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New Indirect Method for Calculation of Flow Forces on Immersed Bodies in Molecular Dynamics Simulation

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Abstract In this paper, two different approaches of calculating forces in molecular dynamics simulation are investigated and a new method is presented. Drag force on a carbon nanotube in uniform liquid argon flow is evaluated using these three methods. Nanotube is modeled as a rigid cylinder and all the interactions are calculated by Lennard-Jones potential function. First of all common method of calculating drag by direct summation of forces in flow direction is used to verify the code and simulation. Then the continuum approach of calculating forces using momentum balance and change in flow velocity profile is implemented and investigated. Results of this approach show that the increase in number of bins used for velocity measurement will decrease the difference with direct method about 5%. Nevertheless the continuum approach at the best underestimate the drag force by about 20% of direct summation and confirm the fact that continuum approaches are not necessarily appropriate at nanoscale flows. Finally a molecular momentum balance method is presented and used for calculating drag force. The new presented method works properly and the difference with the direct summation method can be reduced from 30% to less than 1% by increasing the number of time steps used for data averaging.

Keywords: Molecular Dynamics, Flow Forces, Momentum Conservation, Carbon Nanotube

1. Introduction

In recent years, study of fluid flow in nanoscale has been a subject of interest. Due to lack of experiment in nanoscale the role of numerical methods to achieve this goal is significant [1]. At length scales less than ten molecular diameter the continuum theories breaks down in both gas and liquid flows [2]. Therefore the common numerical methods for the simulation of fluid flow cannot be used. To study the behavior of fluid flow in atomistic level, the powerful method of molecular dynamics (MD) simulation can be used. This method is suitable for the simulation of problems in the order of 100nm or less [3].

One of the first researches for understanding behavior of nanoscale flows passing an obstacle was done by Rappaport and Clementi in 1986 [4]. They performed an MD simulation to investigate 2D fluid flow passing a circular obstacle. In this work they concluded that the MD approach may prove to be a valuable tool for probing of the detailed microscopic flow structures. Vergeles *et al.* [5] studied translational and rotational motions of a sphere in a viscous liquid using molecular dynamics simulation. They reported that the exerted drag and torque on a sphere in an effectively unbounded fluid are found to be in agreement with continuum hydrodynamics. Walther *et al.* [6] presented a non-equilibrium molecular dynamics (NEMD) simulation of water flow passing over an array of carbon nanotubes (CNT) with two different diameters at three flow speeds in the interval 50–200 m/s. They showed that the calculated drag coefficient of carbon nanotubes array is in reasonable agreement with that obtained from macroscopic, Stokes-Oseen solution. A more detailed and systematic study on the drag coefficient of

nanotubes is done by Tang and Advani [1]. They performed NEMD simulations to investigate uniform liquid argon flow around carbon nanotube. Direct summation of molecular forces was used to evaluate the drag forces and its coefficients for two different sizes of CNTs in a wide range of flow velocities between 15-320 m/s. They realized that at low speed flows MD results of drag coefficients are larger than those obtained from FE analysis and empirical equations. A 2D NEMD simulation of flow over two side by side molecular cylinders was presented by Ziarani and Mohamad [11]. They studied the effect of different parameters on the flow field and the hydrodynamic forces of the cylinders. Lift and drag forces are evaluated using two different methods; direct summation of molecular forces and a continuum approach. In the continuum approach, they used the velocity profile behind the cylinders to estimate the drag forces from the momentum change of flow passing the cylinders. They reported 25-30% difference between results of two approaches with no details on how the velocity profile is measured and how the integral form of conservation of momentum is applied in MD simulation.

The objective of this work is to study comprehensively different methods of force calculation in molecular dynamics simulations. The approach of continuum evaluation of forces will be investigated in details and will be compared with the molecular method. In addition, a new molecular method originated from continuum approach will be presented for force calculation in this paper.

2. SIMULATION

In this paper amount of parameters and results will be presented in reduced molecular dynamics units. The molecular mass (m) and diameter (σ) of liquid argon are used as units of mass and length, respectively. The ε which is the strength of interaction for Lennord-Jones Potential, is considered as unit of energy. Amounts of these parameters are given in Table 1. Also the units of time, force and velocity are derived using reference molecular units.

