

# A Molecular Dynamics Study of Energy and Momentum Accommodation Coefficients in the Transitional Knudsen Numbers

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## ABSTRACT

The principal aim of this work is to study the thermophysical aspects of gas interaction on the walls of nano-channels using a three dimensional Molecular Dynamics (MD) model. The complete physics of gas-surface interaction in micro/nano channels in the non continuum regime is yet to be understood. The important parameters that are relevant to such interactions include energy and momentum accommodation coefficients. The study reported in this paper aims at implementing a relatively simple methodology for calculating different accommodation coefficients. Energy accommodation coefficient (EAC) and normal momentum accommodation coefficient (NMAC) are the main focus of the study. The accommodation coefficients for the different equilibrium states are calculated. Instead of tracking individual collision, a collection of collisions are considered for the calculations. The EAC and NMAC reported here are found to be influenced by gas-wall temperature difference and Knudsen number in the transition regime.

## 1. INTRODUCTION

Accurate prediction of the rarefied flows in small devices, such as micro-pumps, micro sensors, MEMS/NEMS, etc., are essential for the development and optimised designs of these systems. This has opened up new challenges in the domain of experimental and computational techniques involving gas flows. Since experimental measurements of gas-surface interaction parameters of rarefied flows [1, 2, 3] are difficult to be achieved in nano sized domains, simulation techniques has emerged as an important tool in the analysis.

The momentum and energy transfer between the gas and the surface are central to all aspects of gas-surface interactions. The main goal of this work is to analyse the gas-surface interaction in a nano channel and study the interaction parameters, such as the energy accommodation coefficient (EAC) and normal momentum accommodation coefficient (NMAC). Molecular dynamics (MD) method [4] is considered as an accurate numerical method to analyse gas surface interaction at micro/nano scale. In this regard, the platinum-argon system is considered and allowed the system to evolve to equilibrium for a specified set of initial conditions. The different states of the system, on its way towards equilibrium, are reproduced by controlling both the wall and the gas temperature. The accommodation coefficients for the different states are calculated by tracking collisions from collision tracking bin.

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## 2. ACCOMMODATION COEFFICIENTS

The accommodation coefficient is a measure to quantify interaction between a wall and a fluid. It indicates the degree to which molecules are accommodated to the surface. The accommodation coefficients can be defined as

$$\alpha_E = (E_i - E_r) / (E_i - E_w) \quad (1)$$

where  $E$  denotes energy and  $i, r$  are for the incident, reflected conditions respectively. The quantities with  $_w$  are determined from thermal wall distributions which are constants for a given wall temperature [5]. The accommodation coefficient based on normal component of velocity is as given as

$$\alpha_n = (P_{ni} - P_{nr}) / (P_i - P_w) \quad (2)$$

where  $n$  denotes the direction normal to the surface. The absolute values of normal momentum are considered for the calculation of normal momentum accommodation coefficient (NMAC) [6].

## 3. DEGREE OF RAREFACTION

The factor which characterises the degree of rarefaction of gases is Knudsen number ( $Kn$ ), which is defined as the ratio of the mean free path of the molecules to the characteristic dimension of the system. The kinetic theory equation for Knudsen number for hard sphere model has been used to calculate  $Kn$ . Even though continuous potential fields are present, the kinetic theory expression is useful to correlate the number density and the characteristic dimension of the system. In the system which we have simulated, the value of  $Kn$  calculated is found to be in the range 0.15-0.34, thereby placing the system in the transitional regime [7].

## 4. MOLECULAR DYNAMICS MODEL

The molecular dynamics method is a deterministic and efficient numerical method to analyse the dynamics of gas surface interaction in a rarefied medium. The real challenge in the MD simulation of gas-surface interaction is to model the wall and accurately define the wall-gas interaction.

### 4.1. Wall models

There have been many attempts to model walls and to extract the gas surface interaction properties. The simplest ones among these are the reflective and the thermal wall models which were originally proposed by Maxwell. He also proposed Maxwell's model which is a linear combination of reflective and thermal wall models. Many authors have used thermal wall models and its variants in their molecular simulation study [8,9,10,11]. The advent of the advanced computational resources facilitated the use of other models also [12, 13, 14, 15]. In this study, the explicit wall model is used to explore the dynamics of gas-surface interaction.

