Implementation of SMC averaging method in a channeled molecular flow of liquids and gases

S M H Karimian\textsuperscript{1} and S Namvar\textsuperscript{2}
Center of Excellence in Computational Aerospace Engineering, Department of Aerospace Engineering, Amirkabir University of Technology, Tehran, Iran

E-mail: hkarim@aut.ac.ir\textsuperscript{1}, soheil_snp@aut.ac.ir\textsuperscript{2}

Abstract. This paper proposes the application of SMC (SAM-Modified-CAM) method of averaging, formerly developed by Karimian et al, in a channelled molecular flow. While they have used periodic boundary conditions in all three directions, in this paper wall boundary condition is used in one direction and in the other two directions, periodic boundary conditions are applied. In order to verify the results obtained from SMC averaging method for the above boundary conditions, velocity profile extracted here is compared to the one obtained by Sofos et al. Results show that very good agreement is achieved. In addition, same trends reported in the work of Sofos et al for the change of velocity magnitude with the spring stiffness and also with the interaction ratio \(k_c/k_w\), have been observed in the present study as well. It is worth to mention that the velocity profile calculated by the present averaging method is much smoother than the one obtained by Sofos et al. This approves that SMC averaging method is capable of being used for cases with wall boundary conditions. To check whether the simulation has reached equilibrium, “convergence history” of maximum velocity in the channel is plotted using both SMC and CAM methods of averaging. A comparison between the two proves that SMC yields a much smoother graph which is more suitable for determination of the time step in which the equilibrium occurs. It can be concluded that SMC method of averaging is a reliable way of data sampling in molecular dynamics simulations.

1. Introduction
In MD simulation where discrete molecules are modeled rather than continuum flow, finding an appropriate way to calculate mean flow characteristics of molecules is inevitable. Two conventional ways of averaging in MD simulation are sample averaged measurement (SAM) and cumulative average measurement (CAM). Having applied these averaging to obtain molecular flow velocity, the following relations is obtained for SAM velocity of N molecules in a selected domain [2]

\[
\langle \vec{V}_s \rangle_{SAM} = \frac{1}{S} \sum_{j=1}^{S} \vec{V}_{av}(t_j)
\]

Where \(S\) denotes the total number of time steps, \(t_j\) indicates that the corresponding variable is a function of time at \(j\)th time step, and \(\vec{V}_{av}\) is defined as

\[
\vec{V}_{av}(t_j) = \frac{1}{N(t_j)} \sum_{p=1}^{N(t_j)} \vec{v}_p(t_j)
\]
In which $\vec{V}_p$ is the velocity vector of a molecule in the selected domain. Moreover, the CAM velocity would be [2]

$$\langle \vec{V}_s \rangle_{\text{CAM}} = \frac{1}{S} \sum_{j=1}^{S} N(t_j) \vec{V}_{av}(t_j)$$

S.M.H Karimian et al [2] have done a comparative study on SAM and CAM method of averaging for the calculation of velocity. They have implied that to obtain averaged molecular property (e.g. velocity) within a selected domain using SAM method, the molecular property is first averaged within the domain and then these average velocities are averaged over the time. This makes SAM highly sensitive to the number of molecules and their thermal velocity. As the number of molecules increase, SAM gives more realistic results. In fact, SAM is vulnerable to statistical errors for low density flows. In contrast, averaging in CAM is performed cumulatively over the molecules during past time steps which means that the number of molecules taken into account increases continuously. However obtaining flow quantities using these methods will be accompanied by large fluctuations. Thus it is difficult to distinguish whether the quantity in question is converged. To resolve this problem they proposed a more efficient method of averaging called SMC (SAM-Modified-CAM) which could be defined as follows [2]

$$\langle \vec{V}_s \rangle_{\text{SMC}} = \frac{1}{S} \sum_{q=1}^{S} \langle \vec{V}_q \rangle_{\text{CAM}}$$

The SMC method of averaging has been validated through different test cases in Ref. 2. Judging by the statistical error in all test cases, it is concluded that SMC method is superior to SAM and CAM. This is because SMC method damps out the oscillations of velocity versus time and makes the curve smoother.