Table 1
Molecular units

Quantity	Symbol	Equivalent in SI
Length	σ	3.40×10^{-10} *(m)*
Mass	m	6.625×10^{-26} *(kg)*
Energy	ε	1.657×10^{-21} *(J)*
Time	$\tau \equiv (\sigma^2 m/\varepsilon^{-1})^{1/2}$	2.15×10^{-12} *(s)*
Force	$F \equiv \varepsilon\sigma^{-1}$	4.873×10^{-12} *(N)*
Velocity	$U \equiv \sigma\tau^{-1}$	1.58×10^{2} *(m.s^{-1})*

2.1. Solution domain

In this work argon flow around a carbon nanotube is simulated. As shown in Fig. 1, the system consists of two types of atoms; the argon atoms as fluids, and the carbon atoms forming the carbon nanotube. Domain of simulation is a box with the dimensions of 50×60 molecular diameter in *z* and *y* direction. Domain width in *x* direction equals the length of nanotube. Periodic boundary condition is applied to the solution domain in all directions. A single walled (12, 0) carbon nanotube with the diameter of 2.76 and length of 6.26 is fixed with an offset of $0.1L_y$ from cell center to the inlet. The nanotube is modeled as a fixed body with rigid structure such that carbon atoms do not move relative to each other.

2.2. Initial setup and flow driving

Argon atoms are initially distributed in the channel in a lattice form of FCC with the liquid argon density of 0.8 consists of 14964 argon atoms. Initial thermal velocities of atoms are assigned according to their initial temperature of 95K. Velocity components of atoms are assigned based on Maxwell–Boltzmann distribution that results in a zero resultant velocity and no flow in the

channel. Initial flow is generated by adding a velocity of U_0=1.0 in *y* direction to all argon atoms. To maintain the flow, velocities of atoms located in the $0.03L_y$ of the inlet region are reset every 50 time steps. Resetting or rescaling of velocity is applied by setting the initial random thermal velocity of the atoms in the inlet region and then adding U_0 to their velocities in y direction. In addition to maintaining the flow, this technique removes excess heat from the system as well [8] and there is no need to use thermostat or other techniques.

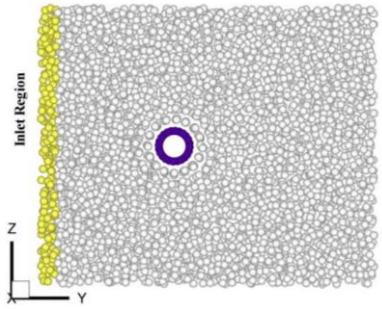

Fig. 1. Schematic of the system

2.3. MD simulation

All the atoms in the system interact by Lennard-Jones (LJ) 12-6 potential function but with different parameters for argon-argon and carbon-argon interactions.

$$V_{ij} = 4\varepsilon\left[\left(\frac{\sigma}{r_{ij}}\right)^{12} - \left(\frac{\sigma}{r_{ij}}\right)^{6}\right] \qquad (1)$$

Where V_{ij} is the LJ potential, ε is the strength of interaction, σ is the molecular diameter of argon and r_{ij} is the distance between a pair of atoms. Based on LJ potential, the interaction force between a pair of atoms is calculated by

$$\boldsymbol{F}_{ij} = \frac{48\varepsilon}{\sigma^2}\left[\left(\frac{\sigma}{r_{ij}}\right)^{14} - \frac{1}{2}\left(\frac{\sigma}{r_{ij}}\right)^{8}\right]\boldsymbol{r}_{ij} \qquad (2)$$

Where $\boldsymbol{r}_{ij}$ is the position vector from atom $\boldsymbol{i}$ to atom $\boldsymbol{j}$ and r_c is the cut-off distance beyond which the forces are neglected. The interaction forces between argon-argon and carbon-argon atoms are computed using the parameters as in Table 2.

Table 2
LJ potential parameters

	Argon-Argon	Carbon-Argon
ε	1.6567×10^{-21} J	1.9646×10^{-21} J
σ	3.4×10^{-10} m	3.573×10^{-10} m

A cut-off distance of 2.5σ is used for both interactions and neighbor list method is used to calculate interactions between atoms. Equation of motion is integrated using a Verlet scheme with a time step of $\Delta t = 0.001\tau$ equals 2.15×10^{-15} s. With these assumptions a parallel MD code is developed using atom decomposition approach to reduce computational time. The simulation is performed for 8×10^5 time steps corresponding to 1.7 ns.

3. RESULTS

First an MD code was developed and the case of Tang and Advani [1] was performed to verify the code. They used MD to calculate the drag force exerted on CNT (12, 0) in uniform liquid argon flow. In order to investigate the effect of domain size on drag force they performed simulations with different domain sizes as listed in Table 3.