### 4.2. Details of simulation

We opt for a three dimensional system in which monatomic gas flows between smooth and parallel surfaces. One of such systems, the most studied due to its simplicity and availability of experimental data, is the flow of argon between infinite parallel platinum walls. Platinum surface is assumed to be smooth and clean and also without any defects. For the system studied in the present case, the characteristic dimension is in the nanometre range thereby placing the rarefaction levels of the flow in the transition regime.

The dimensions of the simulation box are chosen to be  $10 \times 12 \times 10$  nm and the characteristic dimension is identified as 10 nm. The smallest dimension of the simulation box is kept greater than or equal to the characteristic dimension of the system. Total number of atoms in the simulation is around 20000, out of which approximately 19000 are wall atoms. Initially, the atoms are arranged in FCC structure for platinum and in random configuration for argon.

As the primary objective was to study gas-surface interaction, both the wall and the gas are modelled using Lennard-Jones (LJ) potential [4], which is frequently used to represent the interactions. The data reported in the literature from recent studies are used in this simulation. The mass of argon is  $6.63 \times 10^{-26}$  kg and that of platinum is  $3.23 \times 10^{-25}$  kg. The characteristic length parameters for LJ potential are chosen to be 0.340 nm and 0.247 nm for argon and platinum respectively [13]. The interaction strength for the model is taken as  $5.20 \times 10^{-20}$  J for Pt-Pt interaction and  $1.65 \times 10^{-21}$  J for Ar-Ar interaction [16]. The characteristic LJ length belongs to cross type non-bonded interaction and is computed by Lorentz-Berthelot mixing rules [17].

To control the heat produced in the walls due to collisions and to maintain the wall temperature, thermostats ( $T_w$ ) are applied on both walls as shown in Fig. 1. Temperature of the wall is maintained by controlling translational degrees of freedom of the wall atoms [18]. To eliminate any unbalanced force due to computational error, the velocity of centre of mass is made zero without altering the relative positions of the atoms. Parallel algorithms for classical molecular dynamics are used for implementing MD simulations [19]. Visualisation of trajectories has been implemented using VMD [20].

### 4.3. Tracking of collisions

Collisions are tracked by defining a bin near the wall. When the atoms are in the bin, collisions are recorded as done in similar investigations for tracking collisions in a plane [13, 15, 21]. The location of the bin is determined by considering the influence of the wall upon a gas atom and the mean free path of the gas atoms. In the present study, the gas is so rarefied that the possibility of collisions with another gas molecule near the wall is rare. The location of the collision tracking bin is decided to be at wall-gas interaction cut off distance ( $r_c$ ) which is equal to 2.5 times the Lennard-Jones length parameter.

