Materials Science & Engineering A 556 (2012) 420–428

Contents lists available at SciVerse ScienceDirect

Materials Science & Engineering A

journal homepage: www.elsevier.com/locate/msea

Nanomechanics of free form and water submerged single layer graphene sheet under axial tension by using molecular dynamics simulation

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ARTICLE INFO

Article history:
Received 17 May 2012
Received in revised form
29 June 2012
Accepted 3 July 2012
Available online 6 July 2012

Keywords:
Graphene
Single layer graphene
Nano-fluid
Molecular dynamics
Nanomechanics.

ABSTRACT

The mechanical characteristics of single layer graphene sheet are studied in this work via molecular dynamics simulation method. The effect of loading direction, size of the graphene sheet and vacancy defects in the form of slits on the mechanical performance is investigated by subjecting the graphene sheet to tensile loading at various temperatures. The findings show superior tensile characteristics of the graphene sheet loaded in zigzag direction when compared to that of the armchair direction. Furthermore, the sheet size considerably influences the mechanical characteristics of graphene under tensile loading. Our findings reveal that the temperature and the location and quantity of defects significantly impact the elastic properties of graphene. However, the variation in mechanical properties due to defects is less pronounced at higher temperatures. Additionally, we also carried out the tensile loading of graphene submerged in water for its potential applications in nano-level fluid flow. Though, the presence of surrounding water medium weakens the tensile properties, the duration of elastic limit is still exceptional that makes graphene an ideal material for applications in nano-fluidic and nano-biological devices.

1. Introduction

Research in graphene has attracted significant interest in recent years due to its remarkable mechanical [1] and physical properties [2,3]. A single layer graphene sheet has the thickness of only one carbon atom which makes it the thinnest material [4] with a large specific surface area [5]. This feature of graphene makes it an ideal candidate for nanoelectromechanical systems (NEMS) [6] and nano-fluidic devices for applications in nano-level drug delivery [7], fluid separation [8] and nano-filtration [9]. These future applications require a critical understanding of the exceptional mechanical properties of graphene for its deployment in NEMS and nano-level biological devices. The mechanical properties of free-standing graphene was measured by Lee et al. [1] using atomic force microscope who established graphene as the strongest material ever measured. Theoretical studies on graphene are a popular mode of research, employing ab initio calculations or molecular dynamics (MD) simulation technique. Ab initio calculations were employed by Liu et al. [10] to study the ideal tensile strength of flat graphene sheet. They showed that the tensile strength of graphene depends strongly on the chirality of graphene and obtained a high Young's modulus of 1.05 TPa. This study further suggested that brittle cleavage fracture may be an inherent behavior of graphene and carbon nanotubes at low temperatures. Zhao et al. [11] investigated the mechanical strength of graphene subjected to tensile loading using the orthogonal tight-binding method and molecular dynamics simulations method. Their results indicated that the fracture strength and fracture strain is affected by the size and chirality of bulk graphene. The effect of cracks and large defects on the mechanical properties of carbon nanotubes and graphene sheets were studied by coupled quantum mechanics and molecular modeling approach by Khare et al. [12]. The numerical results indicate that the weakening effect of defects and cracks vary only moderately with the shape of the defect. Furthermore, they also showed that the strength of carbon nanostructures depends primarily on the cross section of the defect perpendicular to the loading direction and the structure near the fracture initiation point. Ni et al. [13] made use of MD simulation to confirm the mechanical anisotropy of graphene sheets. They demonstrated that the anisotropic mechanical properties were attributed to the hexagonal structure of the unit cells of the graphene. The variation of mechanical and thermal properties of graphene sheet with the nature of defects were analyzed by Hao et al. [14]. They found that while the variation of defect concentration has a little impact on the Young's modulus, the thermal conductivity of the graphene sheet is more sensitive. Tsai and Tu [15] investigated the mechanical properties of graphite flakes and single graphene layer were

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http://dx.doi.org/10.1016/j.msea.2012.07.008

investigated using MD simulation. They found that the graphene layers exhibit better elastic properties than the graphite flakes and suggested to expand and exfoliate the graphite flakes for providing better reinforcement effect in nanocomposites. The variation in fracture strength of graphene with temperature, strain rate, and crack length were investigated by Zhao and Aluru [16] using MD simulation method. They further deployed the quantized fracture mechanics (QFM) theory to predict the relation between fracture strength and crack length of graphene and compared the theoretical predictions with the classical MD simulations. Their findings show that the temperature plays a more important role in determining the fracture strength of graphene and found that the material becomes softer at temperatures exceeding 1200 K. Reddy et al. [17] modeled the graphene sheet in continuum to determine its elastic properties. They observed that the equilibrium adjustments of atoms have much influence on the computed elastic constants. Zhang et al. [18] investigated the mechanical properties of bilayer graphene sheets coupled by sp^3 bonding using MD simulation method. Their simulation suggested that these sp^3 bonds exert a strengthening influence on the interlayer shear modulus and load transfer rate, thereby enhancing the stability of the graphene sheet under axial compression. The above mentioned literature studies clearly demonstrates the exceptional mechanical qualities of graphene. However, the mechanical characterization of graphene in a fluid medium (for, e.g., water) has been clearly lacking in literature. Study of mechanical properties of graphene in water is of fundamental importance for its application in nano-fluidic devices. Furthermore, the influence of the size of graphene sheet and the combined effect of vacancy defects and the temperature on the mechanical properties of graphene also remains to be explored. In this paper, we employed MD simulation to find the influence of size, defects and temperature on a free form single layer graphene sheet subjected to axial tensile loading. We have further focused on the mechanical characterization of graphene sheet subjected to tensile loading submerged in water which has been compared with those of the free form graphene sheet.

