Composite Structures 134 (2015) 981–988

Contents lists available at ScienceDirect

Composite Structures

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

Tensile fracture behavior of short carbon nanotube reinforced polymer composites: A coarse-grained model

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ARTICLE INFO

Article history:
Available online 7 September 2015

Keywords:
Polymer-matrix composites
Carbon nanotube
Tensile fracture
Coarse-grained model

ABSTRACT

Short-fiber-reinforced polymer composites are increasingly used in engineering applications and industrial products owing to their unique combination of superior mechanical properties, and relatively easy and low-cost manufacturing process. The mechanical behavior of short carbon nanotube (CNT) polymer composites, however, remains poorly understood due to size and time limitations of experiments and atomistic simulations. To address this issue, the tensile fracture behavior of short CNT reinforced poly (methyl methacrylate) (PMMA) matrix composites is investigated using a coarse-grained (CG) model. The reliability of the CG model is demonstrated by reproducing experimental results on the strain–stress behavior of the polymer material. The effect of the nanotube weight fraction on the mechanical properties, i.e. the Young's modulus, yield strength, tensile strength and critical strain, of the CNT/polymer composites is studied in detail. The dependence of the mechanical properties of the composites on the orientation and length-to-diameter aspect ratio of nanotube reinforcements is also examined.

1. Introduction

Short-fiber-reinforced polymers (SFRPs) are a developing class of composite materials, which have attracted intense attention due to their outstanding mechanical, thermal and electrical properties [1,2]. The mechanical properties of SFRP composites can reach stiffness levels attainable with continuous fibers, while the ability of unreinforced polymers to be formed into complex shapes is also preserved [3]. In addition, the production process for short fiber composites is more economical than continuous fiber composites [4]. This compromise between cost and performance makes short fiber polymer composites as excellent alternatives in electronic, automotive, oilfield and chemical industries [5]. Among different types of fibers used in the composites, the outstanding mechanical properties of carbon nanotubes (CNTs) promise ultra-high-strength reinforcements in high-performance polymer matrix composites [6].

In order to develop SFRPs, a quantitative understanding of their mechanical properties is of great significance since the mechanical performance of the materials strongly depends on controllable parameters such as fiber length and orientation [7]. Up to now, a number of experimental studies have been conducted in the literature on the mechanical behavior of reinforced polymer composites [8–12]. The experimental investigations, however, provide insufficient insight into molecular scale processes such as the interfacial interactions between nanotubes and matrix. The main reason for the drawback is rooted in the limited resolution of experimental techniques at the nanoscale. Furthermore, experimental efforts frequently encounter difficulties in fabricating SFRPs with uniformly distributed fibers with desired sizes.

Molecular dynamics (MD) simulations, in contrast, provide detailed information of phenomena at the nanoscale such as stick–slip mechanisms and the interfacial interactions between matrix and reinforcements [13–16]. MD simulations also facilitate the interpretation of experimental data, and additionally open a route to new designs of nanocomposites. Therefore, molecular simulations are necessary in the understanding of mechanical behavior of reinforced polymer nanocomposites. Zhu et al. [17] studied the elastic properties of an epoxy Epon 862 matrix with a size of $4.028 \times 4.028 \times 6.109\ \text{nm}^3$ reinforced by short (10,10) CNTs with

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http://dx.doi.org/10.1016/j.compstruct.2015.09.001
0263-8223/

length-to-diameter aspect ratios of 2.15 and 4.5. They reported that short CNT fibers increase the Young's modulus of the polymer matrix up to 20% compared to the pure Epon 862 matrix. Molecular simulation studies on the elastic properties of short single-walled CNT (SWCNT) reinforced Poly (vinylidene fluoride) (PVDF) matrix composites [18] showed that a (5,5) SWCNT with a length of 2 nm can increase the Young's modulus of a CNT/PVDF unit cell by 1 GPa. The simulation unit cell consists of a (5,5) SWCNT with a volume fraction of 1.6% embedded in 60 PVDF chains. Arash et al. [19] investigated the mechanical behavior of CNT/poly (methyl methacrylate) (PMMA) polymer composites under tension. They proposed a method for evaluating the elastic properties of the interfacial region of CNT/polymer composites. Their simulation results on the elastic properties of a PMMA polymer matrix with a size of $3.7 \times 3.7 \times 8$ nm^3 reinforced by a short (5,5) SWCNT reveal that the Young's modulus of the composite increases from 3.9 to 6.85 GPa with an increase in the length-to-diameter aspect ratio of the nanotube from 7.23 to 22.05.

