Constitutive Modeling of Nanotube-Reinforced Polymer Composite Systems

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ABSTRACT

In this study, a technique has been proposed for developing constitutive models for polymer composite systems reinforced with single-walled carbon nanotubes (SWNT). Since the polymer molecules are on the same size scale as the nanotubes, the interaction at the polymer/nanotube interface is highly dependent on the local molecular structure and bonding. At these small length scales, the lattice structures of the nanotube and polymer chains cannot be considered continuous, and the bulk mechanical properties of the SWNT/polymer composites can no longer be determined through traditional micromechanical approaches that are formulated using continuum mechanics. It is proposed herein that the nanotube, the local polymer near the nanotube, and the nanotube/polymer interface can be modeled as an effective continuum fiber using an equivalent-continuum modeling method. The effective fiber retains the local molecular structure and bonding information and serves as a means for incorporating micromechanical analyses for the prediction of bulk mechanical properties of SWNT/polymer composites with various nanotube sizes and orientations. As an example, the proposed approach is used for the constitutive modeling of two SWNT/polyethylene composite systems, one with continuous and aligned SWNT and the other with discontinuous and randomly aligned nanotubes.

KEY WORDS: nanotechnology, nanotubes, continuum mechanics, molecular mechanics, nanocomposites

1. Introduction

In the last few years, nano-structured materials have excited considerable interest in the materials research community partly due to their potentially remarkable mechanical properties. In particular, carbon nanotube-reinforced polymer composites have shown considerable promise. A Young’s modulus as high as 1 TPa and a tensile strength approaching 100 GPa has been measured for single-walled carbon nanotubes (SWNT) [1]. These properties, in addition to their relatively low density, make nanotubes an ideal candidate for polymer-composite reinforcement. In order to facilitate the development of nanotube-reinforced polymer composites, constitutive relationships must be available to predict the bulk mechanical properties of the composite as a function of molecular structure.

In recent years, many micromechanical models have been developed to predict the macroscopic behavior of composite materials reinforced with fibers that are much larger than nanotubes [2, 3]. These models assume that the fiber, matrix, and sometimes, the interface, are continuous materials and the constitutive equations for the bulk composite material are formulated based on assumptions

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of continuum mechanics. Even though many studies have focused on the fabrication and characterization of nanotube/polymer composites [4-9], a limited number of studies have addressed the applicability of micromechanics to nanotube-reinforced polymer composites. Qian et al. [5] compared the experimental and predicted values of modulus for a nanotube/polystyrene composite. The predicted modulus was estimated by using an approximation that yielded the composite modulus as a function of the physical volume fraction of the nanotubes, the geometry of the nanotubes, and the moduli of the constituent nanotubes and polymer matrix. Shaffer and Windle [4] predicted the modulus of carbon nanotube-poly(vinyl alcohol) composites using a similar approach.

In a recent example, Wise and Hinkley [10] predicted that the local change in the polymer molecular structure and the non-functionalized interface is on the same length scale as the width of a nanotube for a SWNT surrounded by polyethylene molecules. In traditional graphite fiber-reinforced composite materials the diameters of the fibers are about 1×10^4 times larger than a typical nanotube diameter. Therefore, the atomic interactions in the local polymer and at the interface between the local polymer chains and the SWNT will have a more significant impact on the bulk mechanical properties of the SWNT/polymer composites than on traditional graphite/polymer composite materials.

If a micromechanical approach is used to model the constitutive behavior of SWNT/polymer composites, then the assumptions of the model are of critical importance. An important assumption in continuum mechanics is that the densities of mass, momentum, and energy exist in the mathematical sense [11], that is, regardless of length scale. If traditional micromechanical modeling approaches are used to predict the bulk properties of SWNT/polymer composites, then the RVE must represent a material continuum. However, at the nanometer length scale the material more closely resembles an atomic lattice than a continuum. Therefore, an equivalent-continuum model of the RVE (representing the nanotube, nanotube/polymer interface, and the local polymer molecules) needs to be developed for bulk constitutive modeling of SWNT/polymer composites.

