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A semi-continuum mechanical model for analyzing the wrinkling of graphene sheet supported by an elastic substrate

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ABSTRACT

A semi-continuum mechanical model has been developed for analyzing the wrinkling of graphene sheet supported by a soft substrate. An energy based method is adopted, in which the elastic energies of graphene sheet and substrate as well as the interfacial van der Waals energy are considered. The graphene sheet and substrate are modeled by continuum mechanical method. In order to estimate the interfacial interaction directly, molecular models of graphene sheet and Polydimethylsiloxane (PDMS) substrate are combined with the continuum models. Graphene wrinkling due to the stretch-then-release process has been examined. The numerical results reveal that substrate deformation in the thickness direction leads to the initial wrinkled morphology of graphene sheet, which is further enhanced by the nonuniform interfacial van der Waals interaction. Upon releasing the substrate, the wrinkle wavelength of graphene sheet with initial wrinkled morphology decreases linearly with the released strain.

1. Introduction

Graphene possesses superb mechanical, electrical and chemical properties and has been used in high performance sensors and energy storage devices. Wrinkled morphology of graphene sheet which is induced by the stretching-then-releasing process makes it possible to fabricate stretchable devices [1,2]. The morphology of wrinkle is strongly affected by the stiffness of graphene sheet and substrate, and the interfacial interactions [3,4]. Analysis and modeling efforts have helped us in better understanding the mechanism of wrinkling of graphene-substrate system [5], improving the performance of graphene device and identifying the key micro mechanical parameters [6]. Major analysis and modeling methods include molecular dynamics [5,7–19], classical continuum mechanics [6,20–25] and molecular mechanics [26,27]. Molecular dynamics (MD) and classical continuum mechanics are the two most popular methods.

Zheng et al. [16] adopted molecular dynamics to study effect of functionalization on the wrinkling of graphene sheet. They found that functionalization of graphene would reduce the critical strain of wrinkling. Wang et al. [12] studied defect-guided wrinkling by MD and found that defects in graphene sheet can be used to control its wrinkling. Zhu et al. [17] modeled the wrinkling of graphene sheet on substrate-supported nanoparticles. The adhesion of graphene to substrate was assumed to be caused by van der Waals force. They adopted the continuum method developed by Jiang et al. [28] to simplify the process for calculating van der Waals energy. Tian et al. [15] modeled the wrinkling of boron nitride (BN) sheet under shear loading by MD, and determined the critical shear stress for wrinkling. Shen et al. [11] adopted MD to study the effects of functional groups with oxygen atom on the wrinkling of graphene sheet. The numerical results indicated that the functional groups at the edge of graphene sheet play a more important role in determining the wrinkled structure of graphene sheet than those on the face of graphene sheet. Huang et al. [14,13] and Tian et al. [7] adopt MD to study the wrinkling of graphene sheet under shear load and gave the critical load of wrinkling. Klaver et al. [9] constructed a molecular model of graphene sheet and copper substrate. The interfacial interactions were modeled by the charge-optimized many-body potential. The simulations revealed the wrinkling process of graphene-stiff substrate. Liu et al. [8] simulated the pulled out of graphene sheet from polymer substrate by MD. The results showed that the graphene sheet formed a wrinkled shape due to the effect of substrate.

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Even though much progress has been made, the modeling and analysis of wrinkling in graphene-substrate system by MD has been hindered by the constraints in the length and time scale. Compared to MD, the continuum mechanics method is more suitable for simulating the wrinkling of graphene-substrate system [28]. Zhang et al. [21] developed a continuum model to study the onset of wrinkling. The adhesive force is modeled by pair-wise Lennard-Jones 6–12 potential. The parameters of this potential are measured by a pressurized blister test [29]. However, the values of parameters varied with substrate material [25]. Zhang et al. [20] developed a continuum model to simulate the 2D wrinkling network of graphene sheet supported by stiff substrate. The adhesive energy density was given by an effective 3–9 Lennard-Jones potential [29]. Aitken et al. [30] and Teng et al. [31] developed a continuum model to analyze the stability of a graphene sheet on a stiff substrate. The graphene sheet was modeled as a thin elastic film while the energy of substrate is ignored. They assumed that the adhesive force was caused by van der Waals interaction. The concept of atom volume density is adopted to calculate the value of adhesive potential directly [24]. However this method may cause logical contradiction, since the distance between graphene sheet and substrate may be shorter than 0.6 nm [30] which is smaller than the length of covalent bond between carbon atoms of graphene. The continuum models developed by Huang et al. [32] and Pan et al. [22] can be used for analyzing wrinkles in a film supported by a compliant substrate. However these models are not able to calculate the interfacial van der Waals energy directly.

In a recent study by Gao et al. [33], a continuum mechanical method was developed to analyze multi-buckling induced wrinkle on graphene sheet supported by a soft substrate. The graphene sheet was modeled as a thin film; the interfacial adhesive stress was model by an effective 3–9 Lennard-Jones potential, the substrate stiffness was estimated using classical plate theory. The effects of randomly distributed debonded and bonded regions were considered. In order to model the effect of van der Waals interfacial interaction on wrinkling of graphene sheet supported by a soft substrate directly in the present study, a semi-continuum mechanical model is developed. A continuum mechanical method is applied to calculate the elastic energy associated with the deformations of graphene sheet and soft substrate. The interfacial van der Waals energy is evaluated by an atomistic method. An energy based method is adopted to study the morphology due to wrinkling growth.