Table 3
Different domain sizes

Size	L_y	L_z	Number of Atoms
1	30	25	3741
1.33	40	33	6584
1.67	50	42	10475
2	60	50	14964
2.33	70	58	20251
2.67	80	67	26736
3	90	75	33669

To evaluate the drag force of nanotube, simulations are performed to reach an equilibrium time t_{eq} and then continued to a desired time t to get enough data for extracting forces and other macroscopic properties. Some of the methods reported in

the literature to detect system equilibration are to record instantaneous values of the energies and pressure during this period [9], or to monitor the positional disorder and velocity distribution of atoms [10]. In this work trend of kinetic energy is used to detect the equilibrium time. Change of kinetic energy during the simulation is shown in Fig. 2 for 3 different domain sizes.

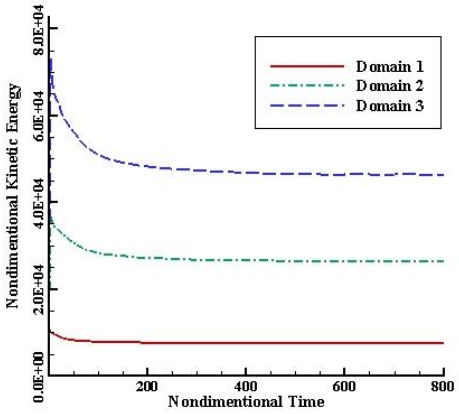

Fig. 2. Kinetic energy during the simulation

As can be seen in Fig. 2, all three domains have reached to equilibrium state after time durations from 100 to 200 nondimensional time which correspond to 2.15×10^{-10} to 4.50×10^{-10} seconds. Therefore t_{eq} equal to 200 is adopted for all the simulations in this work. In all of the cases solution is continued up to 800 nondimensional time.

3.1. Direct Method and Code Verification

In MD simulations, lift and drag forces are calculated directly from the summation of forces exerted on the solid atoms by fluid atoms [1-6]. Forces exerted along the nanotube in *x* direction and along the flow in *y* direction (Drag) are calculated in each time step and are shown in Fig. 3. As seen, both forces fluctuate around their corresponding value. In *x* direction fluctuations are around zero as expected, but in flow direction the drag force fluctuates around a nonzero value.

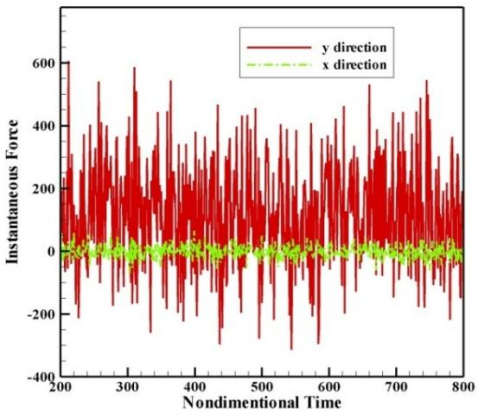

Fig. 3. Instantaneous forces in *x* and *y* direction

Mean value of forces are obtained from the following formulation which averages the instantaneous forces over a period of time after equilibrium.

$$D_{ave}^{dir}(t) = \frac{1}{N_{t_{eq}-t_f}} \sum_{t_{eq}}^{t_f} D_{inst} \qquad (3)$$

Where $N_{t_{eq}-t_f}$ is the number of time steps from equilibrium time, t_{eq}, to final time of averaging, t_f. As mentioned before, in these cases according to the convergence trend of kinetic energy, t_{eq} is set equal to 200. Mean values of forces calculated for the time range of 200 to t_f, where $200< t_f \leq 800$ in three directions are shown in Fig.4. Mean drag force (*y* direction) has converged to its equilibrium value of 115, and in two other directions both forces are about zero as expected.

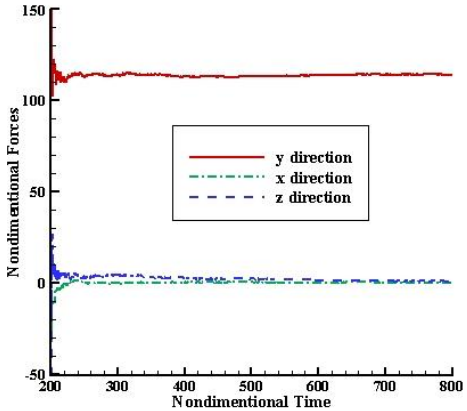

Fig. 4. Averaged forces in *x*, *y* and *z* direction

A thorough verification of present code is done by simulating Tang and Advani's case [1] for all domain sizes of Table 3. Mean drag forces obtained in this study are compared with those of Tang and Advani on domain sizes of Table 3 in Fig. 5.