In this paper, a technique for developing constitutive models for SWNT-reinforced polymer composite materials is proposed. First, a representative volume element (RVE) of the molecular structure of the nanotube and adjacent polymer chains has been determined by using molecular dynamics (MD) simulations. Second, an equivalent-continuum model of the RVE is developed. The mechanical properties of the equivalent-continuum model are determined based on the force constants that describe the bonded and non-bonded interactions of the atoms in the RVE and reflect the local polymer and nanotube structure and the polymer/nanotube interaction. Finally, the equivalent-continuum RVE is used in subsequent micromechanical analyses to determine the bulk constitutive properties of the SWNT/polymer composite with various nanotube orientations and volume fractions.

As an example, the constitutive modeling of a SWNT/polyethylene composite is demonstrated. A MD simulation has been conducted to determine the equilibrium molecular structure of a SWNT surrounded by polyethylene molecules. An equivalent-continuum modeling technique has been used to develop a continuous RVE. Two examples of the incorporation of the RVE into micromechanical analyses for the determination of constitutive properties of continuous aligned-nanotube and random and discontinuous-nanotube composites are presented.
2. Modeling approach

The proposed approach for the constitutive modeling of SWNT/polymer composites is outlined below. The approach uses the energy terms that are found from molecular mechanics and the molecular structure determined from MD simulations. A brief description of the computational chemistry techniques that are used is given first followed by a description of the equivalent-truss and equivalent-continuum model development.

2.1 Computational chemistry

The bonded and non-bonded interactions of the atoms in a molecular structure can be quantitatively described by using molecular mechanics. The forces that exist for each bond, as a result of the relative atomic positions, are described by the force field. These forces contribute to the total vibrational potential energy of a molecular system. In general, the vibrational potential energy for a nano-structured material is described by the sum of the individual energy contributions in the RVE [12]:

\[ E = \sum_{\text{bond}} E_{\text{bond}}^{\text{bond}} + \sum_{\text{bond}} E_{\text{bond}}^{\text{angle}} + \sum_{\text{bond}} E_{\text{bond}}^{\text{torsion}} + \sum_{\text{bond}} E_{\text{bond}}^{\text{nb}} \]  

(1)

where \( E_{\text{bond}}^{\text{bond}} \), \( E_{\text{bond}}^{\text{angle}} \), and \( E_{\text{bond}}^{\text{torsion}} \), and are the energies associated with bond stretching, angle variation, and torsion, respectively, and \( E_{\text{bond}}^{\text{nb}} \) is the energy of the non-bonded interactions, which includes van der Waals and electrostatic effects (Figure 1). The individual energy contributions are summed over the total number of corresponding bonds in the molecular RVE. Various functional forms may be used for these energy terms depending on the particular material and loading conditions considered [12]. Obtaining accurate parameters for a force field often amounts to fitting a set of experimental or calculated data to the assumed functional form.

As an example, the Optimized Potential for Liquid Simulations (OPLS) united atom force field [13-15] was used for the molecular mechanics modeling of a SWNT/polyethylene composite. For simplicity, only the bond stretching, bond-angle variation, and van der Waals parameters were considered. In particular, the total vibrational potential energy of the RVE is:

\[ E_{\text{vib}} = \sum_{\text{type bond}} \sum_{\text{bond}} K_{\text{bond}}^{\text{stretch}} (\rho - P)^2 + \sum_{\text{type bond}} \sum_{\text{bond}} K_{\text{angle}}^{\text{angle}} (\theta - \Theta)^2 + \sum_{\text{type bond}} \sum_{\text{bond}} \left( \frac{K_{\text{torsion}}^{\text{torsion}}}{\rho^{12}} - \frac{1}{\rho^6} \right) \]  

