-th and $j$-th molecules, and $\mathbf{F}_{\text{external}}$ is the external force acting on the $i$-th molecule.

### Nomenclature

- $F$: Interaction force (N)
- $F_{\text{ext}}$: External force (N)
- $\phi_{ij}$: Interaction potential (J)
- $m$: Mass of molecules (kg)
- $N$: Number of molecules (Non-dimensional)
- $\varepsilon$: Depth of Lennard-Jones potential wall (J)
- $\sigma$: Diameter of molecules (m)
- $r_c$: Cut-off radius (m)
- $r_{ij}$: Distance between $i$th and $j$th molecules (m)
- $T$: Temperature (K)
- $t$: Time (s)
- $u$: Molecules velocity (m/s)
- $U$: Molecules velocity in $x$ Direction (m/s)
- $\tau$: Characteristic time (Non-Dimensional)
- $\rho$: Density (kg m$^{-3}$)
- $Kn$: Knudsen number
- $k_B$: Boltzmann constant

### Subscripts

- *: Dimensionless quantity
- W: Wall
- X: $X$-direction
- Y: $Y$-direction

Numerous studies show that for free molecular flow regime, $Kn > 10$, the continuum hypothesis may not be valid, thus the Navier–Stocks and Darcy–Brinkman equations may not lead to proper results. Non-continuous effects increased by decreasing the characteristic length of channel. This issue was investigated
Fig. 1. Schematic of lattices commonly used in 3-D and 2-D simulations.

Fig. 2. Porous structure of $\varepsilon_s = 0.732$.

where $F_{\text{ext}}$ is the inlet external force per molecule. Applying a force to molecules implies a change in their potential energy as they move due to the applied force field. The forces ($F_{ij}$) calculate from potential function (Eq. (2)).

$$F_{ij} = \nabla \phi_{ij} = \frac{\partial \phi_{ij}}{\partial r_{ij}}.$$  \hspace{1cm} (2)

The Leonard-Jones, LJ, potential as a two-body potential model is the most commonly used and simple interaction potential. It is given by:

$$\phi (r_{ij}) = 4\varepsilon \left[ \left( \frac{\sigma}{r_{ij}} \right)^{12} - \left( \frac{\sigma}{r_{ij}} \right)^{6} \right], \hspace{1cm} r < r_c$$  \hspace{1cm} (3)

where $\varepsilon$ is the depth of the potential energy, $\sigma$ is the molecular diameter and $r_c$ is distance which the inter particle potential is negligible. The first term represents the short-range repulsive interactions preventing the overlap of the molecules, while the second term represents a dipole-induced attractive interaction.

Many nanoscale fluid flow simulations had the fluid confined between two walls, and applied periodic boundary conditions in the flow direction. In most of the earlier simulations, standard LJ is applied to all molecules in the computational domain and also, the FCC and SQ2 lattices structure are used in almost all the 3-D or 2-D studies. FCC and Sq2 lattices are shown in Fig. 1. The channel’s height is $40\sigma$, the ratio of the channel length, $L$, to the height is 2.5. The wall is platinum, Pt. Argon molecules confined between Pt wall atoms. The periodic boundary condition is imposed at the inlet and outlet of the channel. The walls and fluid domains arranged on sq2 lattice. The temperature is set to $k_B T = 1$. This represents a compressed fluid about 30% above the melting temperature [33]. The temperature in an N-particle system can be defined through the average kinetic energy.

The molecular mass of Argon is $m = 6.69 \times 10^{-26}$ kg, molecular diameter is $\sigma = 0.3405$ nm and binding energy is $\varepsilon = 1.6539 \times 10^{-21}$ J. For computational efficiency the intermolecular interaction forces are truncated to zero at a cut-off distance of cut-off $r_c = 3\sigma$. Coefficients of the Lennard-Jones potential function are represented in Table 1. Atomic interactions (liquid/liquid, solid/solid and liquid/solid) are described by LJ type potentials give in Table 1.