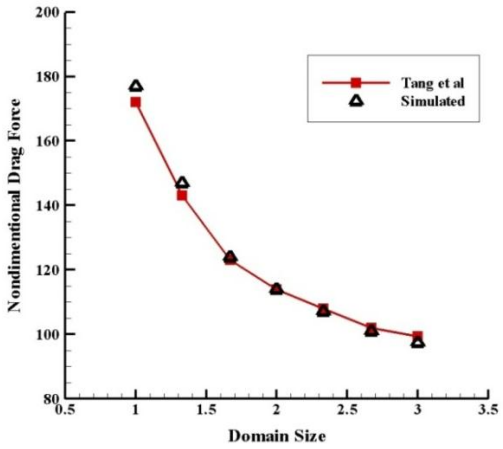

Fig. 5. Comparison of drag forces obtained from present simulation and reference [1]

As seen in Fig. 5, the agreement between present results and those obtained in Ref. 1is excellent. Error between drag forces obtained from present study and Ref.1 are given in Table 4. In the worst case an error of less than 4% exists.

Table 4
Comparison of present work and reference [1]

Domain Size	Tang and Advani	Present Work	Percentage Difference
1	172.37	176.57	2.44%
1.33	142.76	147.05	3.21%
1.67	123.03	124.26	1.00%
2	113.65	114.15	0.43%
2.33	107.73	107.19	0.50%
2.67	102.3	100.91	1.36%
3	99.34	97.36	2.10%

3.2. Momentum Balance Approach

In continuum flow, drag force on a cylinder can be determined using the difference between velocity profiles across the wake behind the cylinder and that of the upstream. From the conservation of linear momentum one can write

$$D = \int \rho b u (U_0 - u)\, dz \qquad (4)$$

Where ρ is the density of fluid, b is the channel width, U is the upstream velocity profile and u is the velocity profile across the wake behind the cylinder. The integral in Eq.(4) is the decrement in momentum flow that occurs across the cylinder. Implementation of Eq. 4 in MD simulation results in the following formulation

$$D_{mean}^{mb} = \sum_{k=1}^{(nbin)_z} \rho_k L_x \overline{u_k} (U_0 - \overline{u_k}) \Delta z_k \qquad (5)$$

Where $D_{mean}^{mb}$is the mean drag force by momentum balance approach, k indicates bin number in z direction, $(nbin)_z$ indicates number of bins in z direction, U_o is the value of velocity in the upstream uniform flow, Δz_k is the height of bin k, and $\overline{u_k}$ is the y component of mean velocity in bin k calculated by Sample-Averaged Measurement (SAM) [11].

From the results obtained, velocity profile is measured at three different sections along the channel, i.e. at the inlet where y=0.10L_y, exactly behind the nanotube where y=0.45L_y, and near the outlet where y=0.80L_y. At each section 25 bins are located in z direction with the size of 2×2×6.26 in z, y, and x directions respectively, to measure velocity profile [12]. As shown in Fig. 6, velocity profile at the inlet where y=0.1L_y, is approximately uniform with a value of U_o=1.0. Exactly behind the cylinder where y=0.45L_y, velocity decreases to zero in the middle of channel and increases to U_o=1.2 near the boundaries. Near the outlet where y=0.8L_y velocity profile shows a trend of gradual return to a uniform profile at the outlet.

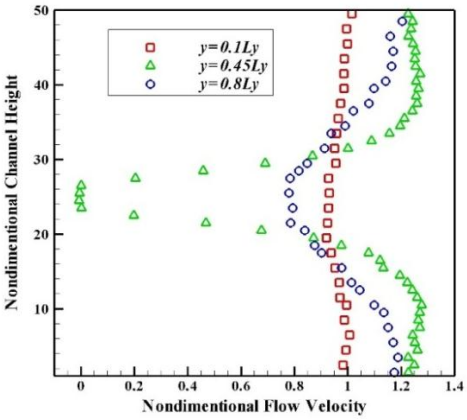

Fig. 6. Velocity profile at $0.1L_y$,$0.45L_y$ and $0.8L_y$

However due to the researches found in the literature, this method is not accurate in nanoscale flows. The difference between the results of two methodologies is reported to be about 30% [7]. In this work two modifications were used to reduce the difference. In previous researches the density ρ in equation (5) was assumed to be constant. But we calculate the average density for each bin and the drag force is calculated with the assumption of variable density. Another parameter which can affect drag force calculated is the number of bins used for determining velocity profile, so three different bin sizes are used to investigate this effect. Results of these two modifications can be seen in Table 5.