(2)

where \( P \) and \( \Theta \) refer to the undeformed bond lengths and bond angles, respectively, and \( \rho \) and \( \theta \) refer to the deformed quantities. The force constants for stretching, bond-angle variation, and van der Waals interactions are \( K_{\text{bond}}^{\text{stretch}} \), \( K_{\text{angle}}^{\text{angle}} \), and \( K_{\text{torsion}}^{\text{torsion}} \), and \( K_{\text{vdW}} \), respectively. The energy terms are summed over the total number of bonds associated with each bond type (e.g. C-C, CH2-CH2, and CH2-CH3 bond stretching). The values of the force constants used for this example are [13-15]:

\[ K_{\text{C-C}}^{\text{stretch}} = 46,900 \frac{kcal}{\text{mole} \cdot \text{nm}^2} \]
\[ K_{\text{CH2-CH2}}^{\text{stretch}} = K_{\text{CH2-CH3}}^{\text{stretch}} = 26,000 \frac{kcal}{\text{mole} \cdot \text{nm}^2} \]
The equilibrium bond lengths and bond angles are:

\[ P_{c-c} = 0.142 \text{ nm} \quad P_{cH_2-cH_2} = P_{cH_2-cH_3} = 0.153 \text{ nm} \]  

\[ \Theta_{c-c-c} = 120.0^\circ \quad \Theta_{cH_2-cH_2-cH_2} = \Theta_{cH_2-cH_2-cH_3} = 112.4^\circ \]  

The MD technique has become an effective tool for studying the physics of condensed matter systems in which the forces acting on particles in a cell are calculated and the classical Newtonian equations of motion are solved numerically [16-18]. In general, each particle is allowed to interact with all the other particles in the simulation.

The molecular structure used for the analysis of the SWNT/polymer composites in this study is a representative configuration taken from an equilibrated MD run (Figure 2). The simulation was performed in a rectangular periodic box (volume \( \sim 36 \text{ nm}^3 \)) at constant temperature (300K) and pressure (1 atm) [19-21]. The cell contained one (6,6) carbon nanotube of length 49.61Å and 100 n-decane chains. The simulation was run for 800 pico-seconds with a time step of 1 femto-second. To avoid biasing the final structure of the system, the run was started at very low density (large periodic cell size) and allowed to reach the prescribed internal pressure (1 atm) during an initial equilibration phase. This approach allows the n-decane matrix to equilibrate into a favorable configuration.

The structure of the nanotube was held fixed during the simulation and interacted with the n-decane molecules through a non-bonded van der Waals potential. The n-decane chains were fully flexible and were modeled by using the OPLS united atom parameters [13-15]. This force field combines the hydrogen atoms with the carbon atom to which they are bound to form one larger effective (or united) atom. This approach greatly reduces the computational work required per time step, allowing much longer simulations to be performed than would be possible with an explicit or all-
atom force field. This approximation is justified in this case because the phenomenon of interest, the ordering of the matrix around the nanotube, occurs on a much longer timescale than the vibrational period of the carbon–hydrogen bonds. The united atom model allows for the effective incorporation of these vibrations by adjusting the size, mass, and interaction parameters of the aggregate repetitive unit.

2.2 Equivalent-continuum modeling

The equivalent-continuum modeling of the RVE consists of two main steps: the development of an equivalent truss model that is based on the molecular model, and the development of the equivalent-continuum model that is based on the truss model.

2.2.1 Background

Odegard et al. [22] developed a method of modeling the bulk mechanical behavior of nanostructured materials that uses an equivalent-continuum model. The method consists of three major steps. First, a suitable RVE of the nano-structured material is chosen. The RVE of a typical nano-structured material is on the nanometer length scale, therefore, the material of the RVE is not continuous, but is an assemblage of many atoms. Interaction of these atoms is described in terms of molecular mechanics force constants, which are known for most atomic structures [12]. Second, an equivalent truss model of the RVE is developed in which each truss element represented an atomic bonded or non-bonded interaction. The moduli of the truss elements is based on the molecular mechanics force constants that describe the contribution of each bonded or non-bonded interaction to the total vibrational potential energy. Therefore, the total vibrational potential energy of the molecular model and the strain energy of the truss are equivalent under the same loading conditions. Third, an equivalent-continuum model of the RVE is developed in which the total strain energy in both truss and continuum models, under identical loading conditions, is set equivalent. The effective mechanical properties or the effective geometry of the equivalent-continuum is then determined from equating strain energies. Therefore, a continuum element was determined that has a mechanical behavior representing the behavior of the nano-structured material, which was in-turn, based on the molecular mechanics force constants.

