MD Simulations of Nano-Scale Gas Flows: A Case Study of Couette Flow at *Kn*=10

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Abstract. Utilizing a recently developed 3D smart wall molecular dynamics algorithm, we investigate argon flow in a 5.4 *nm* channel, which corresponds to the free molecular flow regime. Simulation results are compared with the linearized Boltzmann equation solutions. Significant deviations from the kinetic theory predictions are observed for the velocity, density and average normal stress profiles in the near wall region, where the wall force field effects are dominant. The results clearly show incomplete similitude between the rarefied and nano-scale gas flows, if solely based on the Knudsen number.

Keywords: wall force field effects; normal stresses; shear stress; free molecular flow.
PACS: 47.45.-n, 34.35.+a.

INTRODUCTION

Nano-scale confined gas flows are encountered in the components of micro- and nano-electromechanical systems, and in magnetic disc drive units [1, 2]. For the latter, distance between the head and media is on the order of ten nanometers, and the next generation disc drives strive to reduce this distance to enhance the magnetic storage capacity. Classical approach to study gas flows in such small scales utilizes the kinetic theory. Comparison of the characteristic domain size, H, with the local gas mean free path, λ, enables definition of the Knudsen number ($Kn = \lambda/H$), which shows the extent of the nonequilibrium and/or rarefaction effects. The flow is categorized to be in the free molecular flow regime for $Kn \geq 10$; in the transition flow regime for $0.1 \leq Kn \leq 10$; and in the slip and continuum flow regimes for $0.01 \leq Kn \leq 0.1$, and $Kn \leq 0.01$, respectively [3]. The mean free path for air at standard conditions is about 65 *nm*. As a result, the aforementioned nano-scale scale confined flows are mostly in the transition and free molecular flow regimes. The kinetic theory based analyses of these flows utilize analytical and numerical solutions of the Boltzmann equation [4-6], and the direct simulation Monte Carlo method [7].

A fundamental limitation of the kinetic theory based approaches in nano-scale gas transport investigations is the negligence of surface force field effects. Even for the most simplified case of atomistically smooth non-charged surfaces, van der Waals force field interactions between the wall and gas molecules would induce local variations within the *wall force field penetration length*, which is typically one nanometer from each wall. As a result, 40% of a 5.4 *nm* channel would experience wall force field effects, within which the transport could significantly deviate from the kinetic theory predictions.

Accurate modeling of the wall force field effects requires utilization of molecular dynamics (MD) simulations in three-dimensional computational domains, spanning at least one mean free path per periodic direction. This requirement is important in order not to affect the gas intermolecular collisions, characterized by λ. As a result, MD simulations of gas flows are overwhelmed by the excessive number of wall molecules, and quickly become computationally prohibitive. For this reason MD based studies of nano-scale confined gas flows are quite limited in the literature. In order to address this limitation, we developed a smart-wall MD (SWMD) algorithm, which significantly limits the computer memory utilized by the walls to a stencil of 74 wall molecules for three-dimensional FCC crystal, (1,0,0) plane facing the gas molecules [8].

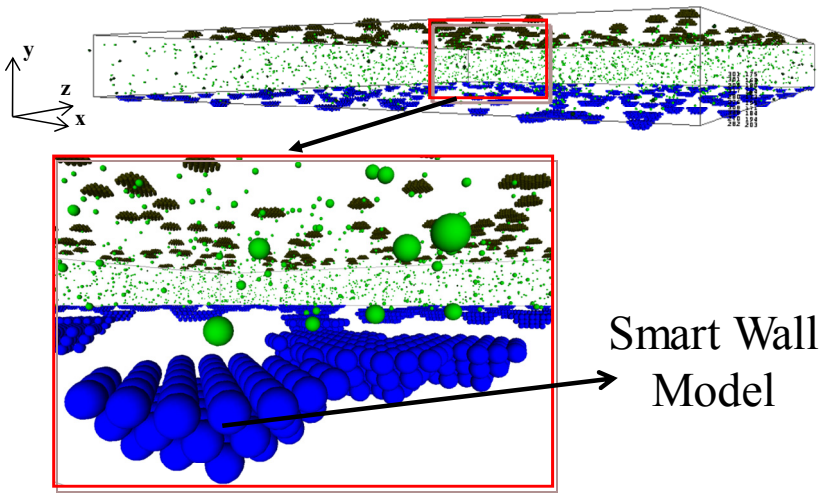

FIGURE 1. Snapshot of three-dimensional SWMD gas flow, and the coordinate system used in this work.

