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Interfacial and mechanical properties of epoxy nanocomposites using different multiscale modeling schemes

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ABSTRACT

In this study, we investigate the interfacial and mechanical properties of carbon nanotube (CNT) reinforced epoxy composite. The work carried out in two stages. In the first, we conducted molecular dynamics (MD) simulations to determine the atomic-level interfacial and mechanical properties of the transversely isotropic representative volume element (RVE) comprised of CNT-epoxy composite. In the second, the Mori–Tanaka micromechanics scheme was used to scale up the mechanical properties of the atomic structure to the microscale level. The work was further extended and used atomistic-based continuum (ABC) multiscale modeling technique, which makes use of constitutive relations derived solely from interatomic potentials to model the same system. Interestingly, the results of our comparative investigation reveals that (i) the ABC technique and MD simulation provide almost identical predictions for the atomic-level interfacial and mechanical properties of the nanocomposite, (ii) both models predict comparable bulk mechanical properties of the nanocomposite containing randomly dispersed CNTs, and (iii) they also reveal that a higher degree of orthotropy of the nanoscale representative fiber significantly influences the bulk mechanical properties of the nanocomposite.

1. Introduction

The remarkable physical and thermomechanical properties of CNTs, such as low density [1], high aspect ratio [2], high Young's modulus [3–6], high thermal conductivity [7–9], low coefficient of thermal expansion [10,11], high strength [12–14] and large fracture strain [13,14] have made them excellent reinforcements for multifunctional composites. It is found that few weight percentages of CNTs can significantly improve the interfacial and mechanical properties of CNT-based composites.

Several experimental studies have been carried out to investigate the interfacial characteristics of CNT-reinforced composites. For instance, Wagner et al. [15] estimated the interfacial shear stress between the multi-walled CNTs and the polymer based on the fragmentation test to be as high as 500 MPa, which is more than one order of magnitude compared with conventional composites. Micro-Raman spectroscopy was used by Ajayan et al. [16] to measure the local mechanical behavior of single-walled CNT bundles in an epoxy nanocomposite. They noticed that the efficiency of stress transfer and hence the enhancement of the mechanical properties is lower than expected due to sliding of the CNTs in the agglomerated bundles. Qian et al. [2] investigated the load transfer in multi-walled CNT–polystyrene composites and reported that addition of 1 wt% of CNTs increases the tensile modulus and strength by ~39% and 25%, respectively. Schadler et al. [17] studied the interfacial characteristics of multi-walled CNT-reinforced composites with both the tension and compression loadings. They reported that the compression modulus is higher than that of the tensile modulus, indicating that the load transfer to CNTs from the matrix is higher in compression. Cooper et al. [18] used scanning probe microscope tip to pull-out individual single- and multi-walled CNTs ropes from epoxy matrix. The ISS of both cases was found to be in the range of 35 to 376 MPa. This relatively high value of ISS was attributed to the formation of a strong ultrathin epoxy layer at the interface. This layer exits as a result of the formation of covalent bonds between CNTs and the surrounding polymer molecules, which originate from the presence of defects in the CNTs.

A significant number of analytical and numerical studies have also been conducted to investigate the interfacial properties of CNT-based composites. For example, Lordi and Yao [19] used force-field based molecular mechanics calculations to determine the binding energies and sliding frictional stresses between CNTs and a range of polymer substrates, in an effort to understand the factors governing interfacial strength. They reported that binding