As an example of this modeling approach, Odegard et al. [22] developed an equivalent-continuum model for a graphene sheet (Figure 3). A RVE of the graphene lattice was subjected to a set of fundamental loading conditions, and it was determined that the effective thickness of a plate that represented the mechanical behavior of the graphene sheet was 0.28 nm and 0.24 nm for in-plane tension and shear, respectively. It was concluded that a wall thickness of 0.28 nm should be assumed in the determination of the Young’s modulus for SWNT from experiment or computations, instead of 0.34 nm, which is the inter-plane spacing of graphite.

2.2.2 Truss model

In traditional molecular models, the atomic lattice has been viewed as an assemblage of masses that are held in place with atomic forces that resemble elastic springs [23]. The mechanical analogy of this is a pin-jointed truss model in which each truss member represents either a bonded or non-bonded interaction between atoms. Therefore, the truss model allows the mechanical behavior of
the nano-structured system to be accurately modeled in terms of displacements of the atoms. The
deposition of each bonded or non-bonded interaction corresponds to the axial deformation of the
corresponding truss element. This mechanical representation of the lattice behavior serves as an
intermediate step between linking the molecular model with an equivalent-continuum model.

The total mechanical strain energy, $\Lambda^t$, of a truss model is expressed in the form:

$$\Lambda^t = \sum_{\text{type}} \sum_{\text{rod}} \frac{AY}{2R} (r - R)^2$$

where $A$, $Y$, $R$, and $r$ are the cross-sectional area, Young’s modulus, undeformed axial length, and
deformed axial length of each truss element, respectively. The term $(r-R)$ is the axial deformation of
the rod under consideration. The summations occur for each individual rod of each truss element
type in the RVE.

In order to represent the mechanical behavior with the truss model, equation (6) must be equated
with equation (2) in a physically meaningful manner. Each of the two equations is the sum of
energies for particular degrees of freedom. The main difficulty in the substitution is specifying
equation (6), which has stretching terms only, for equation (2), which also has bond-angle variance
and van der Waals terms.

Odegard et al. [22] showed that for small deformations, the Young’s moduli of the rods
representing primary bonds and the bond-angle variance interactions may be determined as a
function of the force constants:

$$Y^a = \frac{2K^a R^a}{A^a}$$

$$Y^b = \frac{3K^b}{2R^b A^b}$$

where the superscripts $a$ and $b$ are associated with primary bonding and bond-angle variance
interactions, respectively. The Young’s modulus of the truss element that represents van der Waals
bonding is more difficult to obtain. The simplest approach was to fit the change in the vibrational
potential energy of a van der Waals bond approximated with an appropriate function (such as the
Lennard-Jones equation [24, 25]) with the corresponding strain energy of a truss element given by
equation (6). The modulus of the truss element was adjusted to minimize the sum of the squares of
the errors between the van der Waals function and equation (6), for a limited range of atomic
distances. This was a simple approximation that increased in accuracy for smaller mechanical
deformations. For example, the Young’s moduli for the truss elements representing bond
stretching, variation in bonding angle, and van der Waals bonds for the SWNT/polyethylene
composite are:

$$Y^a_{C-H} = 295 \text{ TPa} \quad Y^a_{C=C} = 6 \text{ TPa} \quad Y^a_{C=C} = 176 \text{ TPa}$$
\[ Y_{c-c}^b = 9 \text{TPa} \quad Y_{c-H_2-C-H_2}^b = Y_{c-H_2-C-H_3}^b = 8 \text{TPa} \]

\[ Y_{c-CH_3}^c = Y_{c-CH_3}^c = 200 \text{MPa} \quad Y_{c-H_2-C-H_2}^c = Y_{c-H_2-C-H_3}^c = 500 \text{MPa} \]

where the superscript \(c\) denotes van der Waals bonding. The values are obtained by using the force constants in equation (3), by using equations (7) and (8), by using the approach described above for the van der Waals bonding, and by assuming an elastic rod radius of 0.01 nm. The finite element truss model for the RVE is shown Figure 2. The finite element model was generated using ANSYS® 5.7 [26]. The different shades of the truss elements correspond to different values of Young’s modulus.

