Composites: Part B 68 (2015) 446–464

Contents lists available at ScienceDirect

Composites: Part B

journal homepage: www.elsevier.com/locate/compositesb

Uncertainty quantification for multiscale modeling of polymer nanocomposites with correlated parameters

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ARTICLE INFO

Article history:
Received 8 July 2014
Received in revised form 25 July 2014
Accepted 13 September 2014
Available online 22 September 2014

Keywords:
A. Polymer–matrix composites (PMCs)
B. Mechanical properties
C. Computational modeling
C. Micro-mechanics
Multiscale modeling

ABSTRACT

We propose a stochastic multiscale method to quantify the correlated key-input parameters influencing the mechanical properties of polymer nanocomposites (PNCs). The variations of parameters at nano-, micro-, meso- and macro-scales are connected by a hierarchical multiscale approach. The first-order and total-effect sensitivity indices are determined first. The input parameters include the single-walled carbon nanotube (SWNT) length, the SWNT waviness, the agglomeration and volume fraction of SWNTs. Stochastic methods consistently predict that the key parameters for the Young's modulus of the composite are the volume fraction followed by the averaged longitudinal modulus of equivalent fiber (EF), the SWNT length, and the averaged transverse modulus of the EF, respectively. The averaged longitudinal modulus of the EF is estimated to be the most important parameter with respect to the Poisson's ratio followed by the volume fraction, the SWNT length, and the averaged transverse modulus of the EF, respectively. On the other hand, the agglomeration parameters have insignificant effect on both Young's modulus and Poisson's ratio compared to other parameters. The sensitivity analysis (SA) also reveals the correlation between the input parameters and its effect on the mechanical properties.

1. Introduction

Carbon nanotubes (CNTs) are ideal reinforcement materials for polymeric composites due to their superior mechanical and electrical properties as well as their light weight [1–3]. Substantial progress obtained from experiment studies were recently summarized by Thostenson et al. [4]. However, there remains a great challenge in the structural characterization and fabrication of polymer nanocomposites (PNCs) at nano-scales [5]. Analytical and numerical simulations have been employed to assist the design of PNCs, and *a priori* prediction of their mechanical properties. Numerous micromechanical models have been developed to predict the mechanical properties of PNCs. As have been shown by Odegard and co-workers [6], the direct use of continuum micromechanical models might be inadequate to predict the mechanical properties of PNCs since the micromechanical models do not consider the lattice structure of the CNT but simplify it as a solid fiber. Furthermore, a continuum micromechanical model often assumes perfect bonding between the fibers and the matrix. Hence, it cannot capture the effect from the interphase between the CNT and the polymer matrix that is dominated by van der Waals (vdW) interactions [7]. In order to address these issues, Shokrieh and Rafiee [8,9] developed a transversely isotropic equivalent fiber (EF).

Due to the high aspect ratio and low bending stiffness of the CNTs, they normally exist in non-straight shape [10,11]. As shown by micrograph images [12], the embedded CNTs exhibit significant curvature. Several studies [12–14] have been carried out to investigate the influence of CNT curvature on the effective modulus of PNCs. Often a nanotube was substituted by a solid continuum fiber. Thus, the effect of the fiber waviness on their mechanical properties was studied instead of that of the CNT waviness. The results showed that the effective modulus of PNCs can be significantly degraded with an increasing fiber waviness. Most of previous researchers limited their studies to a sinusoidal waviness pattern that may improperly estimate the effects of the fiber waviness on the mechanical properties. This issue will be addressed in the numerical results and discussion Section 6. Since the CNTs are concentrated in local regions due to van der Waals (vdW) interactions between CNTs and also due to the non-uniform dispersion of the

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http://dx.doi.org/10.1016/j.compositesb.2014.09.008
1359-8368/

CNTs during processing, local bundles of CNTs within the polymer matrix are formed. The PNCs were modeled as random heterogeneous media and homogenization models have been applied at the lower scale to determine their local material properties. Shi et al. [15] developed a two-parameter approach in combination with the Mori–Tanaka model to determine the influence of the agglomeration of CNTs on the mechanical properties of PNCs.

Several approaches have been developed to predict the mechanical properties of PNCs. However, the predicted mechanical properties were found significantly deviating from the measured values in experiments. The discrepancy was later discussed and explained by multiscale Monte-Carlo finite element simulations [16]. However, in their stochastic multiscale model, only the volume fraction was considered as a random parameter whereas other parameters such as the CNT length, the orientation, the agglomeration, the waviness, and dispersion of CNTs were considered deterministic. A stochastic multiscale approach was also used by [17] to predict mechanical properties of PNCs. This study allowed realizing the scale difference between micro and nano that was not modeled for PNCs on micro-level [18–20].

This paper presents a comprehensive stochastic multiscale method to quantitatively determine the influence of many input parameters on the mechanical properties of PNCs. Sensitivity analysis (SA) methods are developed from the classical Sobol's estimator [21] to quantify the influences of the correlated (dependent) input parameters on the Young's modulus and Poisson's ratio of PNCs. Six important parameters at the corresponding scales are selected: the single-walled carbon nanotube (SWNT) length (L_{SWNT}), the averaged longitudinal modulus (E_{LEF}) and transverse modulus of EF (E_{TEF}) being representative of the waviness parameters, the agglomeration parameters (ξ, ζ) and the volume fraction (V_f). To reduce computational cost, surrogate models are constructed to approximate the mechanical model and the SA was performed on the surrogate models.