Table 5
Effect of modifications on continuum approach

Density	Continuum approach	Direct Method	Difference	Nbin
Constant	85.60	113.65	24.7%	*25*
Variable	88.39	113.65	22.2%	
Constant	87.20	113.65	23.3%	*50*
Variable	90.94	113.65	20.0%	
Constant	87.29	113.65	23.2%	*150*
Variable	91.14	113.65	19.8%	

The increase in the number of bins from 25 to 50 causes the decrease in difference from 22.2% to 20.0% with direct method. But changing the number of bins to 150 does not have a significant effect on the difference. It can be concluded that increasing the number of bins more than this does not decrease the difference necessarily. Although these two modifications on the continuum approach decrease the difference with direct method but the method underestimates the drag by 20% at the best. Hence the method of momentum balance with continuum approach needs a fundamental revision to become an appropriate method for calculating forces in molecular dynamics simulations, which will be discussed in next section.

In this paragraph the effect of different number of bins on velocity profile is discussed. Velocity profile behind the cylinder is evaluated with three different numbers of bins of 25, 50 and 150 corresponding to bin sizes of 2σ, σ and σ/3, respectively. The results are shown in Fig. 7. It shows that the increase in number of bins does not cause a significant change in profile shape but with more precision it is observable that increase in number of bins causes some fluctuations in velocity profile.

When the size of bins is decreased the number of molecules presence in each bin at each iteration also decreases so the number of data used for averaging velocity also decreases and causes a lack of precision in measurement of velocity in each bin. Because of this reason increasing number of bins from 50 to 150 and more than 150 in this case does not increase the precision of the method.

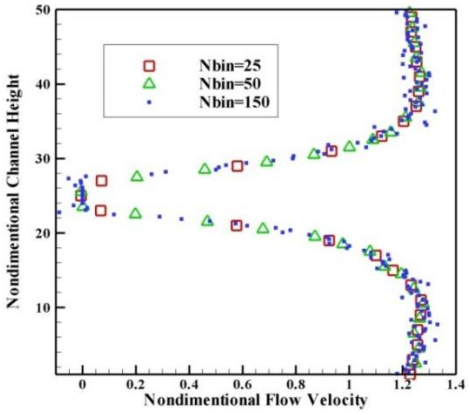

Fig. 7. Velocity profile at $0.45L_y$ using 25, 50 and 150 number of bins

3.3. Molecular Momentum Balance Method

Inspired from the concept of momentum balance in continuum flow, a new method called molecular momentum balance (MMB) is proposed here. As mentioned before uniform flow is maintained by updating flow velocity in the inlet rescaling region every $(\Delta t)_r$ that is equal to 50 solution time steps. In fact by updating velocity to its initial value, we are compensating for the loss of momentum caused by the nanotube in the flow. Based on the second law of Newton this momentum loss during $(\Delta t)_r$ will be equal to the force exerted by nanotube on fluid atoms. Consequently the drag force will be equal to this force but exerted by fluid on the nanotube. Based on this argument one can write

$$\boldsymbol{F} = \sum_{i=1}^{n} m_i \frac{(\Delta \boldsymbol{V})_i}{(\Delta t)_r} \qquad (6)$$

Where $\boldsymbol{F}$ is the resultant force vector, i indicates the molecule number in the inlet rescaling region, n is the total number of molecules in the inlet rescaling region, m indicates the molecular mass (of argon here), $\Delta \boldsymbol{V}$ is the change of velocity vector of atom during each $(\Delta t)_r$ with respect to the rescaling velocity U_o, and $(\Delta t)_r$ is the rescaling step that is equal to 50 solution time steps.

Equation 6 can be applied in three directions after each 50 time steps to calculate the exerted forces. Since the mass of atoms m_i and $(\Delta t)_r$ are constant, the instantaneous drag force can be estimated after each rescaling step from the following formulation

$$D_{inst}^{MMB} = \frac{m}{(\Delta t)_r} \sum_{i=1}^{n} (\Delta V_y)_i \qquad (7)$$

Where D_{inst}^{MMB} is the instantaneous drag force calculated by molecular momentum balance approach. To calculate the mean drag force, SAM is applied here. Therefore

$$D_{mean}^{MMB}(t) = \frac{1}{N_r} \sum_{j=1}^{N_r} (D_{inst}^{MMB})_j \qquad (8)$$

Where N_r is the number of rescaling steps from t_{eq} to t_f. Note that $t_{eq}$of 200 was chosen in all simulations.

