Non-Equilibrium Molecular Dynamics Study on the Thermal and Mechanical Properties of Graphene

B. Mortazavi a,b, A. Rajabpour c*, S. Ahzi b, A. Hadizadeh kheirkhah d

aCentre de Recherche Public Henri Tudor, Department of Advanced Materials and Structures, 66, rue de Luxembourg BP 144, L-4002 Esch/Alzette, Luxembourg
b Institut de Mécanique des Fluides et des Solides, University of Strasbourg/CNRS, 2 Rue Boussingault, 67000 Strasbourg, France
cDepartment of Mechanical Engineering, Imam Khomeini International University, Qazvin, 34194-288, Iran
dDepartment of Physics, Tarbiat Modares University, Tehran, 14115-175, Iran

*arajabp@ut.ac.ir

Abstract: In this study, the thermal conductivity and mechanical properties of Single-Layer Graphene (SLG) are studied using non-equilibrium molecular dynamics (NEMD) simulations. The carbon atoms bonding interactions are introduced using the optimized Tersoff potential proposed by Lindsay and Brodo (Physical Review B, 2010, 82, p. 209903). We found that by increasing the length, the thermal conductivity of SLG increases. Thus the length independent thermal conductivity of SLG was obtained by extrapolation of the obtained results for the infinite length. In this way, the thermal conductivity of SLG is obtained to be around 3,000 ± 100 W/m-K which is in a fine agreement with the latest experimental results. Furthermore, by performing the uniaxial tension tests we studied the Young’s modulus and Ultimate Tensile Strength (UTS) of SLG. We predicted the Young’s and UTS of SLG to be around 985 and 155 GPa, respectively, which are in considerable agreement with the latest experimental results for free of defect SLG. Agreements with the experimental results verify the validity and accuracy of the molecular dynamics modelling architecture developed in this study.

Keywords: Graphene, Molecular dynamics; Thermal conductivity; Mechanical properties.

Introduction
A two dimensional structure of carbon atoms, graphene is an ongoing research interest in low dimensional materials due to its exceptionally high crystal and electronic quality [1]. As a member of carbon family, graphene materials present extraordinary thermal [2], mechanical [3] and electrical [4] properties. In this way, the graphene is considered as an excellent candidate for building block in nano-electro mechanical systems (NEMS), nanoelectronics and nanocomposites. Thus, the fundamental understanding of graphene properties is the critical issue in its future applications.

Molecular Dynamics (MD) is now widely used for the simulation of matter in the molecular-scale. MD simulation computes the motion of individual molecules in the models of solid, liquid, or gas. Then material properties including both physical and chemicals can be evaluated using statistical mechanics theories.

In this way, today MD could be utilised as an efficient tool in atomic scale science. However, it should be noted that the properties obtained by the MD modelling are strongly dependent to the selection of appropriate interatomic potentials, loading conditions and boundary conditions as well. Thus, the validity of the MD modelling design should be first evaluated by comparison with the available experimental results. In the current study we propose the MD architecture for characterization of thermal and mechanical properties of graphene. The validity of our modelling was verified by observing the fine agreements with the latest published experimental results.

Molecular Dynamics Modelling
All simulations in the current study were performed using LAMMPS [5] package. The bonding interaction between carbon atoms was modeled using the optimized Tersoff potential developed by Lindsay and Brodo [6].

The MD simulation model for evaluation of mechanical properties of SLG with chirality angle of 10º (where the 0º is assumed to be armchair) is illustrated in figure 1a. The periodic boundary conditions were used in x and y directions. Prior to applying the loading condition the MLG structure was left to relax anisotropy to zero stresses in x and y directions at temperature of 300K for 20,000 time increments using Nosé-Hoover method. The loading condition was applied by extending the periodic simulation box size in the x direction by constant strain rates. The Young’s modulus, Y, of the structure can be calculated by:

\[ Y = \frac{1}{At} \frac{\partial^2 E}{\partial^2 e} \]  