3. Results and discussion

The simulated 2-D porous media of platinum are shown in Figs. 2–4. Disordered solid blocks are packed between two parallel plates. Porosity $\varepsilon_s$ represents the fraction void volume in the porous structure and it is defined as $\varepsilon_s = (V - V_s) / V$, where $V$ is the total volume, and $V_s$ is the volume of the solid blocks. Isothermal gaseous flows of argon in such nanoscale porous structures are simulated with the presented MD method. It flows in the x direction due to the external force applied on the system. The external driving force is applied along the x-direction to every fluid particle during the simulation. The work produced due to this external force $F_{\text{ext}} = 0.2 \frac{\varepsilon}{\sigma^2}$, $0.3 \frac{\varepsilon}{\sigma^2}$ and $0.4 \frac{\varepsilon}{\sigma^2}$ is converted into heat that must be removed from the fluid through the channel walls to limit the temperature rise. Whenever a particle attempts to cross the boundaries, it is reflected back. The effect of the wall temperature is achieved by randomizing the direction of the reflected velocity (diffusive) and scaling its magnitude to match the boundary temperature where a thermostat is connected to the system. The equations of motion are integrated using a Velocity Verlet algorithm. In order to find the position and velocity of molecules at the next time step, evaluation of intermolecular forces are required. Detailed forces of interactions experienced

<table>
<thead>
<tr>
<th>Type of interaction</th>
<th>$\varepsilon$ (J)</th>
<th>$\sigma$ (m)</th>
<th>$r_c$ (m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ar–Ar</td>
<td>$1.67 \times 10^{-21}$</td>
<td>$3.4 \times 10^{-10}$</td>
<td>2.5$\sigma$</td>
</tr>
<tr>
<td>Pt–Pt</td>
<td>$0.894 \times 10^{-21}$</td>
<td>$3.085 \times 10^{-10}$</td>
<td>2.5$\sigma$</td>
</tr>
<tr>
<td>Ar–Pt</td>
<td>$1.10538 \times 10^{-21}$</td>
<td>$3.2425 \times 10^{-10}$</td>
<td>2.5$\sigma$</td>
</tr>
</tbody>
</table>

Table 1 Coefficients of the Lennard-Jones potential function.
by each of the fluid molecule and wall molecules are calculated separately by using interaction potentials between fluid molecules and fluid–wall molecules.

Fig. 5 compares our results for non-dimensional axial velocity with those of the Xu and Zhou [29]. The dimensionless lengths of calculation domains are 12, 12 and 16. 1372 Argon molecules and 1600 solid molecules of top and bottom wall regions placed.

The materials of both top and bottom solid walls are Platinum; the molecules are arranged in face–centered-cubic structures. The distance between adjacent solid molecules is $2.77 \times 10^{-10}$ m. The Lennard-Jones potential performed in this research with these detail: $\sigma = 3.4 \times 10^{-10}$ m and $\varepsilon = 1.67 \times 10^{-21}$ J. The cut-off distance is set to be $2.5\sigma$. The thermal wall models are employed at the walls. For the non-dimensional gravity force of 5.0, that exert to domain molecules, a time step of $\Delta t = 0.0005\tau$ is used.

A good agreement between the results can be seen, which represents the validation of our results. Note that the non-dimensional axial velocity and driving force can be calculated from the following relations:

$$U^* = \frac{U m}{\varepsilon} \quad (4)$$

$$F^* = \frac{F \sigma \varepsilon}{\varepsilon} \quad (5)$$

where $\sigma$, $\varepsilon$ and $m$ are the molecular diameter, energy depth and molecular mass, respectively. Fig. 6 compares the results of the modified LBM and standard LBM with those of the MDS. The size of LB lattice is $100 \times 40$ in lattice units. Slip and periodic boundary conditions are imposed at solid walls and inlet/outlet boundaries. The figure shows that, similar to the results of our previous works for nano channels, the results of modified LBM have better agreement with those of the MDS in the transition flow regime for porous media. It can be seen from the figure that at lower porosities, properties of atomistic flow show more consistent deviations from the LB results. This is because of the fact that the function $f(Kn)$ in the relation of effective dynamic viscosity of the modified LBM [32,33], depends on the flow geometry. The value $f(Kn) = 2.2Kn$, considered in the present work, is the simplest model for $f(Kn)$ which is obtained for two parallel plates without any solid blocks.

The effect of the driving force on velocity distribution at the channel outlet is shown in Fig. 7 for three porous structures and $Kn = 0.2$. It is found that the mean velocity and the slip near the wall are increased by increasing force, therefore the flow rate is increased but its effect on the slip is weak. Knudsen number is defined as follows:

$$Kn = \frac{1}{2\sqrt{2\pi nl_d^2}} \quad (6)$$
where $d_i$ is the diameter of gas molecule $i$, $l_y$ is width of nanochannel and $n$ is the number of molecules at computation domain.