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energies and frictional forces play only a minor role in determining the strength of the interface, but that helical polymer conformations around the CNT are essential in developing high ISS. Liao and Li [20] studied the interfacial characteristics of a CNT-reinforced polystyrene composite system through molecular mechanics simulations and elasticity calculations. They reported that the CNT-matrix bonding arises from non-bonded electrostatic and van der Waals (vdW) interactions, deformation induced by these interactions, and mismatch in the coefficients of thermal expansion. Their CNT pull-out simulation results suggest that the interfacial shear stress of the CNT-polystyrene system is about 160 MPa; significantly higher than conventional carbon fiber composite systems. Frankland et al. [21] generated stress–strain curves of polyethylene nanocomposite reinforced with long and short CNTs using MD simulations. Both nanocomposites were mechanically loaded in the axial and the transverse directions of the CNT axis. In their study, nanocomposite reinforced with long CNTs showed an increase in the stiffness relative to the polymer. On the other hand, nanocomposites reinforced with short CNTs showed no enhancement relative to the polymer. Xiao and Zhang [22] studied the effects of CNT length and diameter on the distributions of the tensile stress and interfacial shear stress of a CNT in an epoxy matrix. Their work revealed that a smaller CNT diameter has a more effective reinforcement effect and that there exists an optimal tube length at which reinforcement is maximized. They also reported that a CNT has a greater stress transfer efficiency than a solid fiber, and provides toughness and tensile strength to the resulting nanocomposite. An analytical model has been developed by Haque and Ramasetty [23] to study the axial stress and shear stress at the interface of CNT-reinforced polymer composite materials. An expression for the effective length of the CNT has also been established by them for studying the load transfer efficiency in CNT-reinforced composites. A micromechanics model has also been developed by Li and Saigal [24] for assessing the interfacial shear stress transfer in CNT-reinforced polymer composites. Their results indicate that the stress transfer characteristics of nanocomposites can be improved by using sufficiently long CNTs. The load transfer efficiency in the CNT-reinforced nanocomposites was investigated by Tsai and Lu [25] using the conventional shear lag model and the finite element analysis. They revealed that the load transfer efficiency increases with the increment of the aspect ratio of CNTs. Using finite element model, Shokrieh and Rafiee [26] studied the tensile behavior of embedded short CNTs in the polymer matrix in presence of vdW interactions in interphase region. They modeled the interphase using non-linear spring elements capturing the force-distance curve of vdW interactions. They observed that improvement in the Young's modulus of CNT-reinforced composite is negligible for lengths smaller than 100 nm and saturation takes place in larger lengths on the order of 10 μm. Recently, Kundalwal et al. [27] developed a three-phase shear lag model to analyze the stress transfer characteristics of a hybrid hierarchical CNT-reinforced composite. In the first stage of their model, the transversely isotropic properties of CNT-reinforced polymer composite were obtained and subsequently, they were used as inputs to shear lag model development. A significant number of studies have also been carried out to determine the bulk mechanical properties of CNT-based composites. These include experimental measurements (see [28] and references therein), continuum and micromechanical analysis [29–37], multiscale approaches [38–41], and MD simulations [42–45].

2. Multiscale modeling

Evidently, a number of experimental, analytical and numerical works has been reported in the literature to investigate the interfacial and mechanical properties of CNT-based composites. Among all these, we consider the multiscale modeling technique is more appropriate to property evaluation of CNT-based composites. It is, therefore, our objective to develop a multiscale model capable of characterizing both the interfacial and mechanical properties of CNT-reinforced epoxy composite. This task was performed in two steps. In the first, MD simulations were used to study the interfacial and the transversely isotropic mechanical properties of the nanoscale RVE containing CNT embedded in epoxy. In the second, the determined mechanical properties of the nanoscale RVE were used to determine the bulk mechanical properties of the nanocomposite using Mori–Tanaka model. Fig. 1 demonstrates the steps involved in the hierarchical multiscale model. To our knowledge, among the reported multiscale models, a newly developed atomistic based continuum model known here after as ABC [38,41,46] is the first which has successfully described the atomic-property relations of CNT-reinforced polymer composite in a continuum framework. In comparison to other multiscale models, ABC technique distinguished its novelty by incorporating the nanoscale transversely isotropic RVEs in determining interfacial and mechanical. In fact, Meguid and coworkers [38,41,46] reported both the interfacial and mechanical properties of CNT-reinforced polymer composites using ABC technique and succeeded in investigating the influence of different parameters, such as embedded CNT length, thickness of the CNT-polymer interface, CNT diameter and *LJ* cut-off distance. In view of this, our MD-Micromechanics based multiscale modeling approach was compared with this novel ABC approach. For the sake of completeness, we provide below a summary of the two approaches used in modeling the current CNT-Composite system.

2.1. Molecular dynamics modeling and simulations

This Section is devoted fully to molecular dynamics modeling and simulations. All MD runs were conducted with the aid of large-scale atomic/molecular massively parallel simulator

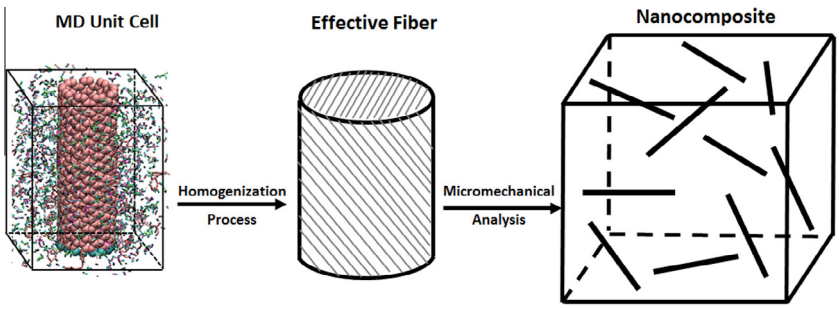

Fig. 1. Modelling steps involved in the developed multiscale model.