In many practical cases in which molecular flow is involved, such as simulating flow in nanotubes, wall boundary conditions should be applied. For this reason, we would like to study the implementation of SMC method in a channeled molecular flow, where the flow is bounded with two layered molecular walls. Fortunately, since the results in the literature where wall boundary B.Cs are used are more than the ones in which periodic B.Cs are applied, comparison of the present results will be straightforward [3-9]. In this paper, flow of 576 molecules of liquid argon with $\rho^* = 0.642 \sigma^{-3}$ in a channel is studied. At each region velocity is evaluated using SMC method of averaging. For the channel mid-point (in all directions), time variation of velocity is plotted to show that how SMC well affects data sampling. Although, the simulation carried out in this paper is for liquid Argon, the implementation of SMC method of averaging is also applicable to molecular flow of gases (low density flows).

2. Molecular modeling and Solution Procedure

This paper presents the simulation of 576 argon molecules which are conducted in a 36.15 Å channel. The Knudsen number of the flow is 0.04 and the pressure is 50 Mpa. The velocity direction of each molecule is determined using Maxwell-Boltzmann distribution function. The interaction between molecules is described by Lennard-Jones 12-6 potential equation:

$$U_{ij} = 4\varepsilon \left( \frac{\sigma}{r_{ij}} \right)^{12} - \left( \frac{\sigma}{r_{ij}} \right)^{6}$$

(1)

Where $U_{ij}$ is the potential energy, $r_{ij}$ is the distance between two interacting molecules i and j, and $\sigma$ and $\varepsilon$ are the distance characteristic length and energy scale, respectively. The intermolecular forces are then calculated from equation 1 through the following relation:

$$F_{ij} = -\frac{dU_{ij}}{dr_{ij}}$$

(2)
And finally the motion of the molecules is determined by Newton's second law through the Verlet’s integrating scheme [10].

As mentioned before, two 2-layered walls are located at Y-constant positions. In the other directions periodic boundary conditions are applied. Each wall molecule is attached to its place with a linear spring to ensure wall solidity. The spring will consequently exert a force of \( F = -K\Delta r \) to every molecule that moves away a distance equal to \( \Delta r \); in this equation \( K \) is the spring stiffness. Figure 1 shows the schematic of initial placement of fluid and wall molecules.

![Schematic of initial placement of fluid and wall molecules](image)

**Figure 1.** Schematic of initial placement of fluid and wall molecules

Wall temperature is kept constant using thermostat. In fact at each time step velocity of wall molecules are rescaled to ensure constant wall temperature. An external force is constantly applied to the fluid molecules in order to develop flow in the channel. It should be noted that all of the initial conditions of the simulation such as temperature, number of molecules, spring stiffness, number of wall layers and external force magnitude are chosen similar to a simulation performed by Sofos et al [9] in order to validate the simulation of this paper. The detailed specifications of the simulation carried out in this paper are presented in table 1.

| Specification                        | Value                  |
|--------------------------------------|------------------------|
| Dimension (Angstrom)                 | 36.15×36.15×36.15      |
| Wall molecules                       | 288 argon molecules    |
| Fluid molecules                      | 576 argon molecules    |
| Spring stiffness ( \( \varepsilon \sigma^2 \)) | 57.15      |
| External force (pN)                  | 0.18                   |
| Thermostat (k)                       | Fixed on 120           |
| Number of bins                       | 15 and 36              |
| Time step (s)                        | \( 10^{-14} \)         |
| Boundary Conditions                  | Periodic in X and Z directions |
|                                      | Wall BC in Y direction  |
3. Results and discussion
As mentioned in Table 1, the solution domain is divided into 15 equal bins in Y direction to evaluate velocity profile. The velocities of molecules are averaged in each bin using SMC method. Based on our examination, this is the minimum number of bins that has resulted in accurate velocity profile. It should be noted that higher number of bins, for instance 36 bins, were also considered and the same results have been obtained; this is shown in figure 2. In this figure velocity profile is plotted after $2 \times 10^6$ time steps using 15 and 36 bins.

![Figure 2. Velocity profile after $2 \times 10^6$ time steps using 15 bins and 36 bins](image)

The effect of the number of time steps of simulation is shown in figure 3. As seen, result of $1.5 \times 10^6$ and $2 \times 10^6$ time steps are almost the same. The relative error between peak values of these profiles is less than 1%.

![Figure 3. Effect of time step on the velocity profile](image)
At this stage results of the present paper are compared with the results of Sofos et al [9] for the same test case in figure 4. Present results are shifted 0.7 nm to the right on the horizontal axis, i.e. in the Y direction of solution domain. Averaging method used by Sofos et al has not been mentioned in his work. In order to make a fair comparison present results are obtained using CAM method. It is worth to mention that approximately the same results are obtained if SMC method is used.

