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Molecular dynamics simulations on deformation and fracture of bi-layer graphene with different stacking pattern under tension

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ABSTRACT

Based on AIREBO (Adaptive Intermolecular Reactive Empirical Bond Order) potential, molecular dynamics simulations (MDs) are performed to study the mechanical behavior of AB- and AA-stacked bi-layer graphene films (BGFs) under tension. Stress–strain relationship is established and deformation mechanism is investigated via morphology analysis. It is found that AA-stacked BGFs show wavy folds, i.e. the structural instability, and the local structure of AB-stacked BGFs transforms into AA-stacked ones during free relaxation. The values of the Young's modulus obtained for AA-stacked zigzag and armchair BGFs are 797.2 GPa and 727.4 GPa, and those of their AB-stacked counterparts are 646.7 GPa and 603.5 GPa, respectively. In comparison with single-layer graphene, low anisotropy is observed for BGFs, especially AB-stacked ones. During the tensile deformation, hexagonal cells at the edge of BGFs are found to transform into pentagonal rings and the number of such defects increases with the rise of tensile strain.

1. Introduction

Monolayer graphene is characterized by the honeycomb lattice structure of hybridized SP^2 carbon atoms [1–3]. It exhibits superior property (e.g. super strength and extreme stiffness) that is promising for a wide range of applications in nanoelectronics, nanodevices and nanocomposites [4,5]. Graphene thus has attracted considerable attention from researchers worldwide.

In particular, the fundamental mechanical property of graphene is a topic of great interest in the areas of both nanomechanics and nanomaterials. In 2008, nano-indentation was performed by Lee et al. [6] to study the mechanical and thermal properties of monolayer graphene, where the Young's modulus $E = 1.0 \pm 0.1$ TPa (associated with thickness of 0.335 nm), the breaking strength about 40 N/m, and the thermal conductivity 5000 $\mathrm{W\,m^{-1}\,K^{-1}}$ (room temperature) were reported. Through experiments on epoxy composites, Rafiee et al. [7] found the influence of graphene on mechanical property, e.g. elastic modulus, tensile strength, fracture toughness, and resistance to fatigue crack, of composite material is remarkable as compared with single- and multi-walled carbon nanotubes. Using density functional theory, Liu et al. [8] found that the Young's modulus and Poisson's ratio for monolayer graphene are 1.05 TPa and 0.186, respectively. Based on orthogonal tight-binding method and MDs, Zhao et al. [9] studied the effects of size and chirality on the elastic property of graphene, where the Young's modulus of zigzag sheets was found to be larger than that of armchair ones. Similarly, size-dependent mechanical properties were studied to predict the non-linear behavior of graphene nanoribbons [10]. Results show that both linear and non-linear properties are strongly dependent on the structure as well as on the size of the graphene strip tested. By MD simulations, Zhang and Gu [11] investigated the effects of layer number, temperature and isotope on mechanical property of graphene which is found to be sensitive to temperature but insensitive to the layer numbers.

Up to now, most studies are focused on monolayer graphene [9,12–19], whereas multilayer graphene films (MGFs) have not received enough attention in spite of the fact that MGFs also hold great promise for various applications in nanotechnology [7,20,21]. Using molecular structural mechanics methods, Hosseini Kordkheili et al. [22] calculated the mechanical properties of double-layer graphene, where non-linear beam and truss elements were introduced to modeling different bonds. In particular, the stacking pattern and the interlayer van der Waals (vdW) interaction may lead to different mechanical property of MGFs from those of monolayer counterpart. Here it is noted that the bi-layer graphene film (BGF) is the simplest case and thus provide an excellent platform for the study of MGF mechanics. BGFs exhibit some prominent attributes, e.g. their optical [20] and electrical properties [21], which enable

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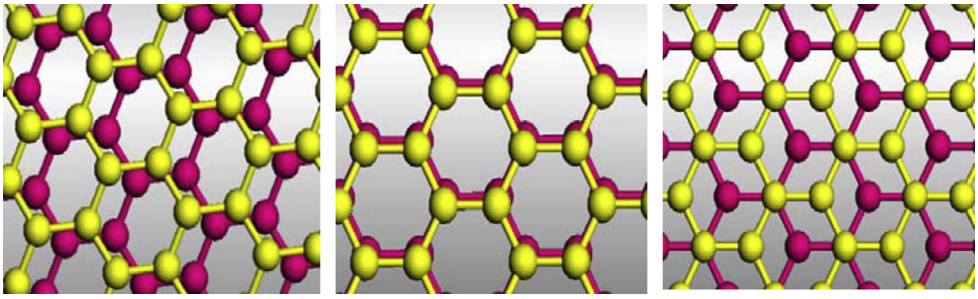

Fig. 1. Three typical stacking patterns of BGFs: (a) Mis-oriented; (b) AA-stacked; (c) AB-stacked. Yellow and red atoms represent top-level and underlying graphene respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

their use in field effect transistors. Furthermore, their double-layer structures can substantially reduce the influence of noise in the transistors by overcoming Hooge's rule which states that as transistors become smaller, the tiny electron charges inside them will threaten to overwhelm a desired signal with unwanted interference [23].

