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A molecular dynamics based cohesive zone model for predicting interfacial properties between graphene coating and aluminum

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ABSTRACT

A cohesive zone model (CZM) based on a traction–separation (T-S) relation is first developed to simulate the interfacial behavior between graphene coating and aluminum (Al) substrate. The CZM parameters, which are very difficult to obtain directly experimentally, are determined using molecular dynamics (MD) simulation. Specifically, the MD simulations under the normal and shear loadings are conducted on the graphene-coating/Al interface to derive its T-S relation and then the relevant interfacial behavior of the composite is identified. The MD results show that the behavior of the interface between graphene coating and Al substrate under normal and shear loading is temperature dependent. The maximum normal tensile stress at the interface decreases gradually while the temperature increases from 150 K to 600 K. But the maximum shear stress increases as the temperature increases from 150 K to 450 K and then decreases as the temperature increases from 450 K to 600 K. Finally, the CZM parameters are determined and then imported into a finite element (FE) model. The blister test results obtained by the FE method are in good agreement with those obtained by the MD simulations. These results suggest that the proposed approach is efficient in determining the CZM parameters of the interfacial behavior between the substrate and the ultrathin coating.

1. Introduction

Though graphene has a Young's modulus of 1.1 TPa and a high intrinsic strength of 125 GPa, its independent use as a structural material is still problematic because it is a two-dimensional crystal of atomic thickness [1]. In contrast, aluminum is widely used as a strengthening structural material because of its high specific strength and light weight. Graphene fragments, as a promising strength enhancer in composites [2], have been demonstrated to improve the strength and toughness of composites, even without ordered arrangements [3,4]. On the other hand, Al always severs in a complex environment. Coating is a popular way to protect the Al products. Recently, Kirkland et al. [5] reported that, compared with alumina coatings, graphene coatings possess many unique properties that are especially suitable for the lightest and thinnest protective barriers of metal components, due to their excellent electrical conductivity, heat resisting property, chemical inertness, and transparency. Chemical vapour deposition (CVD) can produce high-quality graphene films on a metal (for example, Cu, Ni, Pt or alloy) surface at high temperatures, and the films are then transferred to other substrates [6]. It should be noticed that a chemical reaction between Al and graphene can lead to the formation of Al carbide (Al_4C_3) at a higher temperature, which weakens the mechanical properties of graphene-Al composites [7]. So the graphene can be coated on Al surface by using the transferring method.

Many studies have investigated the mechanical properties of graphene/metal nanolayered composites [8–13]. The high intrinsic strength and modulus of graphene dispersed into a metal can effectively constrain dislocation movement in the metal, significantly strengthening the metal. However, graphene-reinforced metal composites exhibit strong interfacial effects under a variety of load conditions. Liu et al. used a molecular dynamics (MD) method to investigate the interfacial strengthening and self-healing mechanism of graphene/copper nanocomposites under shear loading [14]. The interfacial behavior between the graphene and the matrix, especially in the case of a graphene coating on the matrix, plays a significant role in determining the mechanical properties of graphene/matrix composites. In existing finite element (FE) simulations of such composites at micro scale, the interface was often modeled by a cohesive zone model (CZM) in which a traction–separation (T-S) relation between the matrix and the reinforcement phase was employed to describe the matrix/reinforcement

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interaction. However, the cohesive zone law is not universal, and it takes different forms for different material interfaces. Traditionally, the CZM parameters have usually been determined by the quantitative T-S relation that was often obtained from experiments [15,16]. Yet it is very difficult to conduct corresponding experiments with a graphene coating on a matrix. Moreover, the high expense of experimental methods as well as the effect of factors such as lattice mismatch, thermal expansion coefficient mismatch, and crystal defects on the experimental results leads to scattered experimental data and low efficiency. Alternatively, CZMs have been replaced by numerical or analytical modeling of fracture processes. A macroscopic cohesive method introduced by Needleman [17] has been widely used to describe the cohesive zone type interface model, and recently, a modified T-S rule was proposed by Sazgar and Movahhedy [18] to take account of the temperature effect. On the other hand, at atomic scale, MD simulations have been used to predict interfacial behavior between particles and matrix. Gall et al. were the first to obtain the T-S relation by atomistic simulations, when investigating atomic debonding in a silicon/aluminum interface [19]. Subsequently, numerous MD studies were used to investigate the deformation and fracture behavior of single crystal metals such as Al, Cu, Ni, Fe, Cr, W [20], and bi-crystal Al [21,22] under tensile loading. Besides, many MD simulations have been used to study interfacial behavior between two different materials [23–26]. Recent attention has been paid to a multi-scaled CZM. For instance, Dandekar used the local values from MD simulation to derive the global T-S relation for an Al/SiC interface, and imported the resulting higher scaled CZM parameters to the FE model [27]. Sazgar and Movahhedy [18] developed an MD-based modified Needleman CZM for the prediction of equivalent temperature-dependent material behavior in an Al/Al_2O_3 composite. Comparison of MD simulations and experimental results [18,27] demonstrated the effectiveness of the MD-based CZM approach. Near room temperature, however, graphene has a negative coefficient of thermal expansion (CTE), and the absolute values of the CTEs of graphene first increase and then decrease with increasing temperature in range of 0 K to 600 K [28]. The changing trend of the CTEs of graphene with temperature change is completely unlike the behavior of metal, leading to curious interfacial properties when graphene is coated on metal. Research on this anomaly is still unexplored.

