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THE INFLUENCE OF GAS-WALL INTERACTIONS ON THE ACCOMMODATION COEFFICIENTS FOR RAREFIED GASES: A MOLECULAR DYNAMICS STUDY

Shahin Mohammad Nejad*¹, Silvia V. Nede¹, Arjan J.H. Frijns¹

[¹s.mohammad.nejad@tue.nl](mailto:s.mohammad.nejad@tue.nl), [¹S.V.Nede@tue.nl](mailto:S.V.Nede@tue.nl), [¹a.j.h.frijns@tue.nl](mailto:a.j.h.frijns@tue.nl)

Eindhoven University of Technology, Dept. Mechanical Engineering,
PO BOX 513, 5600 MB Eindhoven, the Netherlands

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ABSTRACT

The energy accommodation coefficient (EAC) and the momentum accommodation coefficient (MAC) are two significant parameters determining the gas-solid energy and momentum exchange efficiencies. In this work, molecular dynamics (MD) simulations were employed to study the impact of gas-wall interaction potential on energy and momentum accommodation coefficients between Gold and monoatomic gases (Argon and Helium). The MD simulation setup consists of two infinite parallel plates of unequal temperature positioned at certain distance (12 nm and 102 nm for Argon and Helium gases, respectively) apart from each other, and of gas molecules confined between them. A pairwise Lennard-Jones 12-6 potential was considered at the solid-gas interface. The interaction potential parameters were obtained using the Lorentz-Berthelot (LB) and Fender-Halsey (FH) mixing rules, as well as based on existing ab-initio computations. Comparing the obtained results for the accommodation coefficients with empirical values revealed that the interaction potential based on ab-initio calculations is the most reliable one for computing ACs. Besides, in the case of Au-Ar, the LB mixing rule substantially overpredicts the potential well depth which leads to sticking gas atoms on the solid surface. As a result, computing accommodation coefficients in this case from numerical point of view was not possible.

Introduction

Rarefied gas condition is encountered in numerous modern applications, for example microelectromechanical and nanoelectromechanical systems, semiconductor manufacturing and spacecraft flying over the high altitudes [1]. In such systems, especially adjacent to the solid surface the number of intermolecular collisions is less than the number of collisions between the gas molecules and the wall. Accordingly, the thermo-physical properties of gas (density, thermal conductivity, etc.,) are highly affected by the energy and momentum exchange at the solid-gas interfaces. The energy and momentum accommodation coefficients (AC) are commonly utilized to evaluate the heat and momentum transfer efficiency. Besides the empirical approaches in computing different ACs, numerical methods such as MD simulations considered as a promising tool to determine these parameters. In this work, MD simulation has been carried out in order to study the impact of the interatomic potential on ACs at the solid-gas interface. Having in mind that the solid-gas interatomic potential is one the most salient factors affecting computed accommodation coefficients via MD technique, a

* Corresponding author

pairwise Lennard-Jones (LJ) potential was considered at the solid-gas interface. The LJ potential parameters were computed using different approaches such as the Lorentz-Berthelot (LB) and Fender-Halsey (FH) mixing rules, as well as taking form existing ab-initio calculations [2]. The results were compared with previous experimental values.

Computational methods

The MD simulation setup considered in this work is a three-dimensional system, in which a monatomic gas is confined between two parallel walls (see Fig.1).

