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Strengthening mechanisms of graphene coated copper under nanoindentation

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ABSTRACT

We conduct molecular dynamics simulations to investigate the strengthening mechanisms of monolayer graphene coated copper during nanoindentation. It was found that the fracture of the graphene coating includes three stages: (i) initiation of decagon defects, (ii) growth of decagon defects, and (iii) formation of pearl necklace like structure. After the graphene breaks, the intact graphene coating around the indentation still plays a role in strengthening the copper substrate. The shear modulus mismatch and the lattice mismatch at the interface result in the image and misfit stresses, which act as a barrier to impede dislocations to slip out of the copper surface, thereinto, the image stress is the dominant. In addition, it is interesting to observe pristmatic partial dislocation loop and twinning planes during the indentation.

1. Introduction

Graphene is a single layer material consisting of covalently bonded carbon atoms and has been demonstrated excellent mechanical properties [1–5], thermal and electrical conductivity [6–8], and good anti-oxidation capacity [9]. Consequently, graphene can be used as one layer coating to protect or enhance the surface properties of other materials such as copper (Cu). Recently, graphene/Cu multilayers have been produced to improve the overall mechanical performance [6,10–12]. For example, Koltsova et al. synthesized a Cu/graphene composite and achieved 39% increase in the hardness than pure Cu [11,13].

Kim et al. [12] conducted compression test using nanopillars consisting of alternate layers of Cu and graphene, and found that the improved strength is due to the Cu/graphene interface which blocks the propagation of dislocations. Although the heterogeneous interface is a strong barrier to dislocation propagation, dislocations piled up on one side of interface can induce dislocations nucleation on the other side under shock loading [14]. Liu et al. [15] investigated the graphene-Cu nanolayered (GCuNL) composites under shear deformation and concluded that the enhanced shear stress and toughness of the GCuNL composites result from the constraint from the graphene-Cu interface. The mechanical properties of a single-crystal copper nanosheet and graphene nanoribbon-embedded copper (GNR/Cu) nanocomposite sheet were investigated using MD simulations. It was found that the Young's modulus, tensile strength and fracture strain of the composites are much higher than the single-crystal Cu [16]. The researchers not only studied the strengthening role of graphene as interfaces, but also took into consideration the role of graphene as coatings. The frictional characteristics of monolayer, bilayer and trilayer graphene/Cu(1 1 1) systems under nano-scratch were investigated and it was suggested that the frictional coefficient decreases with the increase of the number of graphene coating layers [17].

However, the underlying strengthening mechanisms have not been well understood. In this work, we conducted MD simulations of nanoindentation on a single layer graphene coated Cu, with a focus on the strengthening mechanism from the graphene coating.

2. Methods and modellings

2.1. Interatomic potentials

The interatomic forces within the graphene layer are modelled using the optimized Tersoff potential which makes the lattice of single layer graphene well match that Cu [18,19]. For copper, the

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well-known embedded atom method (EAM) potential [20] is used to depict the interaction between atoms with the parameters by Mishin et al. [21], with which the elastic coefficient, cohesive energy and stacking faults can be obtained satisfactorily. The interactions between the graphene and copper atoms are described with a L-J potential with the parameters $\varepsilon_{\text{Cu-Graphene}}$ = 19.996 meV and $\sigma_{\text{Cu-Graphene}}$ = 3.225 Å [15,22,23]. The interaction between the diamond indenter atom and copper atom are described with the two-body Morse potential, which is suitable to reveal the surface contact property and has been widely used in the nano-indentation simulations [24–26]. The van der Waals interaction between the diamond indenter atom and graphene atom is modelled with L-J potential with $\varepsilon_{\text{Graphene-C}}$ = 2.967 meV and $\sigma_{\text{Graphene-C}}$ = 3.407 Å [27]. The indenter is assumed rigid so that the interaction between the atoms in the indenter is ignored to save computation time [25,26,28].

