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Understanding the mechanical properties and deformation behavior of 3-D graphene-carbon nanotube structures

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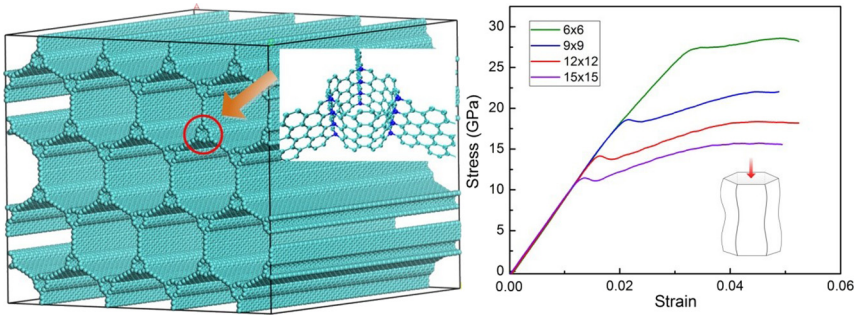
HIGHLIGHTS

- A new three-dimensional graphene-carbon nanotube (3-D GR-CNT) structure has been proposed, which GR and CNT connected using covalent bonds.
- The 3-D GR-CNT structure has outstanding mechanical properties, especially the ultrahigh Young's modulus and the specific strength.
- The 3-D GR-CNT structure dose not have plastic deformation during the compression.

GRAPHICAL ABSTRACT



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ABSTRACT

Here, a new three-dimensional graphene-carbon nanotube (3-D GR-CNT) structure was proposed, and the compressive mechanical properties and deformation behavior of the 3-D GR-CNT structure were evaluated using molecular dynamics (MD) simulations. It was found that the 3-D GR-CNT structure had outstanding mechanical properties, especially the ultrahigh Young's modulus, which was up to 1018 GPa and as high as that of GR. The 3-D GR-CNT structure did not have plastic deformation during the compression. The effects of GR length and CNT diameter were evaluated, and it was demonstrated that when the ratio of CNT diameter to GR length was about 0.6, the ultimate stress of the 3-D GR-CNT structure was the highest. Owing to the low density of the 3-D GR-CNT structure, the structure had outstanding specific strength. At a small compressive deformation, GRs produced buckling deformation with wrinkles to resist compression. After reaching the critical buckling stress, CNTs began to produce wrinkles; and after reaching the failure stress, the destruction started from the junctions. In addition, compared with the sp^3 carbon atoms, the sp^2 carbon atoms were more suitable for the junctions, as more energy could be absorbed by the sp^2 carbon atoms in the junctions.

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1. Introduction

Searching for new carbon allotropes has been an enduring effort for material scientists [1]. Since the discovery of zero-dimensional (0-D) fullerenes (C_{60}) in 1985 [2], other types of carbon allotropes have been discovered, such as 1-D carbon nanotube (CNT) [3] and 2-D graphene (GR) [4]. Among the carbon allotropes, both GR and CNT have unique properties, such as excellent mechanical properties, high thermal conductivity, and large electrical conductivity [5–10], especially, the ultrahigh tensile strength of GR in-plane and CNT along the axis [5,9,11]. However, the compressive strength of GR in-plane and CNT along the axis is relatively low. Scaling these carbon allotropes up to a 3-D structure is an effective way to enhance the compressive strength of GR in-plane and CNT along the axis, and hence, make the 3-D structures more practical in device applications. The 3-D carbon structure has other potential applications. For instance, it can be used as anode materials in lithium-ion batteries, supercapacitors, water treatment membranes, gas and liquid storage and structural composites [12–16].

A number of 3-D GR honeycomb structures have been hypothesized and fabricated [14,15,17–20]. The material properties of the 3-D GR honeycomb structures have also been studied. For example, Kawai et al. [21] verified the formation possibility and investigated the thermal stability of 3-D GR honeycomb structures based on tight-binding molecular dynamics (TNMD). Zhang et al. [18] investigated the electronic and mechanical properties, as well as the localized compression behavior of the 3-D GR honeycomb structures. Using the first-principles density functional theory and molecular dynamics simulations, Pang et al. [13] found that the 3-D GR honeycomb structure had superb mechanical and thermal properties.

However, previous studies focused on the 3-D GR honeycomb structure, which only consists of GR sheets. Compared with GR in-plane, the compressive mechanical properties of CNT along the axis are much better. Therefore, combining the GR and CNT may be an effective way to improve the compressive mechanical properties of 3-D carbon structures. Recent success in the synthesis of carbon honeycomb structures [17] shows a great potential in constructing new 3-D GR-CNT structures, covalently formed by CNT and GR.

