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Mechanical properties of graphene based nanocomposites incorporating a hybrid interphase

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ABSTRACT

The elastic mechanical properties of graphene monolayer based nanocomposites considering a hybrid interphase region between reinforcement and matrix is being investigated via a multi-scale finite element approach. At the first level analysis, the proposed method uses atomistic representation of graphene to extract its elastic mechanical behavior. The output is then utilized on the second level analysis where a representative volume element (RVE) of the nanocomposite is under consideration. The RVE is considered as a three phase composite structure subjected to small strains. The reinforcing phase, i.e. graphene, is modeled in molecular detail via spring based line elements while the reinforced phase, i.e. epoxy, is modeled using continuum mechanics assumptions via the use of solid finite elements. Finally, the third and intermediate phase known as interphase is approached using solid finite elements as well as a hybrid concept. According to this concept, it is considered that the region between the two constituent materials presents anisotropy which is dependent on the distance from graphene and bounded by the surrounding mechanical properties. The influence of imperfect bonding between components is also investigated by introducing an adhesion coefficient within the adopted interphase property equations.

1. Introduction

Single-layer graphene sheets are the one atom thick two dimensional layers of carbon that are predicted to possess exceptionally high mechanical, thermal and electrical properties. With such properties, graphene sheets are considered as ideal materials for composite reinforcement. The investigation of nanomaterial properties is a very active field of research. The investigation of a matrix material reinforced with a nanomaterial such as graphene has recently attracted the attention of numerous researchers due to the exceptional properties of graphene. Evidently, computational mechanics could pose a distinct advance on the research of such a scientific field in comparison with corresponding experimental or analytical approaches since they may treat effectively nanoscale effects. At the same time, however, difficulties arise when nanoscopic and macroscopic formulation concepts should be included in a single computational model.

Plenty of experimental and theoretical studies which are related with the investigation of the mechanical properties of carbon nanotube (CNT) based composites have been performed in the last few years [1–4]. All these approaches investigate the role of CNTs mainly as reinforcing agents of polymer matrix composites. An experimental effort has recently been made to explore the effect of CNT nanoparticles on metallic alloys [5]. On the other hand, the theoretical investigation of CNT–polymer composite systems has been extensively performed via analytical [6–8] and molecular dynamics (MD) [9–11] based methods. A micromechanical formulation linking MD and continuum mechanics approaches has been recently proposed [12]. Finite element approaches has been also introduced [13–24] in the effort of exploring CNT–polymer systems. These finite element based methods may be divided in continuum mechanics studies [13–16], in which all phases are treated as continuum media, and semi-continuum mechanics studies [17–25], in which the CNTs are treated as atomistic structures. The performance of such composite material systems is critically controlled by the interphasial characteristics between the CNTs and the polymer matrix material. The adhesion among the two phases may be derived by complicated phenomena such as chemical bonding, electrostatic and van der Waal's interactions, mismatch in the coefficient of thermal expansion and radial deformation induced by atomic interactions. Some of these complicated nanoscale interphasial mechanisms have been investigated via MD [9] as well as experimental observations [10]. In these studies it has been proved that load transfer between CNTs and a polymer matrix is significant and specifically higher than that observed on most carbon fiber

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reinforced polymer composite systems. In view of the difficulty to define experimentally the interphasial bonding properties between the CNT and the surrounding polymer, several theoretical techniques have been proposed which are mainly based on computational mechanics [26]. Several researchers have used a continuum interphase of an arbitrary chosen, however, modulus of elasticity [14]. Others have utilized exclusively van der Waal's interactions to simulate CNT–polymer interphase [17–21]. Finally, some have discretely applied distance dependent stiffness variations, bounded by composite components, for that purpose [22–25]. Special interest presents a proposed approach [27,28] in which a hybrid model is developed for the interphase between a multi-walled CNT and a polymer.

In contrast with CNT based composites there are fewer studies related with graphene reinforced polymeric materials. Some experimental attempts have been made [29–32] to estimate the mechanical response of graphene–epoxy composites. More are the similar efforts related with graphene nanoplatelets [33–36], the production of which seems to be more approachable. Little has been made at a theoretical level as well. Mechanical properties of nanocomposites consisting of epoxy matrix reinforced with randomly oriented graphite platelets have been studied by the Mori–Tanaka approach in conjunction with molecular mechanics [37]. An analytical study based on nonlocal theory of elasticity has recently been introduced [38] in order to test the stability of composite laminates made of isotropic graphene layers interlaid with bonding agents. A MD based approach [39] has been proposed to characterize multilayer graphene reinforced epoxy composites. In another attempt, a molecular mechanics based approach has been developed in order to simulate interface hydrogen molecules attached to graphene and thus predict the properties of graphane [40]. The finite element method seems to be more attractive in dealing with graphene based composites due to its low computational cost derived by the fact that during modeling the exclusion of some atomistic effects may be permitted. Still, to the author's best knowledge, in the finite element approximations of the specific problem [41–43], van der Waal's interactions have been exclusively used to simulate interphasial phenomena.

