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The effects of CNT waviness on interfacial stress transfer characteristics of CNT/polymer composites

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ABSTRACT

Based on the new simplified 3D Representative Volume Element (RVE) for a wavy carbon nanotube (CNT), an analytical model has been developed to study the stress transfer in single-walled carbon nanotube (SWCNT) reinforced polymer composites. The model is capable of predicting axial as well as interfacial shear stresses along a wavy CNT embedded in a matrix. Based on the pullout modeling technique, the effects of waviness, aspect ratio, CNT diameter, volume fraction, Poisson's ratio and matrix modulus on axial and interfacial shear stresses have also been analyzed in details. The results of the analytical model are in good agreements when compared with the corresponding results for straight CNTs.

1. Introduction

Carbon nanotubes (CNTs) have attracted great interest since their discovery in 1991 [1] due to their unique electronic and mechanical properties such as extremely high elastic modulus, tensile strength and resilience. A detailed summary of CNTs mechanical properties can be found in [2]. The high strength and elastic modulus, fibrous shape and very large aspect ratios of these tubes render them ideal candidate as ultra-strong reinforcement for composites [3]. These polymer based nanomaterial-filled composites usually show remarkable property improvements in various physical properties even at very low content of CNTs compared to virgin polymer or conventional composites. CNT-reinforced polymer composites (NRPCs) find widespread use in many fields such as automotive, aerospace, medical, electronics, optics and materials science due to their superior strength-to-weight ratio, fatigue and fracture resistance, and damping characteristics. Lau and Hui [4] and Thostenson et al. [5] give comprehensive reviews of the manufacturing process, the mechanical and electrical properties, and applications of nanotubes and nanotube-reinforced composites.

However, the mechanical property enhancements which are currently realized with these CNTs are often significantly less than those suggested by simple micromechanical models. This may be caused by a variety of factors, including CNT dispersion within the polymer, the interaction between the polymer and the CNT, the size, shape and orientation of the CNT, and the type of CNT used.

Many experimental observations have shown that, in general, CNTs exist in a curved shape in nanotube reinforced polymer composites (NRPCs) [3,5]. Bogetti et al. [6] developed an analytical model based on 2D laminated plate theory to investigate the influence of ply waviness on the stiffness and strength reduction of laminate constructions. Hsiao and Daniel [7] introduced an analytical constitutive model to determine the elastic properties of unidirectional and cross ply carbon/epoxy composites as a function of fiber waviness. Telegadas and Hyer [8] used the stress analysis technique to predict the failure pressure of imperfect thick multilayer composite cylinders, the imperfection being due to layer waviness. Chou and Takahashi [9] predicted the tensile stress–strain response of flexible wavy fiber composites. Chun et al. [10] theoretically investigated the effect of fiber waviness on the nonlinear behavior of unidirectional composites under tensile and compressive loads. Jortner [11] also reviewed some of the earliest works on the elastic behavior of wavy fiber composites. Many previous studies have only investigated the effect of waviness on percolation threshold and elastic stiffness of composites. For example, Fisher et al. [12–14] and Shi et al. [15] have employed 3D finite element and helical spring model, respectively, to show the effect of waviness on the composite moduli. Yi et al. [16] and Berhan and Sastry [17] considered the effect of nanotube waviness on percolation onset by approximating nanotubes with a sinusoidal shape. More recently, Li and Chou [18] simulated wavy nanotubes using polygons. Shao et al. [19] show that both the waviness and debonding can significantly reduce the stiffening effect of the CNTs.

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The effective moduli are found to be very sensitive to the waviness when the latter is small, and this sensitivity decreases with the increase of the waviness. By combining micromechanics model and finite element simulation, Li et al. [20] found that the nanotube waviness tends to reduce the elastic modulus but increase the ultimate strain of a composite. In a similar study, the influence of the nanotubes curvature on the elastic properties of nanocomposites is studied utilizing the modified fiber model and the approach developed by Mori–Tanaka [21]. The results show that the effect of nanotube curvature on reducing the modulus of the effective fiber is not limited to in-plane curvature but also to curvature in 3D. Tsai et al. [22] found that the inclusion waviness have a great effect on tensile moduli and shear modulus, especially at higher fiber volume fraction, for unidirectional composites. However, if the inclusions were either randomly dispersed or only partially aligned, the degree of waviness effect was smaller. The basic conclusion reached in these studies is that the waviness tends to increase the percolation threshold but reduces the elastic stiffness.

