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Multiscale modeling of effective electrical conductivity of short carbon fiber-carbon nanotube-polymer matrix hybrid composites

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ABSTRACT

Epoxy matrix reinforced with conventional microscale short carbon fibers (SCFs) and carbon nanotubes (CNTs) form a hybrid material system where the characteristic length scales of SCFs and CNTs differ by multiple orders of magnitude. Several recent studies show that the addition of CNTs into a non-conducting polymer matrix improves both structural performance such as modulus, strength and fracture toughness and functional response such as electrical and thermal conductivities of the resulting nano-composite. In this study, a physics-based hierarchical multiscale modeling approach is presented to calculate the effective electrical conductivity of SCF-CNT-polymer hybrid composites. A dual step procedure is adopted to couple the effects of nano- and micro-scale so as to estimate the effective electrical properties of the composite. First, CNTs are dispersed into the non-conducting polymer matrix to obtain an electrically conductive CNT-epoxy composite. The effective electrical conductivity of CNT-epoxy composite is modeled using a physics-based formulation for both randomly distributed and vertically aligned cases of CNTs and the results are verified with the measured data available in the literature. In the second step, SCFs are randomly distributed in the CNT-epoxy composite and the effective electrical conductivity of the resulting SCF-CNT-epoxy hybrid composite is estimated using a micromechanics based self-consistent approach considering SCFs as microscopic inhomogeneities.

1. Introduction

Hybrid composites are gaining the attention of the materials research community as they offer a wide range of possibilities to tailor their properties at various length scales. Hybrid polymer composites consisting of randomly distributed micron-size short carbon fibers and CNTs possess excellent specific mechanical properties [1,2] coupled with inherent multifunctionality [3,4]. Addition of SCFs changes a non-conducting pristine polymer into a conductive polymer composite. As the volume fraction of SCF is increased, the effective electrical conductivity of the composite increases. With the addition of a few percent SCF, the effective electrical conductivity of composite approaches a limiting value and remains within the same order of magnitude with further addition of SCF into the matrix. The maximum achievable limit of electrical conductivity for these microscale carbon fiber reinforced composites is in the range of 0.1–20 S/m. However, this envelope of electrical conductivity of SCF-polymer composites can be pushed further by the addition of CNTs providing another scale of electrical pathways in the composites. In this way, polymer matrix composites reinforced with microscopic carbon fibers and CNTs can exhibit improved electrical conductivity compared to carbon fiber-polymer matrix composites [5]. Addition of nanoscale fibers triggers the mechanisms which are otherwise unavailable in these conventional composites. It enhances the utilization of these composites either by expanding the limits of the available properties or by converting them into a multifunctional composite by imparting one or more previously unavailable functionality. In case of CNT-composites, several such examples exist where addition of CNTs has improved the mechanical response e.g. fracture toughness [6–10] of existing composite system or has introduced/enhanced the piezoelectric behavior in conventional composites for structural health monitoring [11,12]. Addition of even small amount of CNTs into the non-conducting polymer improves the electrical conductivity considerably due to extremely low percolation thershold (0.05–0.5 wt.%) [13,14]. However, the extent of improvement also depends on physical features of the CNTs (singlewall vs multiwall CNTs, aspect ratio), their dispersion in the matrix and inter-CNT contact resistance. In case of CNT-polymer composites, the two electrical conductivity mechanisms, namely, electron hopping (tunneling) and conductive network, complement each other.

The aim of this study is to develop a physics-based modeling framework capable of predicting the effective electrical conductivity of SCF-CNT reinforced polymer hybrid composites. However, predicting the effective electrical conductivity of these hybrid composites remains a challenging task due to the vast difference between the spatial scales

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and electrical conduction mechanisms associated with these two sets of inhomogeneities. Due to this difference, the solution strategy to determine the effective electrical conductivity of SCF-CNT-polymer composite is divided into two parts. Fig. 1 shows the two parts with the schematics of respective microstructure of the hybrid composite and the flowchart for the modeling strategy. In the first part, CNTs are dispersed into the non-conductive polymer to convert it into a conductive matrix [Fig. 1(c)]. The computational framework presented herein is based on the assumption that CNT content in the composite is above the percolation threshold and that the CNT conductive network has already been established. Above the percolation threshold, electron tunnelling mechanism dominates. The effective electrical conductivity of resulting CNT-polymer composite is determined in two steps. In the first step, the CNTs surrounded by a thin layer of polymer are converted into equivalent CNT inclusions and the homogenized electrical conductivity of CNT-polymer composite consisting of these equivalent inclusions is calculated using micromechanics based Mori-Tanaka method. In the second part, SCFs are distrubuted in the resulting conductive polymer matrix [Fig. 1(d)]. The homogenized electrical conductivity of CNT-SCF-polymer composite is determined by a self-consistent approach. Based on the approach described above, the manuscript is organized as follows. Section 2 describes the methodology used for calculating the homogenized electrical conductivity of CNT-polymer composite i.e. the host matrix for SCFs. Following this, Section 3 presents the details of self-consistent formulation used in predicting the effective electrical conductivity of SCF-conductive matrix composite. Section 4 presents the results of homogenized electrical conductivity of CNT-polymer and CNT-SCF-polymer matrix composites. Section 5 concludes with the main findings of this research.