2. Computational model

The numerical simulations in the present work are carried out entirely by using the classical molecular dynamics method [19], in which the interaction between the atoms are described by means of force field equations. Three forms of interaction are considered here, viz. interaction between the carbon atoms of graphene sheet, the interaction between the water molecules and interaction between the water molecules and the carbon atoms of graphene sheet. The Brenner's second generation bond order function (REBO) [20] is used to describe the covalent bonding of the carbon atoms in graphene sheet which is described mathematically as,

$$E_{\mathrm{REBO}} = V_R(r_{ij}) - b_{ij} V_A(r_{ij}) \tag{1}$$

where the repulsive and attractive pair terms are given by V_R and V_A, respectively. The b_{ij} term is used to include the reactive empirical bond order between the atoms.

The interaction between the individual water molecules are described using a flexible three-centered (F3C) water model [21]. The F3C model is capable of accurately fitting the experimental data detected by X-ray and neutron diffraction for different ranges of pressure, temperature, integration time steps and system sizes. An additional advantage of this model is that it employs a short-range truncation which is well-suited for high-speed computation of MD simulations involving large number of water molecules. The F3C water model is described as,

$$E_{\mathrm{F3C}} = E_{bond} + E_{bend} + E_{vdW} + E_{els} \tag{2}$$

where E_{bend} and E_{bond} are intra-molecular potential function that represents the bending energy and bond strength energy in a water molecule, respectively. The terms E_{vdw} and E_{els} denote, respectively, the van der Waals potential and electrostatic potential between water molecules. The complete details of this potential function can be referred to the work by Levitt et al. [21].

The Lennard-Jones (12-6) potential [22] is used to simulate the interactions between the water molecules and the carbon atoms to calculate the interaction of carbon to hydrogen and oxygen atoms. The Lennard-Jones (12-6) potential is given by,

$$E_{LJ} = 4\varepsilon\left[\left(\frac{\sigma}{r_{ij}}\right)^{12} - \left(\frac{\sigma}{r_{ij}}\right)^{6}\right] \tag{3}$$

where $\varepsilon_{\mathrm{CO}}=0.3135$ kJ/mol, $\varepsilon_{\mathrm{CH}}=0.253$ kJ/mol which is the well depth parameter, $\sigma_{\mathrm{CO}}=0.319$ nm, and $\sigma_{\mathrm{CH}}=0.282$ nm denote the collision diameter between carbon to oxygen and carbon to hydrogen, respectively [23].

3. Simulation details

The work described in this paper consists of two parts. In the first part, we focus on the mechanical properties of free form single layer pristine graphene sheet subjected to uni-axial tensile loading as depicted in Fig. 1, in which the graphene sheet is loaded along the armchair and zigzag directions. The effect of size on the mechanical properties of graphene sheet is studied by varying the aspect ratio of the graphene sheet while maintaining almost the same number of atoms. As described in Fig. 1, if the length of the graphene sheet is depicted by L and the width by W, the aspect ratio of the sheet is given by L/W. Tables 1 and 2 lists the dimensions and the number of carbon atoms of the graphene sheet classified based on the aspect ratio for the armchair and zigzag loading configurations, respectively.

The effect of vacancy defects on the mechanical properties of graphene sheet is studied by introducing defects in the form of slits in the graphene sheet. To adequately understand the effect of number and the location of defects on the tensile properties, we have used four forms of defect morphology as described in Fig. 2. Accordingly we define 'C1' defect in which a single slit is formed

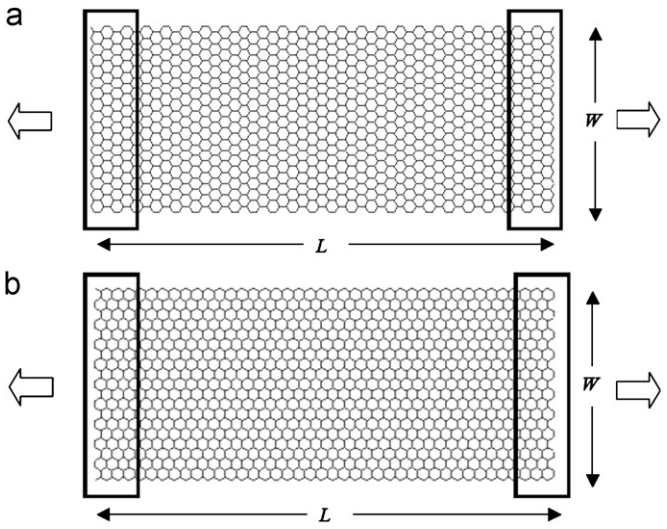

Fig. 1. Procedure of tensile loading of a single layer graphene sheet in (a) armchair and (b) zigzag direction. The end atoms enclosed within the rectangle are subjected to an outward displacement (strain rate=0.001 ps^{-1}) to effect tension.