In the present study, a three dimensional, linear, multi-scale, finite element model is proposed for the simulation and characterization of an epoxy material reinforced with a single-layer graphene. The graphene is treated as a molecule and therefore modeled according to its atomistic structure and interatomic force field appearing between carbon atoms. The epoxy matrix material is modeled as a continuum medium which has typical isotropic properties. In order to approximate effectively the load transfer between the two phases a practical numerical treatment is followed: interphase region is considered as a continuum medium of specific thickness while its mechanical properties are considered as hybrid meaning that are dependent and bounded by the material properties of the basic composite components, i.e. graphene and epoxy. Furthermore, it is assumed that interphasial properties vary exponentially with normal distance and that are correlated with an imperfect bonding parameter [27,28]. In order to define the interphasial property bounds originating from graphene, a numerical study is conducted beforehand in which all anisotropic elastic properties of graphene are defined. According to the proposed model, it is obvious that the anisotropy of graphene leads to a similar anisotropic behavior of interphase close to the reinforcement which, however, is reduced to isotropic near the matrix.

2. Atomistic analysis of graphene

The aim of the present work is to evaluate the stiffness properties that a graphene reinforced polymer may provide by utilizing a computationally efficient numerical scheme. Due to the linear character of these mechanical properties the adoption of nonlinear interatomic potentials was avoided. However, the home-made finite element code, which is extended here to incorporate the hybrid interphase concept, has been successfully used in the past [23] in conjunction with the modified Morse interatomic potential to predict the nonlinear stress–strain behavior of CNT–rubber composites.

The single-layer graphene, denoted as G, is analyzed in atomistic detail in a global Cartesian coordinate system (x, y, z). Therefore, the positions (x, y) of all required carbon atoms c are defined on the plane $z=0$ according to the well-known molecular structure of graphene. The distance between bonded carbon atoms is taken equal to $r=0.1421$ nm while the thickness of graphene is indirectly defined as $t^G=0.34$ nm, i.e. the interlayer distance of graphite. The whole graphene nanostructure may be divided into a series of sub-molecules $c-c-c-c$ consisted of three chemical bonds as illustrated in Fig. 1.

The interatomic force field observed within such a sub-molecule under small strains may be simulated via the use of three different types of two-noded, line, spring elements interconnecting specific carbon atom pairs. These spring elements present three degrees of freedom per node, i.e. three translations. The specific method has recently been presented in every detail and has been validated via a stability analysis concerning graphene nanoribbons [44]. The 6×6 stiffness matrix of those elements is generally expressed in the global coordinate system (x, y, z) as

$$\mathbf{K}^e=\begin{bmatrix}\mathbf{k}^e & -\mathbf{k}^e\\ -\mathbf{k}^e & \mathbf{k}^e\end{bmatrix} \tag{1}$$

where superscript e denotes the type of element. The 3×3 coefficient matrix $\mathbf{k}^e$ is diagonal while its diagonal terms are equal to the stiffness of the element along the three directions. All the utilized spring elements are analyzed in corresponding three dimensional local coordinate systems the normal of which is aligned with z global axis.

Three basic interatomic interactions, i.e. bond stretching, bond angle bending and bond angle torsion, have to be simulated. Firstly, a spring element denoted as $2c$ and analyzed in the $(\bar{x}, \bar{y}, \bar{z})$ local coordinate system (Fig. 1) is utilized for the simulation of bond stretching interaction. The $\bar{x}$-axis of the specific local coordinate system coincides with the line that connects two bonded atoms. The 3×3 coefficient stiffness matrix for the specific

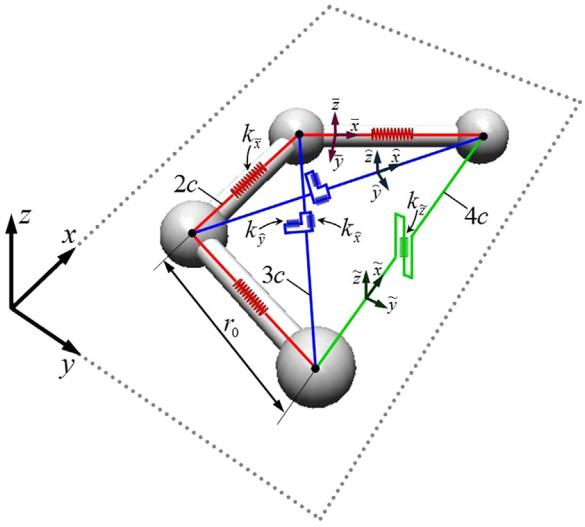

Fig. 1. Numerical formulation of interatomic interactions.