2.2. MD model

The sample for configuration simulation consists of Cu (1 1 1) cubic substrate with three side lengths of 255.5 Å, 256.1 Å and 150.7 Å, respectively, coated with a graphene layer (Fig. 1). In the general, graphene can be coated on different crystal planes of Cu using chemical vapor deposition (CVD) method. It was confirmed high quality monolayer graphene can be produced on Cu(1 1 1) [29] and therefore we selected Cu(1 1 1) in our simulations. A cube corner diamond indenter is used (Fig. 1) and stress concentration at its tip and edges can easily break the graphene coating so that reinforcement efficiency of graphene can be examined. Before indentation, the indenter is placed 8 Å above the graphene coating surface to avoid the interaction between the indenter and the sample. Before indentation, the sample is relaxed using the time step of 1 fs for 100 ps in a NPT ensemble with the periodic boundaries applied in *x*, *y* and *z* directions for equilibrium. The system temperature is kept at 10 K to avoid the random vibration of atoms. During indentation, the atoms in the bottom three layers of the copper substrate are fixed in the *z*-direction to prevent the substrate from shifting vertically, while in the *x* and *y* axes periodic boundary conditions are applied. The NVE ensemble is adopted to keep the number of atoms (N), the volume (V) and the energy (E) of the graphene/Cu nanolayers unchanged during indentation. Langevin thermostat is used to ensure the indentation is performed at 10 K [30]. The height of the cube corner diamond indenter is 60 Å, and it moves downwards at a constant speed of 10 m/s during indentation [31,32]. The MD simulations are proceeded with large-scale atomic /molecular massively parallel simulator (LAMMPS) [33].

2.3. Visualization of defects

Graphene is kind of a two-dimension material, and the defects in which during indentation are easier to be observed. The dislocation extraction algorithm (DXA) [34] constructs the Burgers circuits and divides the region into good and bad areas which is used to visualize various dislocations, stacking faults and twin boundaries, etc. DXA can also be used to determine the Burgers vectors to identify the dislocation patterns. The software OVITO [35] is used to display atomic configurations.

3. Results and discussion

3.1. Fracture of graphene under indentation

The typical indentation load-depth (*P*-*h*) curves are shown in Fig. 2. Overall, the force in *P*-*h* curves for GCL is much higher than that of CuL attributed to the contribution from the graphene layer. There are some sharp load drops at Points c, d, e, f, g and h in the *P*-*h* curve of GCL, which is different from CuL. In the initial elastic stage, the *P*-*h* curve of GCL is higher than that of CuL, attributed to the contribution from the larger elastic modulus of Graphene. A small platform can be observed at Point a, consisting of two small load drops and two valley bottoms, indicated as a_1 and a_2 in the

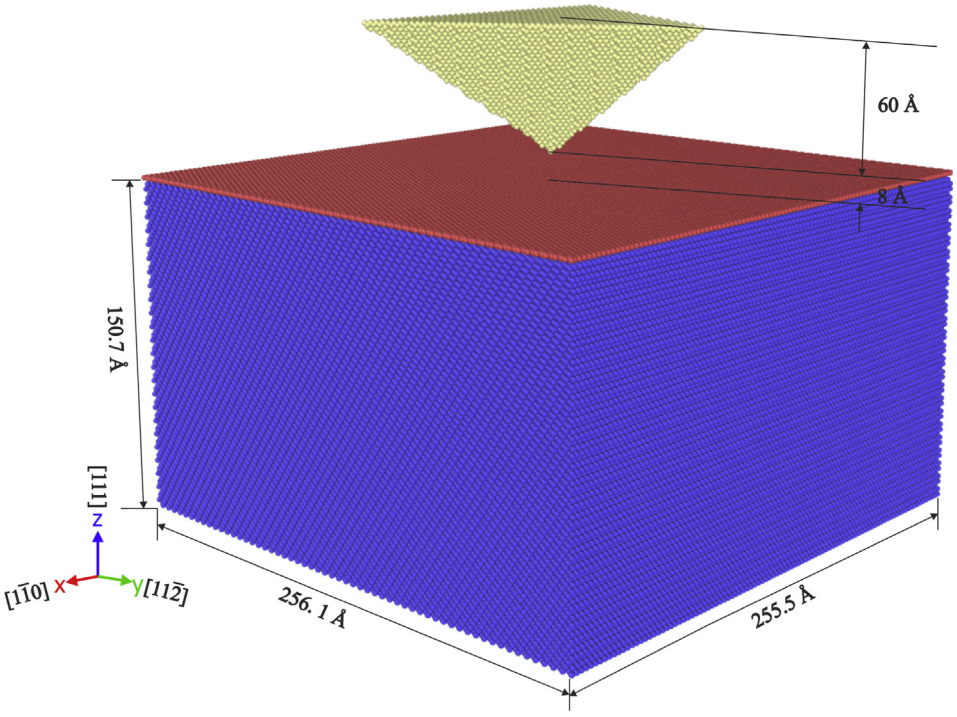

Fig. 1. The MD indentation model. Yellow, red and blue colours represent diamond indenter, graphene coating and Cu layer, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)