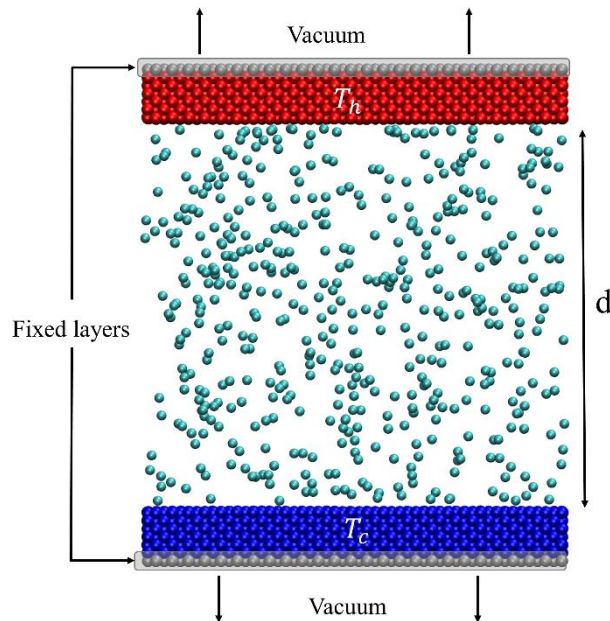


Figure 1: Schematic representation of the simulation setup

The cross section area of the walls is 10 nm by 10 nm, and each of them consists of 8750 atoms arranged in FCC structure. The walls are separated from each other in the y direction. The Knudsen number in the first case is 0.23 and in the latter case is 0.56. Periodic boundary conditions are applied in all three directions. The temperature of the bottom and top walls are maintained at $T_c = 300$ K and $T_h = 350$ K, respectively, using Berendsen thermostats. Besides in each wall, the outermost layer is fixed to prevent the wall from any translational or rotational motion. Since the simulation box is periodic, the walls are not placed directly at the periodic boundary. Vacuum has been considered between the walls at the periodic boundary in order to prevent direct contact and conduction between the hot and cold walls. Herein, the gas molecules are not coupled to an external heat bath, and their temperature change is caused only via collisions with other particles. It should be pointed out that in order to obtain accurate results for ACs, a lot of collisions between gas molecules and the wall surface are required (100,000 collisions have been considered here). Increasing the number of collisions can be achieved by either extending the walls surface area or increasing the gas number density. Considering the computational time, the latter approach is more favorable. The reason lies in the fact that generally the density of atoms in the wall is much higher in comparison to the gas domain. Therefore, more interactions should be modeled within the cut-off radius, which causes more computationally expensive simulation. On the other hand, higher gas number density means higher pressure and the possibility to surpass the gas critical pressure, which is not desirable here, since the properties of a supercritical fluid differs in some aspects from a gas. In this work, in order to increase the gas number density, walls are positioned in the closest distance from each other in such a way that the pressure in the system does not exceed the critical pressure (P_{cr}) value in the system (For Ar $P_{cr} = 4.86$ MPa, and for He $P_{cr} = 0.23$ MPa [3]). Here we chose for separation distance $d = 11$ nm (Ar) and $d = 102$ nm (He), which results in a similar temperature gradient as reported by others e.g. [4]. It has been noticed that further decreasing of the pressure in the system does not



have a significant effect on the obtained ACs. Such a behavior has been also reported in the previous experimental [5] and numerical [6] studies.

The interaction between Gold atoms has been described by the embedded atom model (EAM) potential, whereas the interaction between Ar-Ar and He-He are modeled by the Lennard-Jones (LJ) potential with interaction coefficients ($\sigma_{Ar-Ar} = 3.35 \text{ \AA}$, $\epsilon_{Ar-Ar} = 1.22e^{-2} \text{ eV}$, $\sigma_{He-He} = 2.64 \text{ \AA}$, $\epsilon_{He-He} = 9.4e^{-4} \text{ eV}$) taken from [7]. The Gold-gas interactions were described by LJ potential. For implementing MD simulations, LAMMPS molecular dynamics package was employed [8].

All simulation setups were initially equilibrated for 1 ns (1 fs time step). Afterwards, the production run was started and proceeded for the next 20 and 30 ns for Ar and He cases, respectively.

The accommodation coefficients were calculated by the least-square method proposed by Spijker et al [4]. In this approach, which is based on the correlation between input (impinging) and output (reflection) data obtained from MD simulations, the AC (α) can be computed as follows:

$$\alpha_Q = 1 - \frac{\sum_i(Q_i^i - \langle Q_I \rangle)(Q_R^i - \langle Q_R \rangle)}{\sum_i(Q_i^i - \langle Q_I \rangle)^2}, \quad (1)$$

where Q can be any quantity, such as the gas atom velocity in a certain direction or its total kinetic energy. Q_I and Q_R referred to the considered quantity for the impinging and reflected particles, respectively. The bracket notations denote that the average value for these quantities needs to be calculated.