3D MD SIMULATION DETAILS

We performed SWMD with the isothermal ensemble (NVT) by utilizing (6-12) Lenard-Jones potential for van der Waals interactions. SWMD enables incorporation of wall force field effects in realistic conditions and computational domain sizes [8]. We consider here flow of argon gas between two infinite parallel plates that are H = 5.4 *nm* apart, and moving in opposite directions with a characteristic velocity of U_w = 64 *m/s*. The mass for an argon molecule is $m = 6.63\times10^{-26}$ *kg*, its molecular diameter is σ = 0.3405 *nm* and the depth of the potential well for argon is $\varepsilon = 119.8\times k_b$, where k_b is the Boltzmann constant (1.3806×10^{-23} *J K*$^{-1}$). For simplicity, we utilized classical FCC structured walls having same molecular diameter of argon ($\sigma_{\text{wall}} = \sigma_{\text{argon}}$). Also, potential strengths for gas/gas and gas/wall interactions are chosen to be equal to each other (i.e. $\varepsilon_{wall\text{-}argon}/\varepsilon_{argon\text{-}argon}$ =1). The (1,0,0) plane of FCC wall structures faces the gas. Periodic boundary conditions are applied in the axial (x) and lateral (z) directions. Overall the computational domain spans 5.4 *nm* ×54 *nm* ×54 *nm* ($H\times\lambda\times\lambda$). Based on our previous work, one mean-free-path long domain sizes in the periodic dimensions are sufficient to obtain domain-size independent molecular dynamic (MD) solutions for gas flows [8]. In order to implement the NVT ensemble at constant temperature of 298 *K*, we initially utilized the Nose-Hoover thermostat in the entire domain. However, this approach resulted in significant temperature variations near the wall with increased wall speed (not shown for brevity). In order to maintain a constant temperature system, we divided the flow domain into 10 sub-domains with equal number of molecules (i.e., the Hamiltonian of each sub-domain was the same); and applied the Nose-Hoover thermostat for each sub-domain. This approach was able to maintain constant temperature in the entire flow domain. The number density of gas molecules at 298 *K* and 112.5 *kPa* is 2.86×10^{25} *m*$^{-3}$.

The computational domain is divided into 100 bins of approximately σ/10 in size. Simulations started from the Maxwell-Boltzmann velocity distribution for gas molecules, and ran 10^6 time-steps (4 *ns*) to reach a steady state using 4 *fs* (~0.002τ) time steps, after which, another 2×10^6 time steps (8 *ns*) were performed for time averaging. Longer time averaging has also been performed to confirm convergence of the density and velocity profiles to the steady state. Particularly, the simulation times are compared with the mean collision times (predicted by the ratio of the mean free path λ to the mean thermal speed $c_m = \sqrt{8RT/\pi}$) to result in a state amenable for time or ensemble averaging.

COUETTE FLOW IN A NANO-CHANNEL

In **FIGURE 2-a,** we show the velocity profile within half of the nano-channel. For comparison purposes, the velocity profiles obtained using linearized Boltzmann equation at Kn =10 subjected to the tangential momentum accommodation coefficients (TMAC) of α=1 and α=0.75 are also presented. The MD based velocity profile agrees

well with the kinetic theory predictions that are adopted from reference [4], within middle of the channel. However, the MD results show sudden changes within approximately 1*nm* away from the surface. This distance corresponds to 3σ, and it is due to the influence of the wall force field region. The velocity components in y and z directions have zero average values confirming one dimensional flow (not shown for brevity). The figure includes a schematic of the wall and gas molecules, to indicate the scales involved in the problem. Particularly, the wall is defined at the center of the first row of wall molecules facing the fluid. Although gas velocity increases up to 40 *m/s* very near the wall, the gas molecules cannot penetrate to the several bins neighboring the wall, and hence, the gas velocity goes to zero on the wall. Extrapolation of the velocity profile to the wall indicates "almost" no-slip conditions for this particular case, which could be due to our choice of $\varepsilon_{wall\text{-}argon}/\varepsilon_{argon\text{-}argon}=1$. However, the linearized Boltzmann equation solutions at different TMAC values indicate substantial amounts of velocity slip on the wall. **FIGURE 2-b** shows the density distribution within the channel. Density is a constant in the bulk of the channel. However, it increases within the 3σ region due to the wall force field effects. Particularly, the density starts to deviate from its bulk value around 2.5σ from the wall. Once again the gas molecules cannot penetrate to the bins neighboring the wall, and hence, the gas density goes to zero on the wall.