2. Model

2.1. Combination of continuum method and atomistic method

Based on the pre-stretching and releasing process [2], the substrate is stretched to a strain ε_0 first, and then the graphene sheet is attached onto the substrate. Subsequently, the substrate is released. The graphene sheet is assumed to be perfectly bonded to the substrate and contracts with the substrate. Thus, the released substrate creates a compressive strain on graphene sheet. As shown in Fig. 1, the displacements in x direction at the two ends of graphene sheet are assumed to be identical to the corresponding displacements of the substrate. $\bar{\varepsilon}_{gra}$ and $\bar{\varepsilon}_{sub}$ denote, respectively, the average strain of graphene and substrate in x direction. ε_r is the released strain of the substrate. So $\bar{\varepsilon}_{gra} = \frac{-\varepsilon_r}{(1+\varepsilon_0)}$ and $\bar{\varepsilon}_{sub} = \varepsilon_0 - \varepsilon_r$.

A major difficulty in analyzing wrinkling of graphene-substrate system by classical continuum mechanics is how to directly evaluate the interfacial interaction. This difficulty can be resolved by considering the atomistic structure. The idea of combining continuum mechanical model and molecular model is shown in Fig. 2.

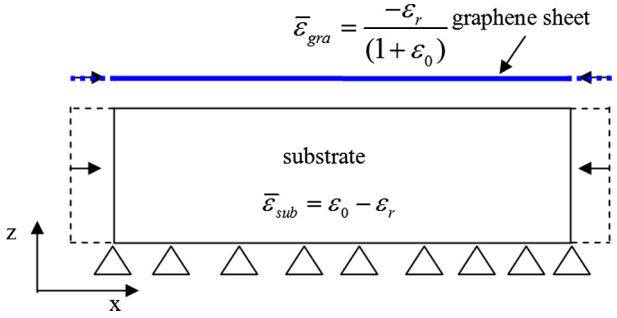

Fig. 1. Model for compressing the graphene-substrate system.

A plane strain condition is assumed for the model of Fig. 2. Beam elements and 4 node plane elements are used to describe the deformation of graphene sheet and polydimethylsiloxane (PDMS) substrate respectively. Even though the graphene sheet and substrate have finite thickness, it is assumed that there is no displacement in y direction and all variables are the functions of x and z only. Each beam element is filled with sp3 carbon atom while each plane element is filled with polymer molecules. In this study, PDMS is adopted as the substrate. The van der Waals force is responsible for interfacial interaction. Then, the system energy includes three parts, namely elastic deformation energy of graphene $U_{g\_elastic}$, elastic deformation energy of PDMS substrate $U_{pdms\_elastic}$ and van der Waals energy between graphene sheet and PDMS substrate U_{vdw}.

$$U_{total} = U_{g\_elastic} + U_{pdms\_elastic} + U_{vdw} \tag{1}$$

$$\text{Here,} \quad U_{g\_elastic} = \frac{E_g}{2}\int_{\Omega 1} \varepsilon^2 \mathrm{d}s \tag{2}$$

and E_g is the elastic modulus of graphene, Ω1 denotes the region of graphene sheet, ε is strain of graphene in the x direction. The elastic energy of PDMS is

$$U_{pdms\_elastic} = \frac{1}{2}D_0\int_{\Omega 2}\begin{bmatrix}\varepsilon_x\\ \varepsilon_y\\ \gamma_{xy}\end{bmatrix}^T\cdot\begin{bmatrix}1 & \nu_0 & 0\\ \nu_0 & 1 & 0\\ 0 & 0 & (1-\nu_0)/2\end{bmatrix}\cdot\begin{bmatrix}\varepsilon_x\\ \varepsilon_y\\ \gamma_{xy}\end{bmatrix}\mathrm{d}s \tag{3}$$

where $D_0 = \frac{E_{pdms}}{1-\nu_{pdms}^2}$, $\nu_0 = \nu_{pdms}$. E_{pdms} and ν_{pdms} are the elastic modulus and Poisson's coefficient of PDMS respectively. Ω2 denotes the region of substrate. The interfacial van der Waals energy is

$$U_{vdw} = \sum U_{ij} \tag{4}$$

where U_{ij} is the van der Waals energy between ith sp3 carbon atom and jth atom in PDMS.

Experimental studies on graphene-substrate deformation of [34], [1,2] concluded that the graphene sheet tend to maintain a wavy shape since the energy of this configuration is lower than that of the flat sheet. Based on the experimental observations, the following deformed configurations which were proposed by Teng and Zhao [31] are adopted for graphene and substrate.

$$w_g(x) = A_g\cos\frac{2\pi x}{\lambda} + h,\ \ w_s(x, z = h_s) = A_s\cos\frac{2\pi x}{\lambda} \tag{5}$$

where w_g is the z-direction displacement of graphene, w_s is the displacement of substrate in z-direction, λ is the wave length, h is the distance between central layer of graphene sheet and the top surface of substrate, h_s is the thickness of substrate, A_g and A_s are the amplitudes of graphene sheet and substrate, respectively. The sinusoidal shape formulations have also been adopted in [21,30], and