Although molecular simulations have been broadly used in modeling reinforced polymer nanocomposites, the massive computational effort required by the simulations strictly limits their applicability to small molecular systems over a limited time scale. These drawbacks hinder MD simulations to study the effect of fiber sizes and orientations on the mechanical behavior of reinforced polymer nanocomposites. In order to overcome these limitations, coarse-grained (CG) models beyond the capacity of molecular simulations have been developed in the literature [20–22]. The principle of CG models is to map a set of atoms to a CG bead, which reduces the atomistic degrees of freedom, resulting in substantial increases in the accessible time and length-scales while partially retaining the molecular details of an atomistic system. Up to now, many CG models have been developed for polymer materials [21,23,24]. Recently, the reliability of these approaches in modeling graphenes and CNTs has been also examined [25–28]. Ruiz et al. [27] established a CG model for the elastic and fracture behavior of graphenes with a ~200 fold increase in computational speed compared to atomistic simulations. Zhao et al. [28] calibrated parameters of the CG stretching, bending and torsion potentials for SWCNTs to study their static and dynamic behaviors. They also derived parameters of non-bonded van der Waals (vdW) interactions between CNTs in a bundle. The CG model was shown to have great potential in the analysis of the mechanical properties of CNT bundles with low computational cost compared to atomistic simulations. Arash et al. [29] developed a comprehensive CG model of CNT reinforced polymer composites capturing the non-bonded interactions between polymer chains and nanotubes. They then employed the model to study the elastic properties of short and long CNT reinforced PMMA polymer composites. Despite the simulation studies on the elastic properties of short CNT reinforced polymer composites, there is still no simulation investigation on the fracture behavior of a polymer matrix reinforced by randomly distributed short CNTs. Furthermore, the effects of nanotubes length and orientation on the mechanical properties of short CNT/polymer composites at large deformations have not been quantified. Hence, a quantitative study on the mechanical properties of short CNT/polymer composites is essential to achieve a successful design, synthesis, and characterization of the nanocomposites.

In this study, the mechanical behavior of short CNT/PMMA composites under tension is investigated in elastic and plastic regimes using a CG model. The applicability of the CG model in predicting the strain–stress behavior of PMMA polymer is examined using experimental results reported in the literature. The effects of the weight fraction, orientation and length-to-diameter aspect ratio of unidirectional and randomly distributed CNT reinforcements on the mechanical properties of the nanocomposites are studied in detail. The mechanical behavior of the CNT/PMMA composites observed in the CG simulations is also interpreted using a micromechanical continuum model.

2. Methods

In this study, we used a CG model that was previously developed and examined for modeling CNT/PMMA polymer composites [29]. In this model, each methyl methacrylate ($C_5O_2H_8$) monomer is treated as a bead with an atomic mass of 100.12 amu as illustrated in Fig. 1(a). The center of the bead is chosen to be the center of mass of the monomer. The pseudoatom is defined as P bead, which enables a 15-fold decrease in the number of degrees of freedom (DOF) of a polymer chain compared to its corresponding full atomistic system. The CG model was also developed for modeling (5,5) CNTs, where each five atomic rings are mapped into a CG bead with an atomic mass of 600.55 amu (see Fig. 1(b)). The bead is defined as a C bead, which decreases the number of DOF of a nanotube 50-fold with respect to full atomistic simulations.

The CG force field is decomposed into bonded and non-bonded potential functions. The total potential energy, E_{total}, of a system is therefore written as the sum of energy terms associated with the variation of the bond length, E_b, the bond angle, E_a, the dihedral angle, E_d, the vdW interactions, E_{vdW}, and the constant free energy of the system, U_0, as $U_{total} = \sum_i E_{b_i} + \sum_j E_{a_j} + \sum_k E_{d_k} + \sum_{lm} E_{vdW_{lm}} + U_0$. The functional forms of the contributing terms for a single interaction are as follows:

$$E_b(d) = \frac{k_d}{2}(d - d_0)^2 \quad \text{for } d < d_{cut}, \tag{1a}$$

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

$$E_d(\phi) = \frac{k_\phi}{2}[1 + \cos 2\phi], \tag{1c}$$

$$E_{vdW}(r) = D_0\left[\left(\frac{r_0}{r}\right)^{12} - 2\left(\frac{r_0}{r}\right)^{6}\right], \tag{1d}$$

where k_d and d_0 are the spring constant of the bond length and the equilibrium bond distance, respectively; k_θ and θ_0 are respectively the spring constant of the bond angle and the equilibrium bond angle; k_ϕ and ϕ are the spring constant of the dihedral angle and the dihedral angle, respectively. D_0 and r_0 are the Lennard–Jones parameters associated with the equilibrium well depth and the equilibrium distance, respectively. A potential cutoff of 1.25 nm is used in calculation of vdW interactions. The parameters of the force field are listed in Table 1. The following simulations were performed using Accelrys Materials Studio 7.0.

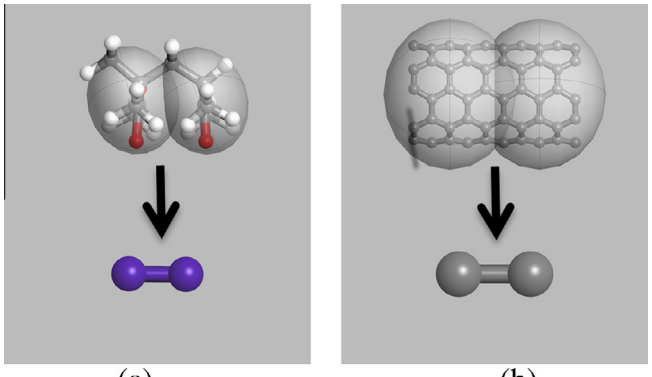

Fig. 1. (a) Two monomers of a PMMA polymer chain and its CG model made of two P beads, and (b) a (5,5) CNT with 10 rings of carbon atoms and its CG model made of two C beads.