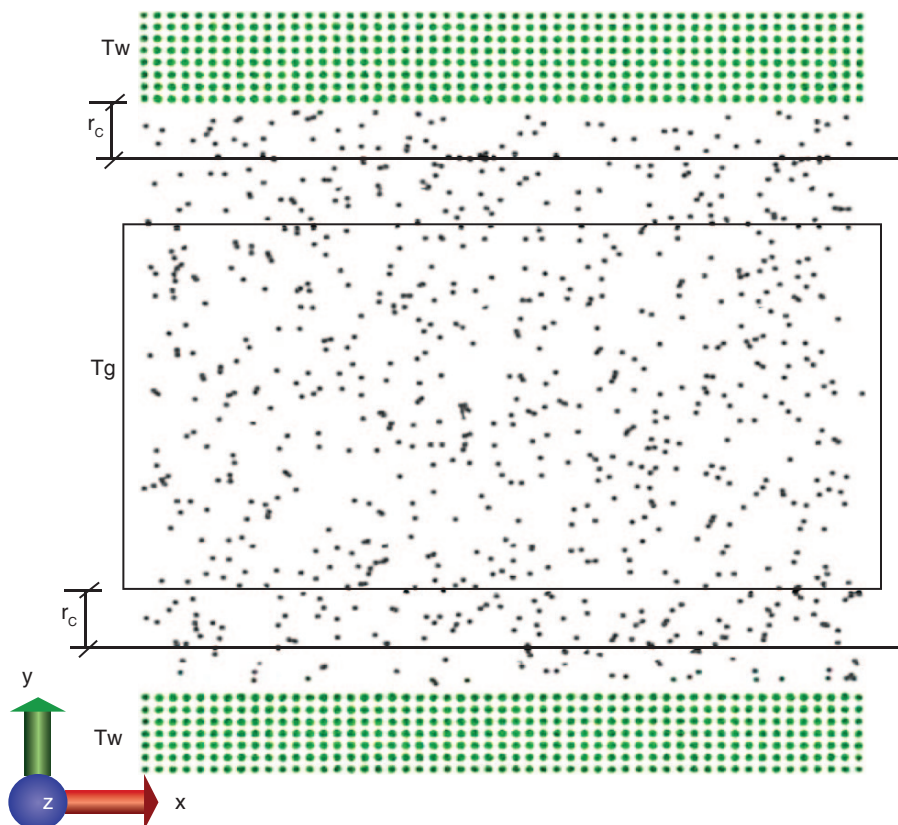


Figure 1. The orthographic representation of simulation model. Location of collision bin, wall thermostat ( $T_w$ ) and gas thermostat ( $T_g$ ) regions are highlighted.  $r_c$  is also shown

## 5. RESULTS

Non Equilibrium Molecular Dynamics (NEMD) is widely used to study accommodation coefficients. The orthographic view of single particle trajectories in the  $y$ -direction for both the solid and the fluid phases are shown in Figure 2. The positions of randomly selected solid atom and a gas atom are plotted with dots for each 100 time steps for a duration of half a million time steps. Initially the gas and the wall are brought to a temperature of 800 K and 300 K respectively.

Then the system is allowed to evolve to an equilibrium state in sufficiently small time step of 1fs. The temperature of the gas is found to decrease drastically in the initial phase and the subsequent decrease is gradual. Finally the gas temperature is found to oscillate about a mean value that is close to the wall temperature.

The procedure is repeated for various Knudsen numbers from 0.1 to 0.8. The time responses of gas temperature are found to follow almost similar paths when the gas-wall temperature difference is appreciably high. Notable differences are observed among various paths when the temperature difference tends to zero. Figure 3 clearly shows that there is no drastic variation in the response of the system as the order of magnitude of the  $Kn$  remains the same.

### 5.1. Variation of gas temperature with wall temperature

Figure 4 shows the variation of wall temperature with respect to the variation in gas temperature for  $Kn = 0.24$ . Similar trends are observed for various other Knudsen numbers under investigation. The curve shows a linear variation. The different gas-wall temperature combinations are selected from the curve, which represents the dynamic states of the above system while reaching towards equilibrium. This would help to model the different states of the system.