For creeping flows, moving with a slow steady velocity ($Re < 1$), the pressure drop ($\Delta p$) through medium and the volumetric flow rate per unit cross-sectional area of porous medium ($U$) are related through the empirical Darcy’s law:

$$U = \frac{k \, \Delta p}{\mu \, L} \tag{7}$$

where $k$ is the permeability, $\mu$ is the dynamic viscosity of the fluid and $L$ is the thickness of the porous medium. According to Darcy’s law, the mean velocity and pressure drop have a linear relation.

The effect of driving force on mean velocity (flow rate) is shown in Fig. 8 for Knudsen number 0.2. As it was mentioned, the velocity increases by increasing driving force. However, for porous structures with relatively small porosity, the flow rate is affected to a less degree than those having higher levels of porosity. For example, the flow rate discrepancy is relatively small between $\varepsilon = 0.805$ and $\varepsilon = 0.732$.

When the driving force is small enough, mean velocity increases linearly with driving force which is in agreement with Darcy’s law because the Reynolds number, $Re$, is about 0.01–0.1, while for higher driving forces, the relationship is no longer linear which shows the Forchheimer flow regime. This plot shows that different flow regimes in porous media (Darcian and Forchheimer) can be simulated using MDS. Also, it can be seen that by increasing porosity, the deviation from linear status occurred sooner. This is because of the fact that the velocity increases by increasing porosity and so the flow gets out of creeping status sooner.

The porosity effect on the velocity distribution at channel outlet is shown in Fig. 9. By increasing the porosity, the obstacles in the flow direction decrease and the velocity increases.

The Kn effect on velocity distribution at channel outlet is shown in Fig. 10. In this study, the change in Knudsen number is achieved by change in fluid particle number which is due to the change in lattice parameter. The parameters related to each case are shown in Table 2 beside related Knudsen number. The applied driving force is exerted on the whole particles. Therefore, by increasing the Knudsen number, the force exerted on each particle increases, thus the velocity increases. Furthermore by increasing Knudsen number the slip velocity near the walls should be increased but, because of the change in wall particles number, the interaction between fluid particles and wall particles also changes and so some reduction in slip velocity near the wall can be seen. According to the figure, by increasing the Knudsen number the mean free path and the freedom of particles increases,
Fig. 7. Velocity distribution across the channel width for various driving force and porosities, $\varepsilon$, (a) 0.805 (b) 0.881 (c) 0.732.

Fig. 8. Axial velocity distribution for different porosities for Darcian and Forchheimer flow regimes.

Table 2
Parameters related to Fig. 10.

<table>
<thead>
<tr>
<th>Porosity</th>
<th>Lattice parameter</th>
<th>Number of atoms</th>
<th>$\kappa n$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\varepsilon = 0.881$</td>
<td>0.5</td>
<td>1707</td>
<td>0.5</td>
</tr>
<tr>
<td>$\varepsilon = 0.881$</td>
<td>0.8</td>
<td>2895</td>
<td>0.3</td>
</tr>
<tr>
<td>$\varepsilon = 0.805$</td>
<td>0.5</td>
<td>1545</td>
<td>0.6</td>
</tr>
<tr>
<td>$\varepsilon = 0.805$</td>
<td>0.8</td>
<td>2720</td>
<td>0.3</td>
</tr>
<tr>
<td>$\varepsilon = 0.732$</td>
<td>0.5</td>
<td>1390</td>
<td>0.6</td>
</tr>
<tr>
<td>$\varepsilon = 0.732$</td>
<td>0.8</td>
<td>2558</td>
<td>0.3</td>
</tr>
</tbody>
</table>

Therefore the oscillations in the velocity distribution function increase.

In Fig. 11 the temperature distributions, obtained via the MD and modified LBM, are compared for a nanochannel without obstacles. Square symbols and dashed line are corresponding to the results of MDS and modified LBM respectively, while the solid line is the curve fit of the MDS results. Important parameters of the molecular dynamic simulation for Figs. 11–13 are listed in Table 3. Good agreement between the results is observed which demonstrates the validity of MD results. However, a little discrepancy appears very close to the walls because for the temperature jump boundary condition of LBM the assumption is
that the wall surfaces completely diffuse while it may be incorrect physically.