In addition, extensive research has been performed to understand the effect of interface on the mechanical properties of NRPCs. Wagner [23] used the Kelly–Tyson (KT) model, which has been widely used to study the matrix-fiber stress transfer mechanism in micron size fiber composites, to study the interfacial shear strength between the NT and polymer matrix. Additionally, Lau [24] has conducted an analytical study on the interfacial bonding properties of NRPCs using the well-developed local density approximation model, classical elastic shell theory and conventional fiber-pullout model, and Gou et al. [25] examined the molecular interactions between the nanotube and thermosetting matrix, epoxy resin, during composite processing. Based on fiber pullout modeling, Zhang and Wang [26] investigated the thermal effects on interfacial stress transfer characteristics of NRPCs under thermal loading by means of thermoelastic theory. Recently, Jiang et al. [27] and Tan et al. [28] established the cohesive law for interfaces between a CNT and polymer that are not well bonded and are characterized by the van der Waals (vdW) forces. A review of the state of the art in mechanics of carbon nanotube polymer composites can be found in [29]. To our knowledge no analytical studies have been reported in the literature predicting the effect of waviness on the stress transfer characteristics of CNT reinforced polymer composites.

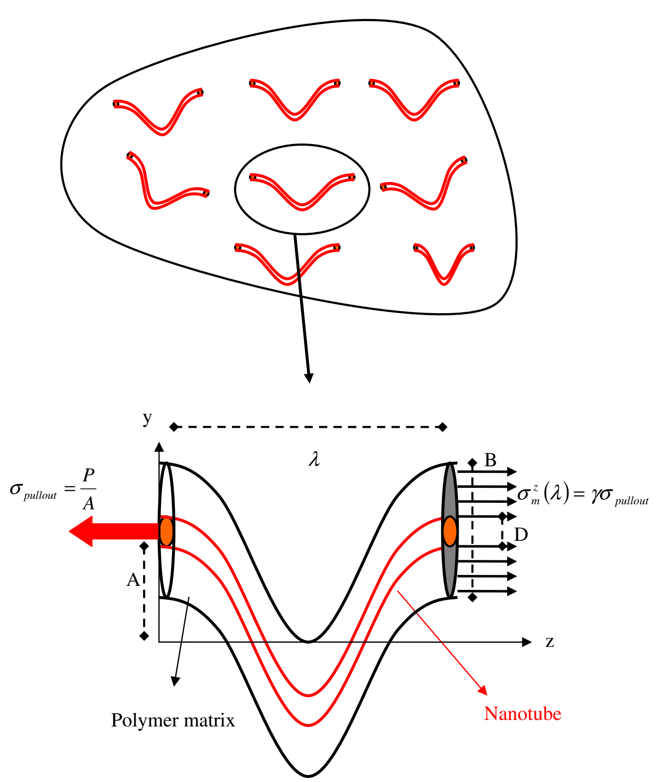

Fig. 1. RVE for Wavy CNT embedded in polymer matrix. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