In the present work, we proposed a new 3-D GR-CNT structure, in which the GR and CNT are connected by carbon carbon covalent bonds, formed by sp^2 and sp^3 carbon atoms. The out-of-plane compression was applied to investigate the compressive mechanical properties and deformation behavior of the 3-D GR-CNT structures. Computational results indicated that the new 3-D GR-CNT structure had ultrahigh Young's modulus and large ultimate stress. Owing to the low density, this structure had high specific strength. Through unloading test, we found that both types of the 3-D GR-CNT structures were elastic. The effects of GR length and CNT diameter were evaluated, and it indicated that when the ratio of CNT diameter to GR length was about 0.6, the ultimate stress of the 3-D GR-CNT structure was the highest. When the compressive stress was lower than the critical buckling stress, GRs produced buckling deformation with wrinkles to resist compression. After reaching the critical buckling stress, CNTs began to produce wrinkles, and after reaching the failure stress, the destruction started from the junctions. Owing to the sp^2 carbon atoms in the junctions absorbed more energy than the sp^3 carbon atoms, the Young's modulus and ultimate stress of type-I were better than that of type-III.

2. Computational method

In this work, the GR and CNT were used to construct a new type of 3-D GR-CNT structure, and C—C covalent bonds were used to connect the GR and CNT. The computational structure can be seen in Fig. 1 (i.e., type-I). As inspired by the work of Stampfer [22], the motivation for venturing into this new 3-D carbon structure stems from that it is possible to fabricate it by using the following process: first, creating a 3-D GR-CNT structure template in substrate using photolithography, based on the created template and then applying chemical vapor deposition (CVD) [23], which is similar to the fabrication of GR. In the junctions, each carbon atom (represented by atoms in blue) connects with *three* neighboring atoms (i.e., *three* covalent bonds), as illustrated in Fig. 1(b), indicating that the sp^2 carbon atoms were used to connect the CNT and GR in the junctions. According to the change of GR length and CNT diameter, the in-plane dimensions of the simulation systems ranged from 169 Å to 283 Å in the *x*-axis, and 180 Å to 367 Å in the *y*-axis. For the out-of-plane dimension, i.e., the *z*-axis, the height was about 150 Å.

Large-scale molecular dynamics simulations were conducted using the LAMMPS package [24]. The adaptive intermolecular reactive empirical bond order (AIREBO) potential [25] was applied to describe the interatomic interactions. To avoid a non-physical post-hardening behavior during the deformation computations of carbon materials, the first nearest cut-off radius (r_{cc}) in the AIREBO potential should be modified in the range of 1.92–2.0 Å [26–29]. For the present work, 1.92 Å was adopted in all simulation systems [26,27]. In order to eliminate the boundary effect, the periodic boundary conditions were imposed in all directions. To ensure zero stress conditions in all three directions, the computational models were relaxed in isothermal-isobaric (NPT) ensemble at a temperature of 300 K for about 500 ps, and then a constant compressive strain rate was applied in the out-of-plane, i.e., the *z*-axis. At the same time, the time step of 1 fs was applied to all simulation systems. In order to understand the thermal stability, a higher temperature, i.e., 1000 K, was used to relax the 3-D GR-CNT structure. The computational results indicated that the configuration of the 3-D GR-CNT structure in Fig. 1 did not change at such a high temperature.

3. Results and discussion

3.1. Mechanical properties

The strain rate is critical for the mechanical properties and deformation behavior of materials [30–32]. To eliminate the effect of compressive strain rate on the computational results, five different compressive strain rates were applied to the simulation systems, i.e., 5×10^{-2} ps^{-1}, 10^{-2} ps^{-1}, 10^{-3} ps^{-1}, 10^{-4} ps^{-1} and 10^{-5} ps^{-1}. The computational results indicated that the ultimate stress of the 3-D GR-CNT structures was insensitive to the compressive strain rate when the compressive strain rate was lower than 10^{-3} ps^{-1} as shown in Fig. 2. Therefore, a compressive strain rate of 10^{-3} ps^{-1} was applied to all computations, as it can reduce the computational cost.

In order to understand the effect of GR length on the mechanical properties of the 3-D GR-CNT systems, four different models were built, which consist of 6×6 armchair CNTs and different lengths of GR sheets in the in-plane (7–13 carbon hexagons were chosen to construct different GR lengths). From Fig. 3, we observe the variation of stress σ during the compressive deformation, and σ can be obtained by [33]:

$$\sigma_{ij} = \frac{1}{V}\sum_{\alpha=1,n}\left(m_\alpha v_\alpha^i v_\alpha^j + \frac{1}{2}\sum_{\beta=1,n}^{n} r_{\alpha\beta}^i f_{\alpha\beta}^j\right) \tag{1}$$

where, *i* and *j* denote the indices in Cartesian coordinate systems, i.e., 1, 2, and 3, while α and β are the atomic indices, m_α and v_α denote the mass and velocity of atom α, $r_{\alpha\beta}$ is the distance between atoms α and β, $f_{\alpha\beta}$ is the force on atom α due to atom β, *n* represents the number of atoms, *V* is the total volume occupied by all atoms and can be obtained using Eq. (2).

$$V = N(A_{GR} + A_{CNT})H = N(4\pi Dt + 6Lt)H \tag{2}$$

where, A_{GR} and A_{CNT} are the area of GR and CNT for a unit cell, respectively, *N* is the number of unit cell, *D* represents the diameter of CNT, *L*