2. Effective electrical conductivity of carbon nanotube-epoxy composites

The effective electrical conductivity of CNT-polymer composite or the 'conductive matrix' is largely limited by the interfacial contact resistance between CNTs and the matrix. Several physics-based computational approaches have been developed for calculating the homogenized electrical conductivity of CNT-polymer composites [15–21]. In CNT-polymer composites, the CNT-CNT and CNT-polymer interactions are governed by weak non-bonding interactions. These interactions create a shell around CNTs which is referred to as interphase layer. This interphase layer provides interfacial resistance to CNTs. The properties of the interphase layer depends upon CNT concentration, CNT-CNT potential-well depth and the average separation distance between the CNTs. The pristine CNTs together with the surrounding interphase layer is replaced by a homogenized CNT inclusion whose effective properties include the effects of the interphase. Fig. 2 shows the strategy adopted in determining the electrical conductivity of the homogenized CNT inclusions.

Consider an isolated CNT surrounded by an interphase layer on its lateral surface as shown in Fig. 1(a). Due to the difference in resistive contributions from the interphase layer at the ends and on the lateral surface of the CNTs, the electrical conductivity of homogenized inclusion becomes transversely isotropic. The effective electrical conductivity of the homogenized inclusion is therefore calculated in two steps. In the first step, the CNT is considered to be surrounded by the interphase layer of uniform thickness along its length only as shown in Fig. 2(a). The longitudinal and transverse electrical conductivities of this inclusion are calculated separately (see [22]). Consider the intermediate inclusion as shown in Fig. 2(b), subjected to a uniform electrical field, E_0, which is directed perpendicular to CNT axis of symmetry [Fig. 2(d)]. Considering the length of CNT along the z-axis, the CNT cross section that lies in the x-y plane is shown in Fig. 2(d). Due to uniform thickness of the interphase layer around the CNT, the two transverse electrical conductivities, σ_x and σ_y for the intermediate inclusion can be considered to be equal. In order to calculate the transverse electrical conductivity of the intermediate inclusion, $\sigma_x(=\sigma_y)$, the Maxwell's equation in cylindrical coordinates is applied to intermediate inclusion [Fig. 2(b)] subjected to uniform electrical field E_0 [see Fig. 2(d)].

$$\nabla^2\phi = \phi_{,rr} + \frac{1}{r}\phi_{,r} + \frac{1}{r^2}\phi_{,\theta\theta} = 0 \tag{1}$$

where, ϕ is the electrical field potential. The plane containing coordinate (r, θ) forms the plane of isotropy of the homogenized intermediate inclusion. The general solution for the Laplacian given by Eq. (1) is given as:

$$\phi = \sum_{n=1}^{\infty}\left(\left(A_n r^n + \frac{B_n}{r^n}\right)(C_n \cos(n\theta) + D_n \sin(n\theta))\right. \tag{2}$$

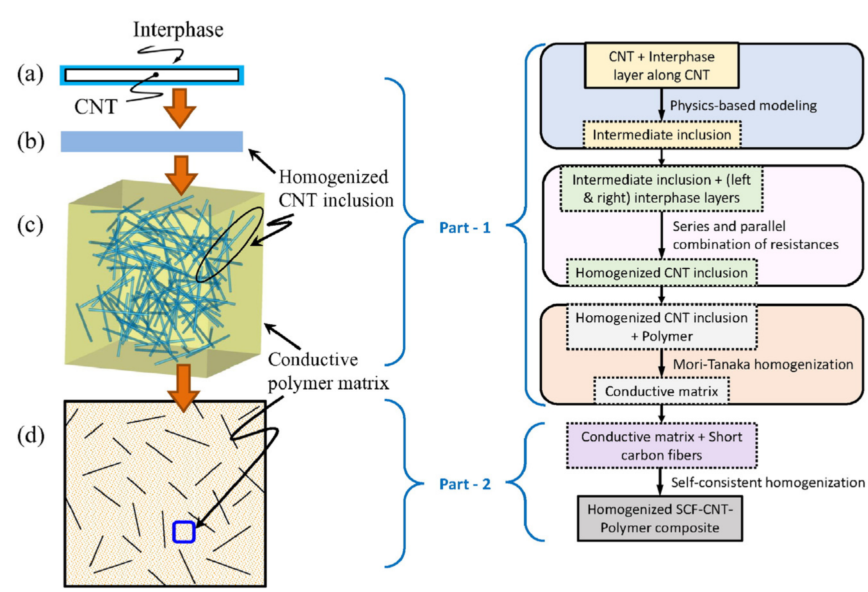

Fig. 1. Schematic representation of the computational approach involved in calculating the effective electrical conductivity of CNT - SCF - polymer matrix hybrid composites: (a) CNT surrounded by the interphase; (b) Homogenized CNT inclusion; (c) Representative volume element for the conductive matrix (CNT + polymer); (d) Representative volume element for SCF-CNT-polymer hybrid composite.