(1)

where E and A are the deformation energy and initial surface of the structure, respectively. t is the thickness of SLG which is equal to 3.4 Å and e is the strain.
Thermal conductivity is computed by non-equilibrium molecular dynamics method. In which a heat flux is imposed on the specimen to form a temperature gradient. It reverses the usual cause-and-effect picture with the advantage of fast convergence of the temperature gradient in a non-equilibrium steady state. We predict the effective longitudinal thermal conductivity, $k_x$, of the SLG using a one-dimensional form of the Fourier law:

$$q_x = -k_x A_c \frac{dT}{dx}$$  \hspace{1cm} (2)

Where $q_x$ is the heat flow in the x direction, $dT/dx$ is the temperature gradient and $A_c$ is the cross-section area of the SLG. As shown in Figure 2b, we introduce a heat flow by transferring a known and constant quantity of kinetic energy from carbon atoms in the hot reservoir to those in the cold reservoir at each simulation time increment. We fix three rows of carbon atoms at the two ends of the SLG to prevent atoms from sublimating and periodic boundary conditions were applied in the y direction.

Results and Discussion

In Figure 2, strain energy for armchair SLG is plotted as function of strain for different loading strain rates acquired by optimized and original Tersoff potentials. Assuming low strain levels (up to 0.05) part as the linear elasticity part for the stress-strain response, the Young’s modulus was obtained using equation 1. Based on Figure 2 observations, the Young’s modulus of the armchair SLG are independent of the strain rate values.

We also performed uniaxial tensile tests for other chirality directions and we found that the reported Young’s modulus is acceptably independent of loading chirality direction. Moreover, the obtained Young’s modulus of SLG predicted by the optimized and original Tersoff [7] potentials in armchair direction are 0.985 and 1.05 TPa, respectively, which both are in good agreement with experimental Young’s modulus of 1 TPa for defect-free SLG reported by Lee et al. [3]. While the obtained Young’s modulus is independent of the loading strain rate, the predicted tensile strength which corresponds to the maximum strain energy decreases by decreasing the strain rate. By calculating the Virial stresses, we found that at the strain rates of $1 \times 10^8$ s$^{-1}$ and the UTS of armchair SLG’s acquired by optimized and original Tersoff potentials are around 155 and 235 GPa respectively. Taking into consideration the experimentally obtained UTS of 130 ± 10 GPa for SLG by Lee et al. [3], we conclude that the optimized Tersoff potential predicts the mechanical response of SLG in a better agreement than those predicted by original potential.

In Figure 3, the inverse of thermal conductivity of armchair SLGs as a function of the inverse of the length is illustrated. As it can be observed, by increasing the length, the thermal conductivity of SLG increases. The length dependency of the obtained thermal conductivity of SLG can be eliminated by extrapolation of the obtained results for infinite length. For this, a simple approach is to define the effective phonon mean free path ($\Lambda_{eff}$) as $1/\Lambda_{eff} = 1/\Lambda + 1/L$. Since the thermal conductivity is proportional to $\Lambda_{eff}$, the thermal conductivity of the infinite system can be obtained by extrapolating to $1/L = 0$. In this way, the size independent thermal conductivity of SLG is obtained to be around 3,000 ± 100 W/m-K. This estimate is close, but underestimates the experimental
results [8]. It is worth mentioning that in a similar study in [9], using the second generation REBO potentials the thermal conductivity of SLG was predicted to be around 350 W/m-K. Using the original version of the Tersoff potential, Wei et al. [10] obtained the thermal conductivities of around 850 W/m-K for SLG. This shows that the optimized Tersoff potential is an accurate potential for analyzing thermal conductivity in carbon structures.

Figure 3. Measured and extrapolating curves of inverse of thermal conductivity as function of inverse of single-layer graphene length

Conclusions
In this study, non-equilibrium molecular dynamics modeling were carried out using optimized Tersoff potential developed by Lindsay and Broido [6] to investigate the mechanical and thermal properties of graphene. The summary of the obtained results are as follows:

1- Using the optimized Tersoff potential, the predicted Young’s modulus and tensile strength of graphene are close to experimental results.

2- The Young’s modulus of graphene sheet is acceptably independent of loading chirality direction

3- The optimized Tersoff potential predicts thermal conductivity of graphene in a much better agreement with the experiments than the predictions based on the original Tersoff and REBO potentials

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