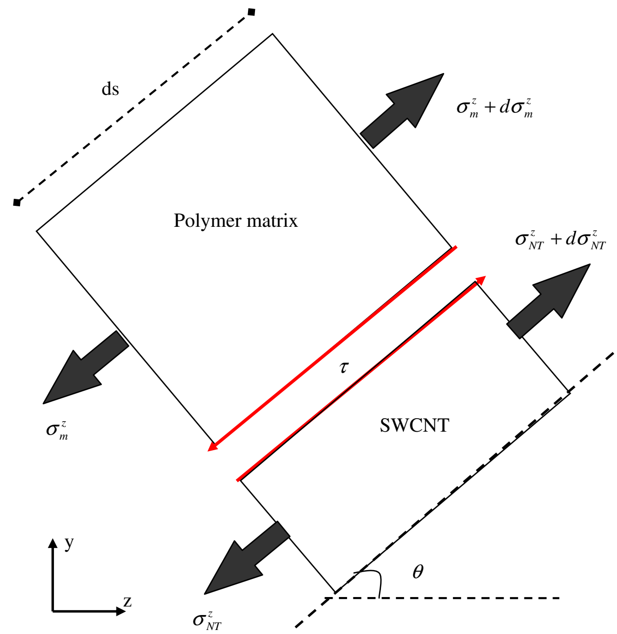

Fig. 2. RVE element for interfacial shear stress between polymer matrix and SWCNT. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

In this study an analytical model is developed to investigate the effects of nanotube waviness on interfacial stress transfer characteristics of single-walled carbon nanotubes (SWCNTs)/polymer composites, based on conventional fiber pull-out models. Solution for the stress distribution can be obtained which satisfy all equilibrium equations, boundary and continuity conditions at the CNT/matrix. The numerical results show that the waviness of CNTs significantly influences the interfacial stress transfer characteristics of CNT/polymer composites which tend to reduce the stiffening effect of the CNTs though they have exceptionally high modulus.

2. Analytical formulation

Using high magnification electron microscopy images, embedded CNTs are often characterized by a certain degree of waviness along their axial dimension. To take advantage of the very high Young's modulus and strength of the CNTs, the mechanical load must be transferred efficiently between matrix and CNTs. Hence, the stress transfer between the reinforcing CNTs and the matrix materials at the interface is an important phenomenon which critically controls the mechanical properties of NRPCs under various loading conditions.

2.1. Modeling the wavy nanotube

Despite the discrete nature of atomic structure of CNTs, we will treat the CNTs as a solid circular cross-sectional area, which implicitly introduces two simplifications into the analysis. First, this assumption neglects the hollow nature of the CNTs. Second, by modeling the nanotube as a continuum we are neglecting any