Results and discussion

The comparison between interaction potentials obtained using different methods is shown in Fig.2. It can be seen that the LB mixing rule always overestimates the potential well depth. In the case of Au-Ar the estimated value by the LB mixing rule is two order of magnitude higher than the value computed by the ab-initio approach. Due to such a big well depth, during MD run all Argon atoms are adsorbed on the solid surfaces (see Fig.3a) and they do not leave them anymore. Therefore, for none of the impinging gas particles an outgoing velocity can be recorded, and this issue makes ACs numerically unobtainable. In addition, the normalized number density distributions in the case of aforementioned system using varied interaction potentials are depicted in Fig.3b. Herein, initially it can be understood that the gas density adjacent to the wall surfaces is higher than the bulk density (n_0) in the central part of the system, and the stronger is the gas-wall interaction the higher is the density profile peak near to the wall. Such a behavior has also been reported in [9]. Furthermore, in the case of the LB potential except in the vicinity of the walls in the other parts of the simulation box the gas density is zero.

In order to compute ACs, the atomic velocity components are recorded before and after bouncing with the wall at the wall-gas interaction cut-off distance which was set at 12 \AA from the wall. Afterwards, a line is fitted to the collision data based on least-square approximation, and the slope of this line can be used to compute the AC (see Eq. (1)). Some velocity clouds are represented in Fig.4, for Au-Ar and Au-He, respectively. The obtained ACs for the aforementioned systems at the bottom wall ($T_w = 300 \text{ K}$) are reported in Tab.1. As we can see for both noble gas cases, the ACs increase with increasing the potential well depth. The reported empirical values for the EAC (α_E) for Au-Ar and Au-He are 0.85 and 0.31, respectively [10]. In the case of Au-Ar, the obtained EAC using interaction potential based on the ab-initio calculations is in excellent agreement with experimental one. Agrawal and Prabhu [11] have reported that the tangential-MAC (TMAC) for Ar on commonly employed surface materials is 0.893. This value is consistent with obtained results for TMAC in different directions (α_x and α_z) in our case. The EAC using FH mixing rule is higher than unity, which is unphysical. In the case of Au-He, the results for EACs using potentials obtained via ab-initio simulations and LB mixing rule are both in good agreement with experimental value. Herein, applying the FH mixing rule leads to an underestimation in the EAC value.

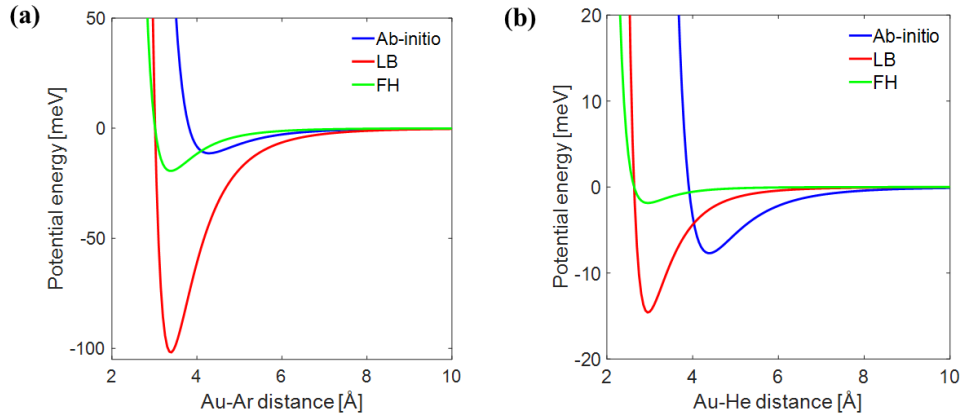


Figure 2: Potential energy plot of noble gases interaction with Gold surface: (a) Au-Ar; (b) Au-He

