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Tuning thermal conductivity of porous graphene by pore topology engineering: Comparison of non-equilibrium molecular dynamics and finite element study

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ABSTRACT

Tuning thermal conductivity of porous graphenes has attracted much interest in the thermal management of nanoelectronics devices due to the promising multifunctional properties of engineered nanomaterials. To explore the potential of tuning thermal properties of monolayer porous graphenes in multiple scales, non-equilibrium molecular dynamics (NEMD) and finite element method (FEM) are implemented to manipulate their thermal conductivity and temperature distribution by the engineering of pore topology. Results indicate that the thermal conductivity of porous graphenes can be significantly lower than a pristine graphene. The thermal conductivity reduction is attributed to phonon scattering at the boundaries of defects described by the phonon density of states analysis. It is found that the thermal conductivity and the temperature distribution of a porous graphene can be desirably tuned by the simultaneous engineering of relative density, pore topology, and pore orientation. Then, the effect of unit cell periodicity on the thermal conductivity of periodic porous graphenes, called phononic graphene or graphene metamaterial, is explored. Finally, comparing the results of continuum mechanics approach through the implementation of FEM and NEMD simulation presents the advantages of NEMD for predicting the thermal conductivity of engineered porous graphenes with characteristic length of lower than 50 nm.

1. Introduction

Tightly packed honeycomb arrangement of carbon atoms with covalent sp^2 bonds [1] generates a highly-strength structure, named as graphene, with outstanding thermophysical properties. Graphene [2] as a one-atom-thick planar sheet possesses high thermal conductivity [3,4]. Because of the distinct properties of graphene, e.g. high thermal/electrical conductivity and high strength and stiffness, they have found many potential engineering applications in nano-sensors [5,6], nano-actuators [5,7], memory devices [8], graphene-based gas/bio-sensors, ion batteries [9–11], electrical double layer capacitors (EDLCs) [10], solar cells [10,12], energy storage and production [10,13–15], water purification [16], and additive or stand-alone lubrications [17,18]. In addition to the abovementioned applications, graphene is also known as a top potential nano-filler for improving multifunctional properties of nanocomposites, energy harvesters, and 3D printed advanced materials [19–22] with distinguished mechanical stiffness and thermal/electrical conductivity [5,8,10,23–26] and thermal expansion [27].

Experimental studies report the thermal conductivity of graphene as high as 4800–5300 W/mK near room temperature (RT) for suspended graphenes [28–34] grown on metallic scaffolds in vacuum or air and without the effect of any underlying substrate. For supported graphenes, i.e. graphene sheets synthesized on substrates, thermal conductivity decreases due to the interface scattering effects [35] between the graphene sheet and the substrate. Due to the extremely high thermal conductivity of graphene, tuning its thermal properties is of great importance for electronic

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industry in order to enhance the efficiency of advanced integrated electrical circuits through controlling their operating temperature. In order to tune thermal properties, many researchers have employed surface modifications through doping adatoms [36], generating vacancies [37–39] and holey nanostructures [40,41], isotope substitutions [42], hydrogenation [43] and hybridization [44], oxidization [45,46], existence of strain [47] and grain boundaries [19,48,49], bending of graphene sheets [36], and graphene laminates [50].

Porosity (P) or void volume fraction is defined by the existence of pores/holes/voids in the nano/microstructure caused by multiple vacancies in the atomic structures. Voids or defects generally appear during casting in the graphene fabrication/preparation process [16,51]. Defective nanostructures usually have single/double/mono cluster vacancies [52] or have other types of defects like Stone-Wales or adatoms [53]. In order to quantify the value of porosity, a few parameters like relative density or porosity are used. Relative density represents the ratio of the number of defective atoms/bonds in the atomic structure to the whole number of atoms/bonds in its pristine periodic unit-cell. In opposite, the porosity indicates the ratio of void space/volume in the porous unit-cell to the whole space/volume of the pristine form. Recently, it has been reported that manipulating the nanostructure or nanoarchitecture of defected graphenes could tune their chemical/electronic [54–61] and thermal [42,62–71] properties. However, there is still lack of comprehensive research on the effect of defect/pore topology, i.e. size and orientation of porosity, on the thermal properties of engineered porous graphenes. The review paper of Benhart et al. [40] presented a comprehensive overview of different types of possible defects in graphenes. It has been found that mono vacancy defects [52] have the most significant influence on the thermal conductivity reduction of graphenes. Results show that the defects breaking in-plane covalent sp^2 bonds are more effective in decreasing the thermal conductivity of graphene compared to the other defect types. The effect of single- and double-vacancy defects and Stone-Wales defect on the thermal conductivity of graphenes using NEMD has also been studied [72]. It has been reported that the thermal conductivity of graphene is less sensitive to temperature with increasing the defect concentration. Investigating the effect of single and double vacancies [73] made by the low-energy electron beam irradiation (20 keV) on the thermal conductivity of suspended graphenes has also been conducted; reduction of thermal conductivity as a function of defect concentration using the Boltzmann transport equation and molecular dynamics (MD) simulation has been reported.