Fig. 12 shows the temperature distributions for $Kn = 0.44$. Symbols and dashed line are the results of MDS and modified LBM respectively and the solid line is the polynomial regression of the MD results. As it is mentioned in Ref. [32], by increasing $Kn$, the accuracy of the modified LBM decreases and so, the agreement between the MD and generalized LB results, reduces. Comparison between Figs. 12 and 13 shows that by increasing $Kn$ the fluctuations in the MD results increases.

Fig. 13 shows the $Kn$ effect on the temperature distribution across the nanochannel. Symbols are the results of MDS and dashed line and solid line are the polynomial regression of the results. Increasing $Kn$ has two opposite effects on the fluid temperature.

The first effect is that it leads to increase in velocity. Therefore larger amount of heat is transferred from the fluid to the walls due to increase in convection effects, so the fluid temperature decreases. The second effect is that it leads to increase in molecular diffusion and kinetic energy of the fluid particles. Therefore the fluid temperature increases. The figure shows that the effect of molecular diffusion is dominant at the fully developed region of the channel.

Fig. 14 shows the effect of driving force on the temperature distribution at the outlet of the nanochannel filled with porous media. Increasing the driving force, causes the molecular diffusion and the kinetic energy to increase. Therefore the temperature increases.

Fig. 15 shows the effect of porosity on the temperature distribution. Similar to the previous results, by increasing the porosity, the flow velocity increases so the kinetic energy and temperature increases. Attention to Figs. 15 and 16 proves that driving force and porosity have no significant effect on the temperature jump near the walls because the fluid velocity near the walls has no significant changes under the effect of wall absorbents.

### Table 3

<table>
<thead>
<tr>
<th>Figure</th>
<th>$T^*$</th>
<th>Number of Argon particles</th>
<th>$L/\sigma$</th>
<th>$H/\sigma$</th>
<th>$Kn$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fig. 11</td>
<td>120</td>
<td>4416</td>
<td>20</td>
<td>17</td>
<td>0.017</td>
</tr>
<tr>
<td>Fig. 12</td>
<td>120</td>
<td>812</td>
<td>80</td>
<td>20</td>
<td>0.44</td>
</tr>
<tr>
<td>Fig. 13</td>
<td>120</td>
<td>4416</td>
<td>20</td>
<td>17</td>
<td>0.017</td>
</tr>
<tr>
<td>Fig. 13</td>
<td>120</td>
<td>1683</td>
<td>20</td>
<td>17</td>
<td>0.045</td>
</tr>
</tbody>
</table>

**Fig. 10.** Velocity distribution at channel outlet for different $Kn$.

**Fig. 11.** Comparison between temperature distributions obtained via MDS and modified LBM (slip flow regime). $H$ is width of channel.
Fig. 12. Comparison between temperature distributions obtained via MDS and modified LBM (transitional flow regime). $H$ is width of channel.

Fig. 13. $Kn$ effect on the temperature distribution.

Fig. 14. Driving force effect on the temperature distribution for $\varepsilon = 0.881$.

Fig. 15. Porosity effect on the temperature distribution, $F^* = 0.4$.

Fig. 16. Mean velocity along the channel for different porosity and $F^* = 0.2$.

Fig. 16 shows the mean velocity distribution along the channel. Because of the periodic boundary condition, for the same porous structure, the inlet and outlet velocities are equal. The figure shows that with increasing the porosity, the mean velocity increases.

4. Conclusion

A molecular dynamic method is presented which can take into account the influence of gas rarefaction. The transport characteristics of porous media was calculated with two different techniques, namely, with the molecular dynamics method and with the LB method, properly modified to account for the dependence of the gas viscosity on the local Knudsen number. The results show that the MD method has good ability for investigating different flow phenomena such as permittivity and Darcian flow in porous media. It was found that the results of modified LBM have better agreement with those of the MDS in the transition flow regime.

The main findings of this work can be summarized as follows:

- The Knudsen number has significant influence on the gas permeability.
• For the same porous structure, the flow rate increase with the increase of Knudsen number and driving force.
  
• The effect of Knudsen number, driving force and porosity on the slip velocity near the walls compared to their effects on the mean velocity is insignificant.
  
• By increasing the Knudsen number the oscillations in the velocity and temperature distribution functions increase.
  
• It is found that increase in driving force, porosity and Knudsen number causes the increase in the mean velocity and flow rate. Furthermore by increasing the Knudsen number the oscillations in the velocity distribution function increase.

References

  
  
  
  
  
  
  
  
  
  
  
  
  
  
  
  
  
  
  
  
  
  
  
  
  
  
  
  
  
  
  
  
  
  
  
  
  