The content of the paper is outlined as follows. In the next section, we briefly describe the multiscale model. Section 3 presents the complete statistical distribution for each input parameter. Different SA methods are described in Section 4. The surrogate model is presented in Section 5. The coefficients of the surrogate model and sensitivity indices will be shown in Section 6 before the discussion on the numerical results. Finally, we close the manuscript with concluding remarks.

2. Multiscale model

In this section, we briefly describe the bottom-up hierarchical multiscale method for PNCs connecting all scales of the nano-, micro-, meso- and macro-scales, see Fig. 1. The stochastic multiscale approach is employed to consider the effective parameters of different scales as uncertainties. In the nano-scale, material properties and the structure of the SWNT are considered. In the micro-scale, the SWNT embedded in the polymer matrix in the presence of the interphase is modeled and replaced by an EF. The SWNT waviness, the agglomeration and the SWNT orientation are taken into account in the meso-scale. In the macro-scale, we consider the volume fraction as an uncertain variable.

2.1. Nano-scale model

The investigated unit-cell at nano-scale consists of an armchair SWNT (10,10) in a polymer matrix. The SWNT is modeled as a space-frame structure. The C–C covalent bonds in the frame-like structure are represented by 3D beam elements. Nodes representing carbon atoms are used to connect the beam elements to form the SWNT structure.

For covalent systems, the total energy can be obtained as the sum of energies consisting of bonded and non-bonded interactions [22]

$$U = \sum V_r + \sum V_\theta + \sum V_\phi + \sum V_{nb}. \tag{1}$$

where V_r, V_θ and V_ϕ are the energy associated with bond stretching, bond-angle bending, torsion (dihedral angle and out-of-plane), respectively; V_{nb} is the energy due to non-bonded van der Waals interactions, see also Fig. 2.

The potential energy is approximated by simple harmonic potential, see [24], as:

$$V_r = \frac{1}{2}k_r(r - r_0)^2 = \frac{1}{2}k_r(\Delta r)^2, \tag{2a}$$

$$V_\theta = \frac{1}{2}k_\theta(\theta - \theta_0)^2 = \frac{1}{2}k_\theta(\Delta\theta)^2, \tag{2b}$$

$$V_\phi = \frac{1}{2}k_\phi(\Delta\phi)^2, \tag{2c}$$

where k_r, k_θ and k_ϕ are the bond stretching force constant, bond angle bending force constant and torsional resistance, respectively; $\Delta r, \Delta\theta$ and $\Delta\phi$ denote the bond stretching deformation, the bond angle bending and the angle change of bond twisting, respectively.

Based on structural mechanics, the total strain energy of a uniform beam with length L, cross-sectional area A and moment of inertia I, subjected to axial load P, bending moment M and twisting moment T, respectively, is given by

$$U = \int_0^L \frac{1}{2}\frac{EA}{(\Delta L)^2}dx + \int_0^L \frac{1}{2}\frac{EI}{(2\alpha)^2}dx + \int_0^L \frac{1}{2}\frac{GJ}{(\Delta\beta)^2}dx, \tag{3}$$

with $\Delta L, \alpha$ and $\Delta\beta$ being the corresponding axial stretching deformation, the bending angle and the twist angle.

Fig. 2 illustrates the equivalence of molecular mechanics (molecular forces) and structural mechanics (beam elements) for bonded interactions. It can be shown [23] that the parameters of the beam model (EA, EI and GJ) are related to the molecular mechanics constants (k_r, k_θ and k_ϕ) by

$$\frac{EA}{L} = k_r, \quad \frac{EI}{L} = k_\theta, \quad \frac{GJ}{L} = k_\phi, \tag{4}$$

where k_r, k_θ and k_ϕ are the bond stretching force constant, bond angle bending force constant and torsional resistance respectively. The C–C bond length L is selected as $0.1421nm$ [25,26].

When the force constants k_r, k_θ, and k_ϕ are known, the sectional stiffness parameters $EA; EI$, and GJ can be obtained. As suggested by [17,23], $k_r = 6.52 \times 10^{-7}$ N/nm, $k_\theta = 8.76 \times 10^{-10}$ N nm/rad^2, $k_\phi = 2.78 \times 10^{-10}$ N nm/rad^2 are adopted in this paper.

A non-bonded vdW force of the truncated Lennard–Jones (LJ) 6–12 with $\epsilon = 0.4492$ kJ/mol, $\sigma = 0.3825$ nm and $r = 0.85$ nm [9,27] is used to describe the interphase between the SWNT and the polymer matrix. The interphase thickness is chosen the same as half of thickness of SWNT (0.34 nm). The vdW force field is expressed as:

$$F_{vdw} = 24\frac{\epsilon}{\sigma}\left[2\left(\frac{\sigma}{r}\right)^{13} - \left(\frac{\sigma}{r}\right)^{7}\right] \tag{5}$$

As presented by Shokrieh and Rafiee [9], the surrounding polymer matrix was simulated as continuum medium and modeled by isotropic solid elements with Young's modulus of 10 GPa and Poisson's ratio of 0.3.

2.2. Micro-scale model

The micro-scale model is considered as a SWNT embedded in a polymer matrix modeled by the continuum mechanics approach [23]. The 20-node hexahedral elements are used to model the polymer matrix. 3-D nonlinear springs connecting the C atom on