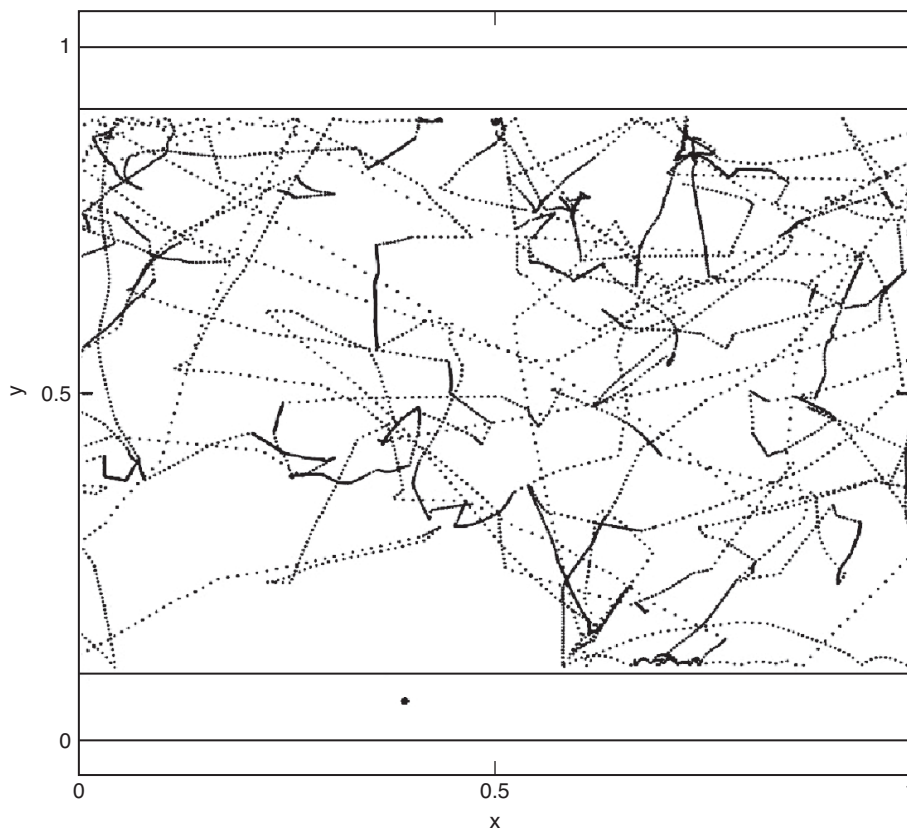


Figure 2. Single particle trajectories for both the solid and the fluid phases

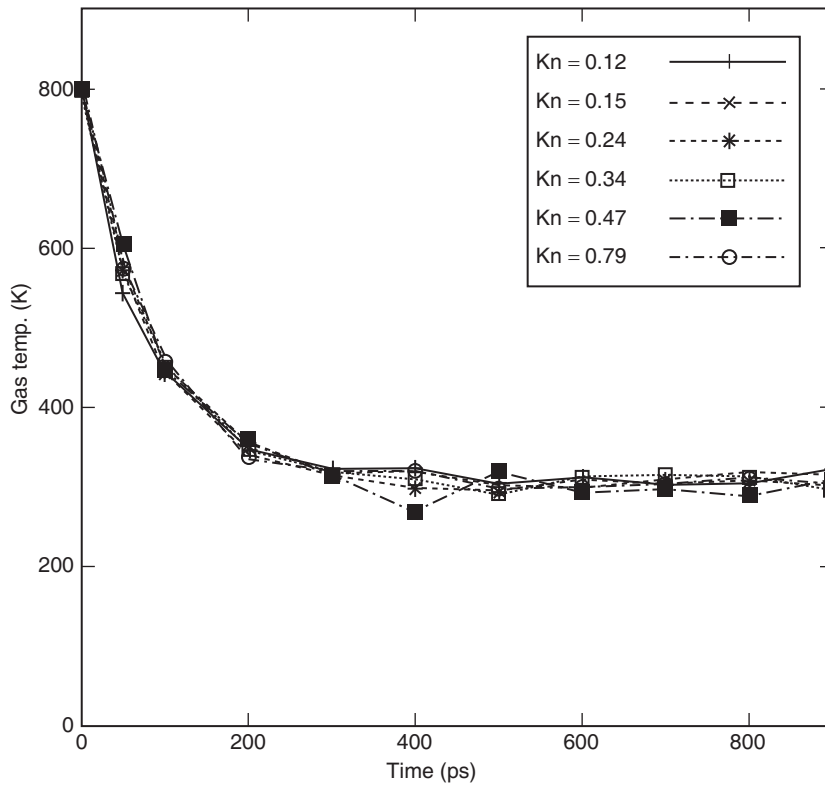


Figure 3. Time response of gas for different Kn numbers

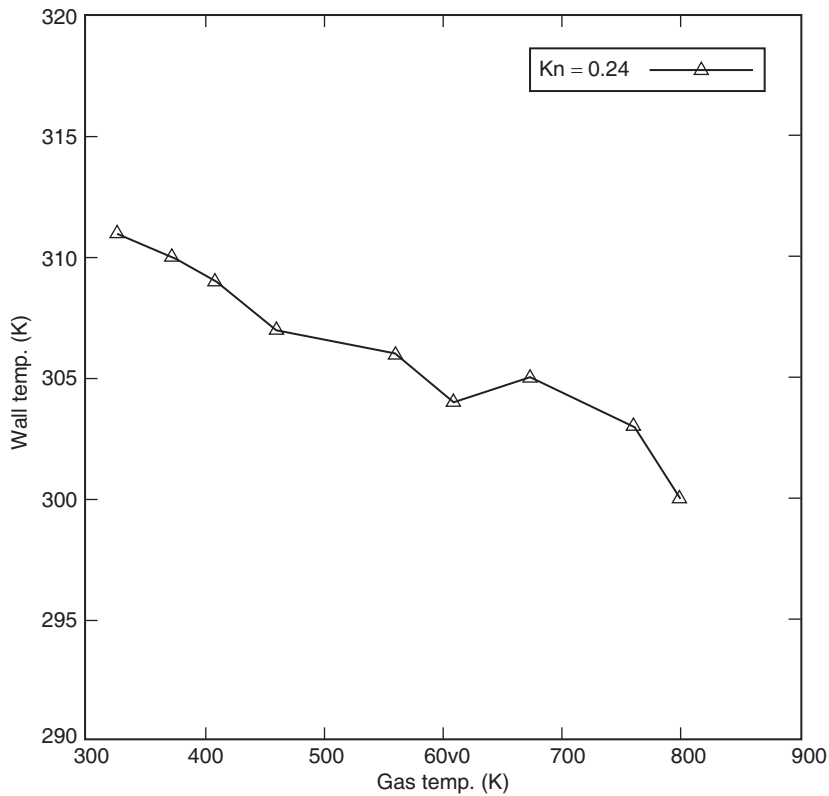


Figure 4. The variation of gas temperature with respect to wall temperature while reaching equilibrium for Kn = 0.24

## 5.2. Calculation of EAC and NMAC

We believe that the random selection of gas-wall temperatures to the system will shadow the actual properties of the system and also it may not appeal to a practical situation of importance. According to Eqs. 1 & 2, EAC and NMAC can be calculated for known quantities of incident and reflected energies of the gas atoms and wall temperature.

The different states of the system are modelled by controlling gas and wall temperature. When we set different wall temperatures for calculating the accommodation coefficients the temperature of the gas is found to be oscillating, as the flow is in a non equilibrium rarefied state. In the present study, we control the gas temperature by applying thermostat to the middle portion of the gas ( $T_g$ ) as depicted in Fig. 1. The thermostat for gas is kept well away from the collision tracking bin so as not to influence the energy accommodation mechanism.

Accuracy of the results from MD, largely depends on the accuracy of the physical model and the number of atoms used in the simulation. As the number of atoms increases MD yield better results, although it increases the round off error. In this context, larger number of collisions would give a better estimate of accommodation coefficients which in turn depends on the size of the system and total duration of simulation. In this study, we choose a system with large number of atoms and longer duration within the available resources. Since it is hard to track individual collisions for a larger system, a cluster of collisions are considered for the calculations.

The incident and reflected atoms are tracked from the bin and energies of each kind are recorded. Incident and reflected atoms are divided into equal number of classes and the class averages are used to calculate the EAC and NMAC. To account for the statistical noise due the longer stay of slowly moving atoms, the collisions are tracked at definite time intervals. The average normal component of velocity will be different for different cases depending on the gas temperature. The collision data are collected from the bin based on the average stay of atoms in the bin. (Another approach is to rescale the quantities to be averaged with respect to the normal component of velocity [5]). However, this type of data collection doesn't account for the sign of collision which is important for the calculation of tangential momentum accommodation coefficient.