Rajasekarsn et al. [74] studied the effect of porosity, including point and line defects, on the mechanical and thermal properties of graphenes. Thermal conductivity of graphene nanomeshes [71] utilizing equilibrium MD (EMD) simulation has recently been investigated; it has been found that the thermal conductivity of nanomeshes increases by enhancing the periodicity for constant porosities. The reason has also been addressed to, as it is called, "necking effect". It suggests that when the distance of adjacent pores is smaller than phonon mean free path, a phonon can be "trapped" behind the pore, which creates a local negative temperature gradient opposing the linear temperature gradient along a free channel [75]. The effects of porosity on reducing the thermal conductivity of phononic graphenes [76] have also been studied atomistically. It has been found that the thermal conductivity of a phononic graphene is significantly lower than that of pristine graphene which can be desirably tuned by changing the porosity and periodic length. An experimental study [51] has been carried out to observe the significant reduction of thermal conductivity and consequently improved thermoelectric properties of single- and bi-layer engineered porous graphene due to the confined geometry of nanomeshes.

Despite of many publications reporting the influence of porosity on the mechanical properties of graphenes, there is a lack of knowledge on the impact of defect engineering on tuning the thermal properties of graphenes and nano-architected materials. In specific, there are parameters like orientation, elongation and topology of porosities which their influence on the thermal properties of engineered porous graphenes has not been yet examined. As a result, in this paper, we conduct extensive NEMD simulation to predict the effective thermal conductivity of engineered porous graphene sheets. We study the effect of topology, elongation, and orientation of pores to observe how the thermal conductivity of phononic engineered porous graphenes can be tuned. We investigate steady-state temperature distribution over the engineered porous graphene sheet to detect the maximum and the minimum temperature occurred around the pores. We compare the results of NEMD simulation and finite element method (FEM) to discover if an agreement can be observed between the effective thermal conductivity of engineered porous graphenes predicted by NEMD and FEM approaches. This research paves the way for further exploiting the potential application of phononic porous nanomaterials in thermal management for electronic devices and nanoelectromechanical systems.

2. Non-equilibrium molecular dynamics simulation

To better understand the heat transport phenomenon around the pore of the engineered porous graphene, the two-dimensional (2D) temperature distribution is obtained by employing the NEMD simulation. The NEMD simulation is implemented by LAMMPS package [77]; AIREBO [78] potential function has been employed to describe C—C and C—H interatomic interactions. However, it is known that pores in graphenes are unstable against filling by carbon adatoms, thus, the stabilization of such pore is a critical issue [79]. Hydrogen may be expected to stabilize pores in graphenes by passivating dangling bonds at the pore boundary [80]. Passivating dangling bonds by hydrogen atoms makes employing Tersoff [81] potential function for simulating the porous graphene samples impossible. Moreover, since the focus of this research is on the effect of pore topology on the normalized effective thermal conductivity, rather than its absolute magnitude, in this study, the AIREBO potential function has been used for predicting the thermal conductivity of graphenes. A schematic view of the developed atomistic model for defected engineered graphene sheet, for evaluating the thermal conductivity, has been shown in Fig. 1. In order to apply adiabatic thermal boundary con-

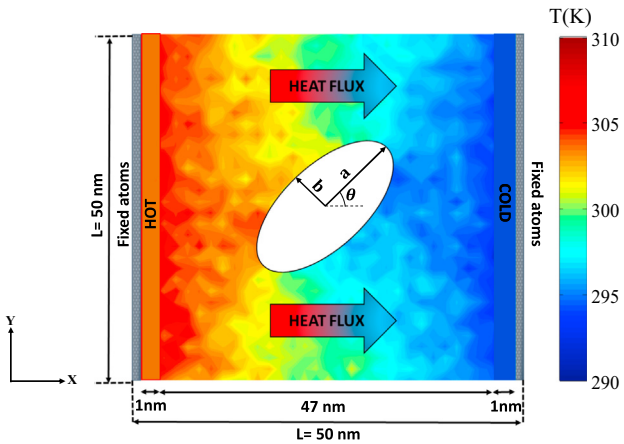

Fig. 1. 2D temperature distribution of an engineered porous graphene calculated by NEMD: major diameter, minor diameter and angle (orientation) of pore are represented by 2a, 2b and θ, respectively. Dimensions of the unit cell for the simulation are 50 nm × 50 nm × 0.314 nm [83].