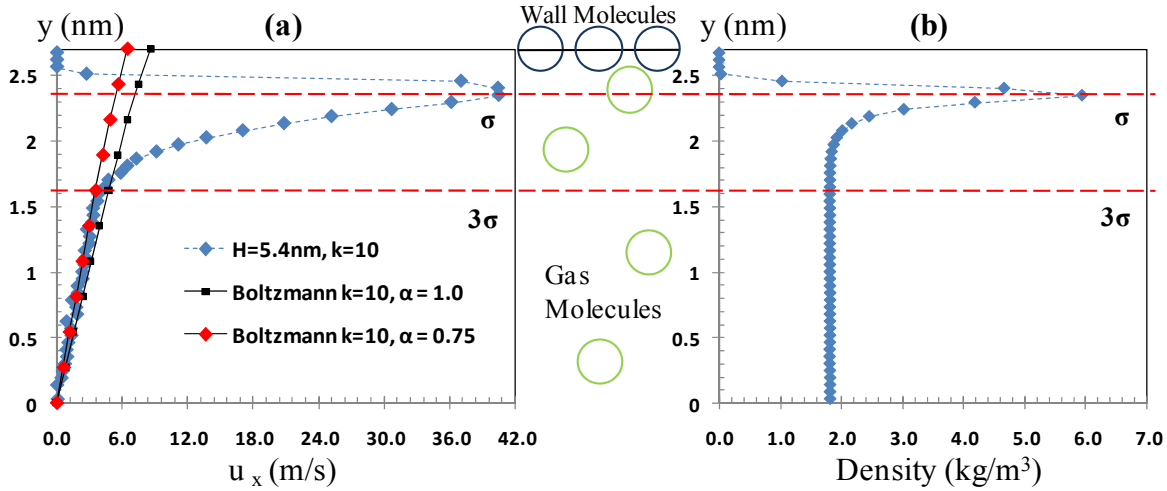

FIGURE 2. (a) Velocity (u_x) distribution in the half of the 5.4*nm* height channel with linearized Boltzmann solution [4]. Wall velocity is equal to 64 m/s. (b) Density distribution in 5.4*nm* height channel. Schematic view of wall position and gas molecules are shown for scale comparison purposes.

Density variations within the nano-channel are induced by the wall force field. It is important to investigate the effects of these force fields within the perspective of normal stresses, which can be eventually related to pressure for this simple compressible system. We utilized the Irving-Kirkwood (I-K) expression to compute the stress tensor components [9]. Average streaming velocity within each bin was initially calculated, and subtracted from the local molecular velocities in the calculations. I-K expression has kinetic and virial components. The kinetic component is a function of the particle velocities, while the virial is an internal contribution from intermolecular forces between the molecules. **FIGURE 3** shows the distribution of mutually orthogonal normal stresses (S_{xx}, S_{yy}, S_{zz}) in the channel. In the bulk flow region, the normal stresses are isotropic. Virial terms in the I-K expression is computed separately and their contributions are less than 0.05%. As a result, pressure can be defined as the average of three normal stresses, and its value can be predicted using the ideal gas law inside the bulk flow region. However, the wall creates anisotropic normal stresses inside the force penetration region.

Shear stress variation within the domain is shown in **FIGURE 4-a**. The attractive component of wall force field increases the shear stress in the near wall region, similar to the behavior of density and normal stresses. The attractive wall force field is the primary reason of inducing the shear driven flow. Therefore, we expect to observe a relation between the gas/wall interaction potential strength and the resulting shear stress in the system. Based on the kinetic theory of gases, Couette flow shear stress in the free molecular flow regime ($Kn \rightarrow \infty$) can be written as [10]

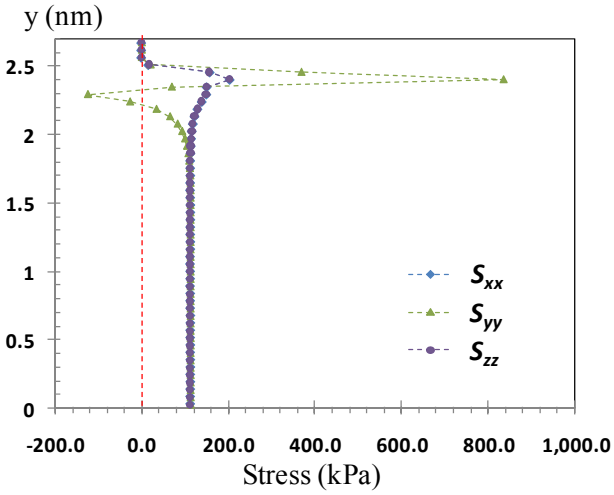

FIGURE 3. Distribution of normal stresses in the channel.

$$\tau_{\infty} = -\frac{\alpha}{2-\alpha}\rho U\sqrt{\frac{RT_w}{2\pi}} \tag{1}$$

where α is the tangential momentum accommodation coefficient (TMAC), ρ is the density, U is the relative velocity between walls, R is the specific gas constant for Argon (208.132 $J\,K^{-1}\,kg^{-1}$), and T_w is the wall temperature. The TMAC value can be interpreted as α portion of molecules reflecting diffusively, while and (1-α) portion reflecting specularly. As can be seen, pure specularly reflecting wall will not drive the fluid, and result in zero shear stress, while lower TMAC values will result in lower shear stress. We cannot directly apply equation (1) to predict the TMAC value for our simulations at Kn = 10. In order to consider the finite Knudsen number effects we utilized the following correction uniformly valid in the free molecular to slip flow regimes [7].