The procedure is repeated for different Kn numbers to study its dependence on Kn. Values of EACs and NMACs thus calculated are tabulated in Table 1. The standard deviation of accommodation coefficient is given in brackets. The overall value of standard deviation is low, except for some odd cases, indicating the consistency of calculation.

Table 1. Calculated values of EAC and NMAC

<b>Kn = 0.15</b>		<b>Kn = 0.24</b>		<b>Kn = 0.34</b>	
<b>Energy Accommodation Coefficient</b>					
<b>Temp. Diff.</b>	<b>EAC</b>	<b>Temp. Diff.</b>	<b>EAC</b>	<b>Temp. Diff.</b>	<b>EAC</b>
59	0.211 (0.019)	59	0.232 (0.032)	60	0.238 (0.032)
145	0.190 (0.011)	147	0.218 (0.010)	149	0.234 (0.015)
242	0.191 (0.006)	244	0.207 (0.022)	247	0.220 (0.014)
291	0.188 (0.005)	293	0.204 (0.005)	296	0.207 (0.012)
435	0.184 (0.005)	440	0.202 (0.005)	447	0.206 (0.011)
<b>Normal Momentum Accommodation Coefficient</b>					
59	0.218 (0.024)	59	0.230 (0.028)	60	0.244 (0.033)
145	0.209 (0.011)	147	0.235 (0.011)	149	0.249 (0.015)
242	0.203 (0.006)	244	0.233 (0.008)	247	0.240 (0.013)
291	0.196 (0.007)	293	0.236 (0.007)	296	0.245 (0.011)
435	0.189 (0.004)	440	0.220 (0.005)	447	0.239 (0.008)



### 5.3. Variation of EAC and NMAC with Kn and temperature difference

Variation of EAC and NMAC with respect to Kn is shown in Figures 5 and 6. It is interesting to note that the value of EAC decreases as the temperature difference between wall and gas increase. When the temperature difference between wall and gas decreases, the standard deviation is found to increase. This could be due to the low energy of the atoms and subsequent longer stay in the collision tracking bin. The reduction in EAC is not consistent with the temperature difference, i.e., for the same temperature difference it does not yield the same difference in EAC. This shows the dependency of accommodation coefficient on the absolute values of wall temperature and gas temperature. At higher temperatures, the energy of the molecules is high and hence the velocities. This would affect the relaxation time and consequently the accommodation coefficients. It can also be observed that, on an average, EACs are less than the accommodation coefficient based on the normal component of velocity. To some extent, NMAC also follows a similar trend as EAC as shown in Figure 6. A similar trend was reported by Spijker et al. albeit the conditions were, to an extent, different from the present study [13].

Figures 5 and 6 also show the variation of EAC and NMAC with Kn. Both EAC and NMAC increase as the Kn increases. As the Kn increases the gas-gas interaction decreases and probability of gas molecule finding the wall increases. Furthermore, when Kn increases the influence of gas atoms upon another gas atoms decreases which affects the collisions. A direct comparison of these results with available experimental and simulation works could not be made possible as the simulation conditions and methodology used in the present study are different.