In both cases time step of $10^{-14}$ second is used. The bin values of velocities in the present study (denoted by solid squares) are fairly symmetric, as expected. In contrast, velocity values of Sofos et al are not symmetric at all. As seen, both velocity values are close to each other except at the right side of the peak region, where the present velocity values follow a physically meaningful path. Approximately, 12% difference is noticed in the peak value of velocity. From the above observations it can be concluded that the present velocity values are correct and more realistic than those obtained by Sofos et al.

Although both methods approximately result in the same profiles, SMC reveals its advantage when time variation of velocity (convergence history) is examined to see whether the simulation is reached equilibrium. Convergence history of velocity in the middle bin of the above test case is plotted in figure 5, having used both SMC and CAM averaging methods. These curves are plotted based on every 1000 time step results.

![Figure 4. Comparison of velocity profiles obtained in this paper and by Sofos et al [9]](image_url)
Figure 5. Velocity of the middle bin versus time obtained by both CAM and SMC method of averaging

Convergence history can be used to distinguish whether equilibrium is reached. It is obvious that convergence history using CAM method has oscillations all over its curve. Conversely, the curve obtained using SMC method has almost no oscillations. Obviously any kind of convergence criteria such as $\frac{\Delta V}{V} < \varepsilon$, where $\Delta V$ is the difference between velocity values within some successive time steps and $\varepsilon$ is the maximum acceptable error, will work excellent on the curve obtained from SMC. Due to the excessive oscillations in the curve obtained by CAM, implementation of any kind of convergence criteria using this curve will face substantial difficulties. For a suitable convergence criterion in MD simulation see Ref. 2. Although the velocity difference between the two methods is small, based on the error analysis discussed in Ref. 2 present authors believe that velocity value predicted by SMC is more accurate.

4. Conclusion
A simulation of 576 monatomic argon molecules in a channel with two 2-layered walls is reported in this paper. All of the initial conditions have been chosen similar to [9]. The velocity profiles obtained using SMC and CAM methods are approximately the same. For validating the results the velocity profile of the present paper is compared to the one obtained by Sofos et al. For both velocity profiles the values of velocity are roughly the same except for the peak region. It is also observed that the velocity profile obtained in this paper is much smoother and symmetric. CAM and SMC methods are then used to obtain the convergence history. It is observed that SMC yields a graph with no fluctuations which is suitable for obtaining the time step in which equilibrium occurs. This is while the use of CAM results in a convergence history in which oscillations make the use of convergence condition very difficult. The privilege of SMC method is it damps the oscillations of the quantities in the domain of simulation. Being simple to implement without adding to the CPU time of simulation, the SMC method is preferred when mean flow characteristics are to be calculated.
5. References

[1] Fan X, Phan-Thien N, Teng Yong N and Diao X 2002 Molecular dynamics simulation of a liquid in a complex nano channel flow Phys. Fluids 14 1146-53

[2] Karimian S M H and Izadi S 2010 A study on the measurement of mean velocity and its convergence Int. J. Numer. Meth. Fluids 67 2130-40

[3] Abraham F 1978 The interfacial density profile of a Lennard-Jones fluid in contact with a (100) Lennard-Jones wall and its relationship to idealized fluid/wall systems: A Monte Carlo simulation J.Chem. Phys. 68 3713-16

[4] Koplik J, Banavar J and Willemsen J 1989 Molecular dynamics of fluid flow at solid surfaces Phys. Fluids 1 781-794

[5] Thompson P and Robbins M 1990 Shear flow near solids: Epitaxial order and flow boundary conditions Phys. Rev. A 41 6830-37

[6] Jabbarzadeh A, Atkinson J D and Tanner R I 1997 Rheological properties of thin liquid films by molecular dynamics simulations J. Non-Newtonian Fluid Mech. 69 169-193

[7] Jabbarzadeh A, Atkinson J D and Tanner R I 1997 Wall slip in the molecular dynamics simulation of thin films of hexadecane J. Chem. Phys. 110 2612-19

[8] Bing Mi X and T. Chwang A 2003 Molecular dynamics simulations of nanochannel flows at low reynolds numbers Molecules 8 193-206

[9] Sofos F, Karakasidis T E and Liakopoulos A 2009 Non-Equilibrium Molecular Dynamics Investigation of Parameters Affecting Planar Nanochannel Flows Contem. Eng. Sci. 2 283-298

[10] Verlet L 1967 Computer "Experiments" on classical fluids. I. Thermodynamical properties of Lennard-Jones molecules Phys. Rev. 159 98-103