$$\frac{\tau}{\tau_{\infty}} = \frac{0.5297Kn^2 + 1.206Kn}{0.5297Kn^2 + 1.6277Kn + 0.603} \tag{2}$$

Utilizing equations (1) and (2) we predicted the TMAC value for our simulations to be α=0.75. This result shows that the atomically smooth FCC wall structures using $\varepsilon_{wall\text{-}argon}/\varepsilon_{argon\text{-}argon}$ = 1 does not exhibit full diffusive reflection. Our studies with other ε ratios (not shown for brevity) have consistently shown that the TMAC value and shear stress decreases with decreased $\varepsilon_{wall\text{-}argon}$ value. We verified the consistency of our shear stress results with the velocity profile. **FIGURE 2-a** had shown the kinetic theory prediction of velocity distribution for Kn = 10 flow at TMAC values of α=1 and α=0.75. In **FIGURE 4-b** we zoom on the velocity profiles shown in **FIGURE 2-a**, and observe that the linearized Boltzmann solution for α=0.75 matches the MD based velocity predictions better than the α=1 case in the bulk flow region. Further investigations of different Kn flows are also performed, and TMAC values are found to be independent of Kn (Not shown for brevity).

CONCLUSIONS

The SWMD results of shear driven channel flow at Kn=10 have revealed significant wall force field effects on the velocity, density and normal stress distributions in the near wall region, which is about 3σ (=1.08 *nm*) for the Lennard-Jones 12-6 potential. Outside the "near wall" region kinetic theory based predictions are valid. Emergence of the wall force field effects show incomplete similitude between the rarefied and nano-scale gas flows, if solely based on the Knudsen number. Therefore, nano-scale confined gas flows also experience near wall force field effects. Given this fact, one can define a new dimensionless parameter as the ratio of the force field penetration and

characteristic channel length scales. Wall force field effects cannot be neglected for large values of this dimensionless number.

Another important aspect of our findings is that one can predict the TMAC value using 3D SWMD simulations. As a result, atomistically smooth FCC crystal walls with (1,0,0) plane facing the gas resulted in TMAC value of 0.75 for the $\varepsilon_{wall\text{-}argon}/\varepsilon_{argon\text{-}argon} = 1$ case presented here. We have also verified that the TMAC value is independent of the Knudsen number (not shown for brevity). Our future work will concentrate on characterization of TMAC values as a function of the $\varepsilon_{wall\text{-}argon}$ value. Finally, we have shown that the ideal gas law is applicable in the bulk flow region. However, the wall force field induces anisotropic normal stresses in the near wall region.

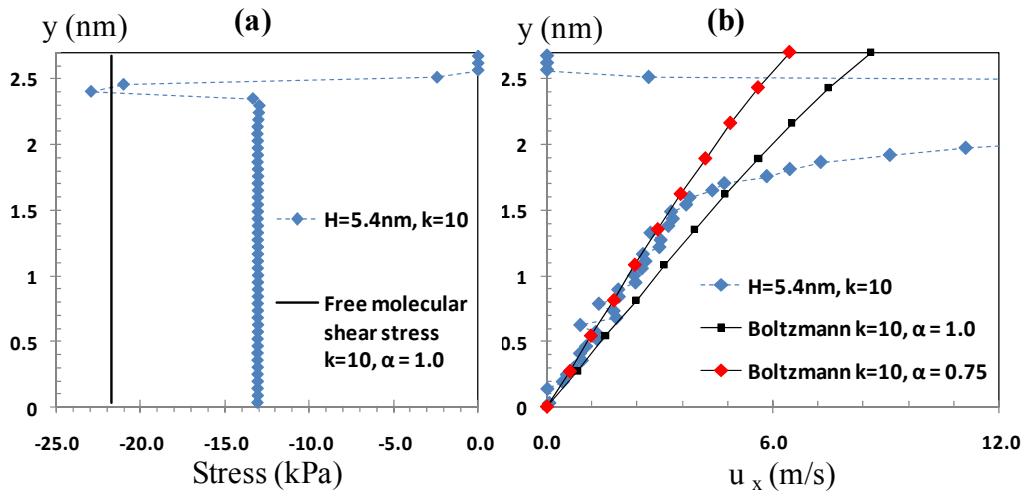

FIGURE 4. (a) Shear stress distribution in 5.4*nm* height channel. (b) A zoomed version of the velocity distribution in the channel verifies that the MD results match better with the linearized Boltzmann solutions using TMAC value of α=0.75.

ACKNOWLEDGMENTS

This work was supported by the National Science Foundation under Grant No: DMS 0807983.

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