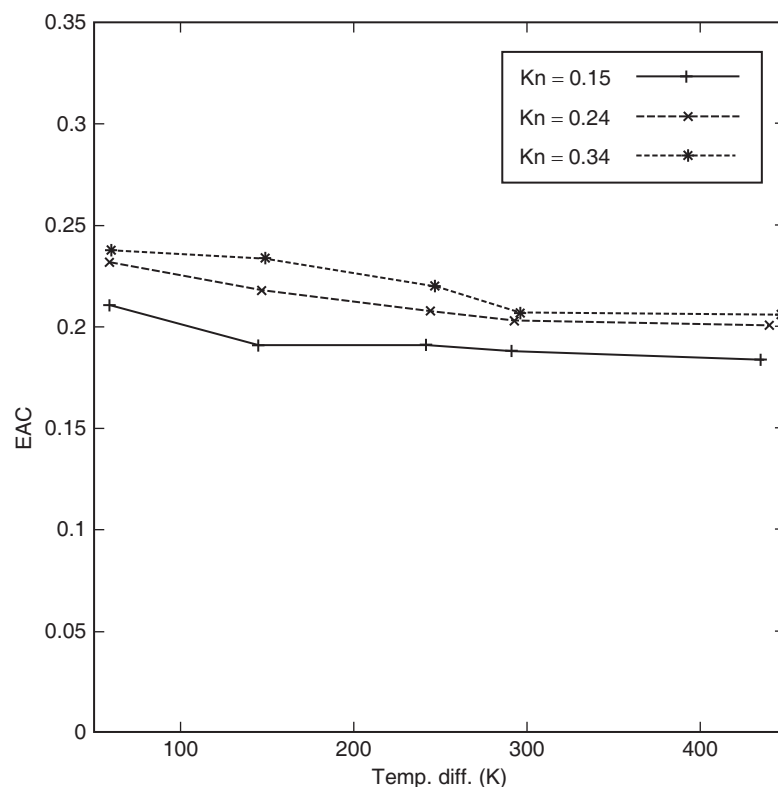


Figure 5. The variation of EAC with Kn

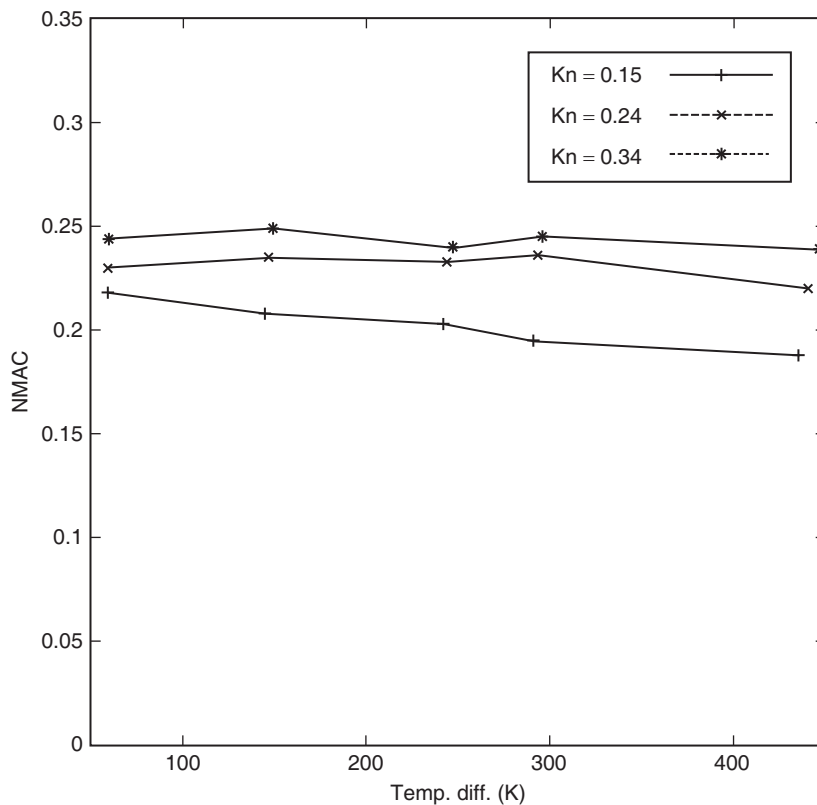


Figure 6. The variation of NMAC with Kn

## 6. CONCLUSIONS

A molecular dynamics study of energy and normal momentum accommodation coefficient in the transition regime is conducted. A general methodology has been presented to calculate EAC and NMAC to very good degree of precision. NEMD simulations of rarefied argon between smooth platinum plates were used to investigate the gas surface interaction. The platinum-argon system is allowed to evolve to equilibrium states and accommodation coefficients for the different equilibrium states are calculated. The methodology presented in this work could be useful for systems containing large number of atoms and collisions. This method also proposes a comparatively simpler algorithm for calculating EAC and NMAC without influencing the physics of the system. The system is also studied by varying the Knudsen number, i.e., by varying number of atoms. The major influencing parameters of EAC and NMAC identified are the Knudsen number and the temperature difference between the wall and the fluid.

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