### 2.2.3 Continuum model

The geometry of the equivalent-continuum RVE was assumed to be cylindrical, similar to that of the truss model (Figure 2). With this approach, the mechanical properties of the solid cylinder are determined by equating the total strain energies of the equivalent truss and equivalent-continuum models under identical loading conditions. The entire set of elastic constants for a cylindrical and homogeneous material is determined from several fundamental loading conditions [27]. Once the mechanical properties of the equivalent-continuum model of the RVE is determined, then it is used in micromechanical analyses as an effective fiber.

### 2.3 Micromechanical methods

Once the equivalent-continuum RVE is established, constitutive models of SWNT/polymer composites is developed with a micromechanical analysis using the mechanical properties of the effective fiber and the bulk matrix material. While the polymer molecules in close proximity to the SWNT (Figure 2) are included in the effective fiber, it is assumed that the remaining polymer has mechanical properties equal to those of the bulk resin. Since the bulk polymer molecules and these local polymer molecules included in the effective fiber are physically entangled, then perfect bonding between the effective fiber and matrix may be assumed.

Numerous modeling approaches have been proposed for constitutive modeling of composite materials based on the mechanical properties of the constituent materials [2, 3]. The elastic, bulk composite behavior is described by:

\[ \{\sigma\} = [C]\{\varepsilon\} \]

where \(\{\sigma\}\) and \(\{\varepsilon\}\) are column vectors that contain the components of stress and strain tensors for the material, respectively, and \([C]\) is the stiffness matrix. The components of the stiffness matrix are dependent on the properties, concentrations, orientation, and interaction of the constituents, and are quantitatively determined by using micromechanical analyses.

As an example, the constitutive behavior of a SWNT/polymer composite with continuous and aligned nanotubes is modeled as shown in Figure 4. Using the concentration and mechanical properties of the effective fiber and the bulk matrix, the stiffness matrix components are determined.
by using analytical or numerical techniques [2]. Similarly, the constitutive behavior of a SWNT/polymer composite with discontinuous and randomly aligned nanotubes is also modeled as shown in Figure 4. The information concerning the concentration and mechanical properties of the constituents, as well as the alignment of the effective fibers, are used in a micromechanical analysis for constitutive model development [3].

3. Summary

Carbon nanotube-reinforced polymer composites have generated excitement in the materials research community partly due to the potentially remarkable mechanical properties that can be achieved with a highly stiff reinforcement embedded in a tough, light-weight polymer matrix. In order to facilitate the development of nanotube-reinforced polymer composites, constitutive relationships must be available to predict the bulk mechanical properties of the composite as a function of molecular structure. Since the polymer molecules are on the same size scale as the nanotubes, the interaction at the polymer/nanotube interface is highly dependent on the local molecular structure and bonding. At these small length scales, the lattice structures of the nanotube and polymer chains cannot be considered continuous, and the bulk mechanical properties of the SWNT/polymer composites can no longer be determined through traditional micromechanical approaches that are formulated using continuum mechanics.

In this study, a technique has been proposed for developing constitutive models for polymer composite systems reinforced with single-walled carbon nanotubes (SWNT). It is proposed herein that the nanotube, the local polymer near the nanotube, and the nanotube/polymer interface can be modeled as an effective continuum fiber using an equivalent-continuum modeling method. The effective fiber retains the local molecular structure and bonding information and serves as a means for incorporating micromechanical analyses for the prediction of bulk mechanical properties of SWNT/polymer composites with various nanotube sizes and orientations.

As an example, the constitutive modeling of a SWNT/polyethylene composite has been demonstrated. A MD simulation has been conducted to determine the equilibrium molecular structure of a SWNT surrounded by polyethylene molecules. An equivalent-continuum modeling technique has been used to develop a continuous RVE. Two examples of the incorporation of the RVE into micromechanical analyses for the determination of constitutive properties of continuous aligned-nanotube and random and discontinuous-nanotube composites have been developed.

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References


Figure 1 – Molecular mechanics modeling

Figure 2 – Equivalent-continuum modeling of nanotube and local polymer molecules.
Figure 3 – Equivalent-continuum modeling of a graphene sheet.

Note: the effective fibers are the equivalent-continuum RVEs.

Figure 4 – SWNT/polymer composite materials
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### Abstract
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