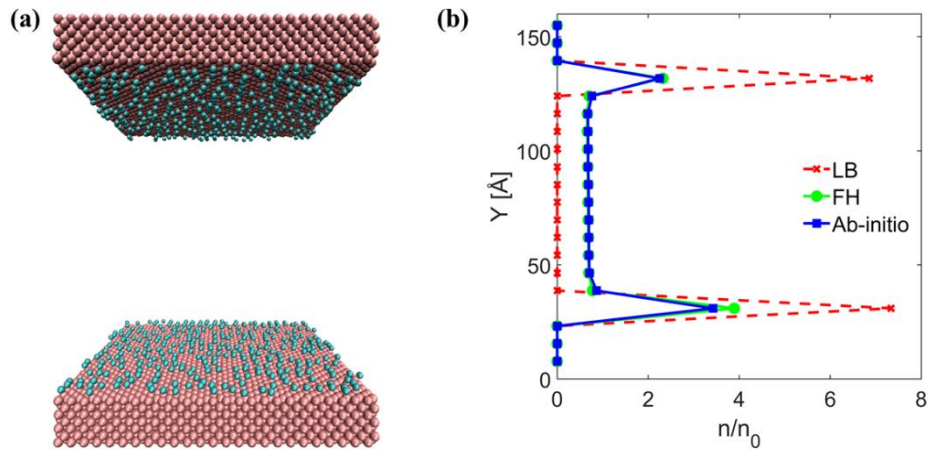


Figure 3: (a) adsorption of Ar gas on Gold surface for the Au-Ar interaction potential based on LB mixing rule; (b) Normalized number density for different Au-Ar interaction potential

System	Pair potential	α_x	α_y	α_z	α_E
Au-Ar	Ab-initio	0.82	0.91	0.83	0.87
	FH	1.00	0.98	1.00	1.2
Au-He	Ab-initio	0.32	0.47	0.32	0.19
	FH	0.2	0.34	0.22	0.07
	LB	0.65	0.74	0.65	0.23

Table 1: MAC and EAC results for Ar and He colliding with Gold surface at T=300 K

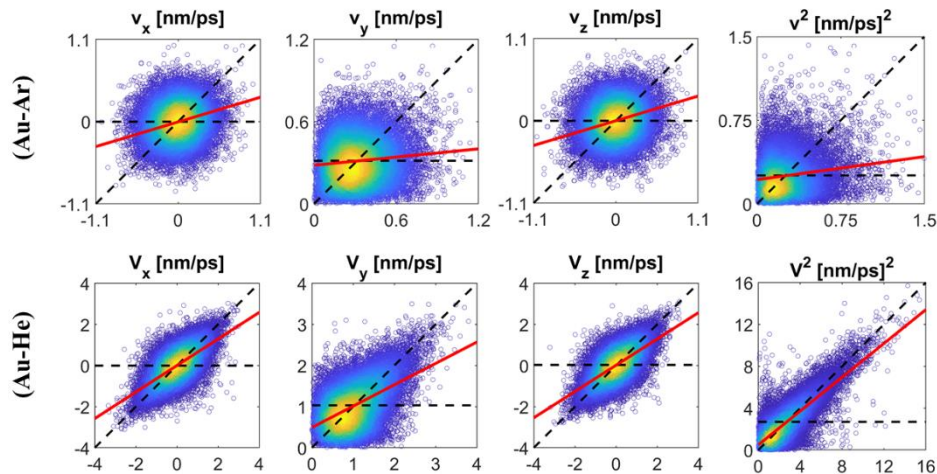


Figure 4: Velocity correlations of impinging (x axis) and reflected (y axis) velocity components of Ar and He for 300 K Gold surface. The dashes horizontal and diagonal lines indicate fully diffusive and specular conditions, respectively. The red line refers to the linear fit of the collision data.

Conclusions

It has been noticed that the accommodation coefficients obtained based on ab-initio potential are always in a good agreement with empirical values. In addition, in the case of Au-Ar the LB mixing rule highly overestimates the potential well depth. This issue causes a fully saturated solid surface in which computing the ACs from numerical point of view is not possible.

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