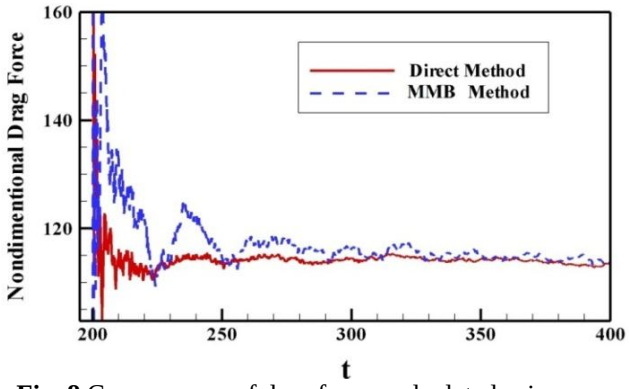

Fig. 8. Convergence of drag forces calculated using MMB method and direct method on the base domain

Drag force is calculated using MMB method and direct method on the base domain. Convergence history of drag force is shown in Fig. 8 for nondimensional time of 200 to 400. In Table 6, drag force calculated by two methods are compared with each other for different values of t_f. As it can be seen error of the MMB method quickly decreases as $t_f$and consequently N_r increases. Finally, at t_f=400 drag of two methods approach 113.5 with an error of 0.2%. Therefore, it can be concluded that MMB method can be used with confidence for the calculation of forces in MD simulations, and will result in accurate data.

Table 6
Comparison of MMB and direct method

t	$N_r(t)$	MMB	Direct	Difference
205	100	153.95	119.91	28.40%
210	200	133.05	114.66	16.00%
220	400	121.65	112.6	8.00%
250	1000	114.58	113.71	0.80%
300	2000	116.44	114.52	1.70%
350	3000	115.18	114.3	0.80%
400	4000	113.61	113.44	0.20%

As it can be seen in Fig. 9, by increasing time from 200 to 400 the error reduces from 40% to less than 2%. These results confirm the fact that this method can be used in molecular dynamics simulations for evaluating forces on immersed bodies. As in direct method of force calculation, increasing number of iterations used for data averaging reduces the difference with final value of forces. As mentioned before in MMB method, the instantaneous drag forces are estimated every 50 time steps. Hence for t<250 number of instantaneous forces used to estimate mean drag is less than 500. This insufficient number of data used for averaging, causes an error of 10%. For t=250, number of forces used for averaging is increased to 1000 and the error is reduced to 1%.

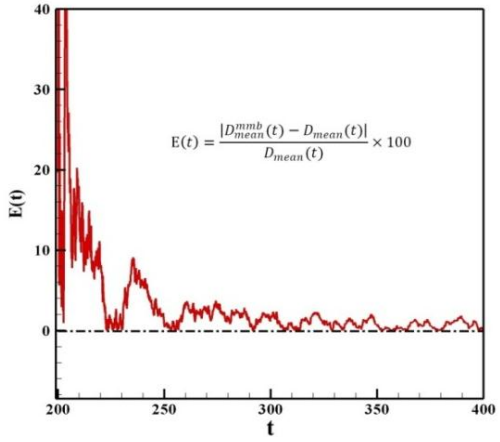

Fig. 9. Relative error of MMB method

The new method can be used for estimating flow forces exerted on nonmolecular bodies such as skin friction on bouncing walls. The computational time does not change significantly with respect to direct method when using MMB method.

4. CONCLUSION

Liquid argon flow over a stationary carbon nanotube was simulated using molecular dynamics simulation. The flow was driven by rescaling particle velocities at the inlet. Drag force on the nanotube calculated using two more common methods including direct summation of forces and continuum approach of momentum balance and a new indirect method is presented. The results of continuum approach show that the increase in number of bins used for velocity measurement will decrease the difference about 5%. Nevertheless the continuum approach at the best underestimate the drag force by about 20% of direct summation and confirm the fact that continuum approach and methods of classical fluid mechanics are not necessarily appropriate for nanoscale flows. But the new presented method works properly and the difference between indirect and direct method can be reduced from 30% to less than 1% by increasing the number of time steps used for data averaging.

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