On the basis of the abovementioned research, MD and theoretical analysis are used in this work to study the interfacial behavior of graphene coating on substrate Al under tensile and shear loadings. Then, the Lennard-Jones (LJ) potential function is applied to estimate the quantitative T-S relation used in the CZM model, after which the relevant CZM parameters are imported into the FE simulations to investigate the deformation of graphene coating on the substrate Al. Finally, to verify the proposed method, the blister test, a well-known method for measuring the adhesion of thin films to their substrate, is simulated by using both the FE method with the CZM parameters from the proposed approach and the MD method.

2. Methods and models

In this study, a monolayer graphene is coated on substrate aluminum. It is well known that the in-plane Young's moduli of graphene are very strong, whereas the out-of-plane Young's moduli are quite weak [29]. Therefore, the effect of the chirality of graphene is ignored because of the weak out-of-plane interaction between the coating and substrate. The typical (1 0 0) and (1 1 1) stacking planes of substrate Al are taken into account for investigating the interfacial behavior between the graphene coating and the substrate Al. The two crystal orientation planes of aluminum and graphene are shown in Fig. 1a. Thus, there are two assemblies of graphene-coating/Al composites (GA) with different crystal stacking between the graphene and aluminum layers: graphene-Al (1 1 1) (GA1 1 1) and graphene-Al (1 0 0) (GA1 0 0). To arrive at an acceptable size, various dimensions for the simulation box along the directions with periodic boundary conditions and free boundary conditions are examined, and finally the dimensions of $196.8 \times 42.6 \times 201.71$ Å are obtained for the simulation box. During the tension process, the boundary in z-direction is set as free, and the boundaries in x- and y-directions are set periodic. While during the shear process, the boundaries in x- and z-directions are set as free, and the boundary in y-direction is set periodic. The graphene is set as a rigid body while applying tensile or shear loadings in our simulations, and this setup has been used by Xu and Buehler [30] to calculate the binding energy of graphene/metal via the first-principle method. Fig. 1b shows the initial configuration of the MD simulation built within the MD package of a Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) [31].

The interactions between carbon atoms are described by the adaptive intermolecular reactive empirical bond order (AIREBO) potential [32,33], and those between aluminum atoms are described by an embedded atom model (EAM) [34–36]. Because aluminum and carbon atoms near the interface do not form chemical bonds in the transferring process, the Lennard-Jones (LJ) 12-6 potential is used to describe the interactions of carbon/aluminum [37], which is rather useful in describing the adhesive interface between different components than the Morse potential [27]. The LJ interaction is written as $U_{LJ}(r_{ij}) = 4\varepsilon[(\sigma/r_{ij}{}^{12}| - (\sigma/r_{ij})^6]$ $(r_{ij} \leqslant r_c)$ [38,39], where $\boldsymbol{r_{ij}}$ is the distance between those atoms not forming bonds, $\boldsymbol{\varepsilon}$ is the well depth, $\boldsymbol{\sigma}$ is the size parameter, and $\boldsymbol{r_c}$ is the cutoff distance. $\boldsymbol{\varepsilon}$ and $\boldsymbol{\sigma}$ are 0.03507 eV and 3.0135 Å, respectively [40,41]. The cutoff distance $\boldsymbol{r_c}$ should be taken to be 3 $\boldsymbol{\sigma}$ or greater, as recommended in [41], so here we employ a cutoff distance of 9.0405 Å in our simulations. Initially, both aluminum and graphene are kept in contact along the interface in the x-z plane with a separation distance of 3.014 Å. The initial interface separation distance is chosen to be close to the equilibrium bond length of Al-C. The equilibrium bond lengths from ab initio calculations and experiments for the Al-C are in the range of 2.31–3.36 Å [35]. Fig. 2 gives the pair distribution function in the GA111 at 300 K, from which it can be seen that, after relaxation, the interface separation distance of the graphene/Al system is measured in the range of 2.37–2.52 Å. This result is consistent with the equilibrium bond length of Al-C. The simulation box is first kept at the constant temperature for 40 ps by Langvin thermostat to permit a reasonably equal partitioning of the kinetic energy at the beginning. Then, the isobaric-isothermal ensemble (NPT) is utilized for 50 ps to maintain a constant temperature and then impose the pressure of 1 bar to obtain the initial physical state of the material. After equilibrium, the load is finally applied to the simulation box to obtain a T–S relationship curve. All simulated time steps are selected as 1 fs.

To apply shear, tensile, and fixed loads to certain atoms, the areas 8 Å thick at the bottom of substrate Al and the graphene layer are considered to be the loading region (regions **a**, **b**, and **c** in Fig. 1), wherein the graphene layer (region **a** in Fig. 1(c)) is subjected to a tensile force in the z-direction, and to a shear force (region **c** in Fig. 1(d)) in the x direction. Region **c** in Fig. 1(c) and (d) is fixed during loading. Due to the abrupt change in the velocity of the atom, the uniform stretching of the system keeps the atom free from computational shock [42]. The values of tensile and shear stresses at the interface are obtained by averaging the values of atomic virial stresses in the entire simulation box. In order to reduce or even eliminate the influence due to elastic deformation, the open displacements of the interface are estimated by the mean atomic displacements of the graphene minus those of the three-layer Al atoms closest to the interface (that is, the thickness of a primitive cell of single crystal Al). Atomic quantities are ensemble averaged both in time and space every 100 fs. Visualization is performed using OVITO [43].

3. Results and discussion

3.1. Traction–separation model

Tensile and shear tests are performed to obtain the T-S relationship