In this paper, MDs are performed to study the free relaxation and deformation mechanism of both AA- and AB-stacked BGFs subject to uniaxial tension. The whole paper is organized as follows: In section 2, geometrical models and simulation details used in the present study are briefly introduced. Results and discussions are included in section 3 where the stress–strain evolutionary relationship of BGFs is achieved, showing the tensile deformation process for BGFs of different stacking patterns. Finally, conclusions are summarized in section 4.

2. Geometrical models and simulation details

Two single graphene layers can be stacked in an arbitrary pattern as shown in Fig. 1(a). Such stacking order is referred to as mis-oriented or twisted pattern which is extremely unstable due to its high free energy. Thus, BGFs are usually found in the following two stable stacking modes, i.e. AA- and AB-stacked structures shown in Figs. 1(b) and 1(c), respectively. In the AA-stacked structure shown in Fig. 1(b), the vertical projections of the corresponding carbon atoms on the two layers completely coincide with each other. While for the AB-stacked structure shown in Fig. 1(c), half carbon atoms of the top layer sit directly on the top of the bottom layer atoms while the other half sit at the centers of the hexagonal rings in the bottom layer. Both AA- and AB-stacked structures are stable due to their relatively low energy and thus are the focus of some research [23].

In nanomechanics, MDs is considered as a standard technique that can be efficiently used to quantify the mechanical property and capture essential characteristics of nanostructures and materials. The basic principle of MD is to work out the force acting on individual atoms through the atom–atom interaction potential, and calculate their trajectory, acceleration and velocity by solving Newtonian mechanics equations. Here a certain number of molecules (atoms) under initial conditions are considered in the calculation which will be done with selected time step and boundary conditions. Subsequently, the statistical average of the obtained results will be outputted, from which the required macroscopic physical and mechanical quantities can be obtained. Also, it is a key issue to select a proper atomic potential function which determines the simulation accuracy for the calculation of MDs. AIREBO (adaptive intermolecular reactive empirical bond order) potential function [24] is mainly for carbons atoms and used to describe the atomic interaction within the same layer as follows:

$$E = \frac{1}{2}\sum_{i}\sum_{i\neq j}\left[E_{ij}^{RB} + E_{ij}^{LJ} + \sum_{k\neq j}\sum_{l\neq i,j,k} E_{kijl}^{TORS}\right] \tag{1}$$

where the second and third items in bracket represent a term for long-range interaction and a twisting term relying on dihedral angle, respectively.

The interlayer cohesion maintained by vdW force is characterized by 12-6 Lennard-Jones potential [25]:

$$V(r) = 4\varepsilon\left[\left(\frac{\sigma}{r}\right)^{12} - \left(\frac{\sigma}{r}\right)^{6}\right] \tag{2}$$

where $\varepsilon = 2.968$ meV and $\sigma = 0.3407$ nm.

In the present simulations, a uniformly distributed tensile force is applied on one side of rectangular BGFs in the perpendicular direction and the atoms on the opposite side are fixed. The geometrical model is square with length of 7.0 nm and the total number of atoms is 3944. Periodic boundary conditions are imposed on the two sides along horizontal direction, as shown in Fig. 2. Nose–Hoover method [26,27] is applied for isothermal adjustment at the temperature of 0.001 K. The low temperature is considered in order to avoid complex changes of mechanical property caused by atomic thermal activation. During the simulation process, unconstrained relaxation is first carried out on initial configuration to keep the system in an equilibrium state with the lowest energy. Then uniform tensile deformation is conducted on the BGFs. Here a time step 1.0 fs is selected and an increment of tensile strain 0.001 is imposed, followed by 1000 steps of relaxation for 1.0 ps. The calculated strain rate is of 1.0×10^9 S^{-1}. The stretching and relaxation process will be repeated to deform the BGFs. All simulations in this paper were conducted with the LAMMPS code developed by Sandia National Laboratory in US [28].

Young's modulus, strain and stress are traditionally defined in the framework of continuum mechanics. In the present work, these concepts will be extended to discrete BGFs consisting of two one-atom thick sheets. The equivalent thickness of graphene needs to be defined in an appropriate manner. The thickness of monolayer graphene is assumed to be 0.335 nm which is the interlayer spacing of multilayer graphene. Following this way, here we define the equivalent thickness of BGFs as $d = 0.670$ nm.

3. Results and discussion

To validate the MDs program described in section 2, we first studied the tensile behavior of a monolayer graphene and obtained its Young's modulus and tensile strength. Assuming the thickness of single-layer graphene is 0.335 nm and based on the stress–strain curves achieved for the sample graphene sheet, the obtained value of Young's modulus is 897.0 GPa, which is in agreement with available theoretical calculations of 0.8–1.1 TPa [6,29,30]. Tensile strength and fracture strain obtained for zigzag graphene are