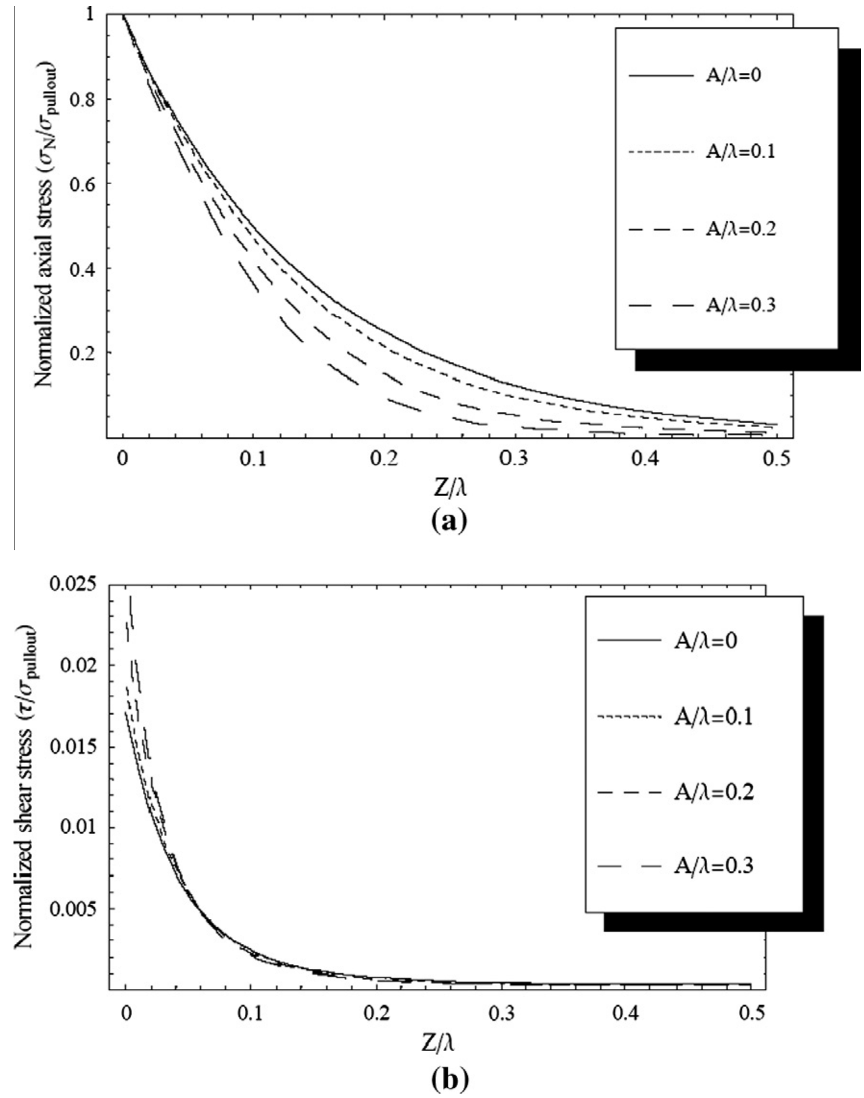

Fig. 3. Normalized CNT stresses vs. normalized length of CNT at different values of waviness: (a) axial stress and (b) interfacial shear stress.

possible relative motion between individual shells or tubes in a multi-walled nanotube (MWNT) and an NT bundle, respectively. In addition, individual phase materials are modeled as linear elastic and isotropic and also perfect bonding between polymer and CNTs is assumed. Because of its simplified equations and methodology for the evaluation of interface parameters from the experimental results, this model is widely used by many researches. Also using nonlinear fiber pullout modeling technique, Zhou and Mai [30] and Xiao and Liao [31] have investigated the effect of anisotropic behavior of fiber and straight CNTs on the interfacial stresses of NRPCs, respectively. More recently, Chen et al. [32] formulated the bonded stage of the pull-out process based on classic shear lag model assumptions and develop a 3D finite element model to verify assumptions.

Consider the wavy CNT of solid cross-section shown in Fig. 1. Nanotube waviness was modeled by prescribing a sinusoidal CNT shape [14], y, of the form:

$$y = \mathrm{A}\cos\left(\frac{2\pi z}{\lambda}\right) \tag{1}$$

where A, λ and z are the sinusoidal amplitude, sinusoidal wavelength and fiber longitudinal direction, respectively. An increment of arc length, ds, of CNT can be described as follows:

$$ds = \frac{dz}{\cos\theta} = dz.\sqrt{1 + 4\pi^2\left(\frac{\mathrm{A}}{\lambda}\right)^2 \sin^2\left(\frac{2\pi z}{\lambda}\right)} \tag{2}$$

2.2. Fiber pull-out modeling

Single fiber pullout as a technique of measuring fiber/polymer adhesion has been in use for many years [33,34]. Assuming that the CNT is undertaking the pullout force, P the boundary conditions at two ends of this model, i.e. $z = 0$ and $z = \lambda$, are (see Fig. 1):

$$\sigma^z_{\mathrm{CNT}}(0) = \sigma_{pullout}, \quad \sigma^z_{\mathrm{CNT}}(\lambda) = 0, \quad \sigma^z_m(0) = 0, \\ \sigma^z_m(\lambda) = \gamma\sigma_{pullout}, \quad \sigma_{pullout} = \frac{P}{A} \tag{3}$$

where σ, P and A represents the stress, axial load and cross sectional area of CNT, respectively. Also subscripts 'CNT' and 'm' refer to the materials of the CNT and matrix, respectively. The superscript 'z' stands for the longitudinal direction and γ is the area ratio of the CNT and matrix, i.e.

$$\gamma = \frac{A_{CNT}}{A_m} = \frac{D^2}{B^2 - D^2} \tag{4}$$

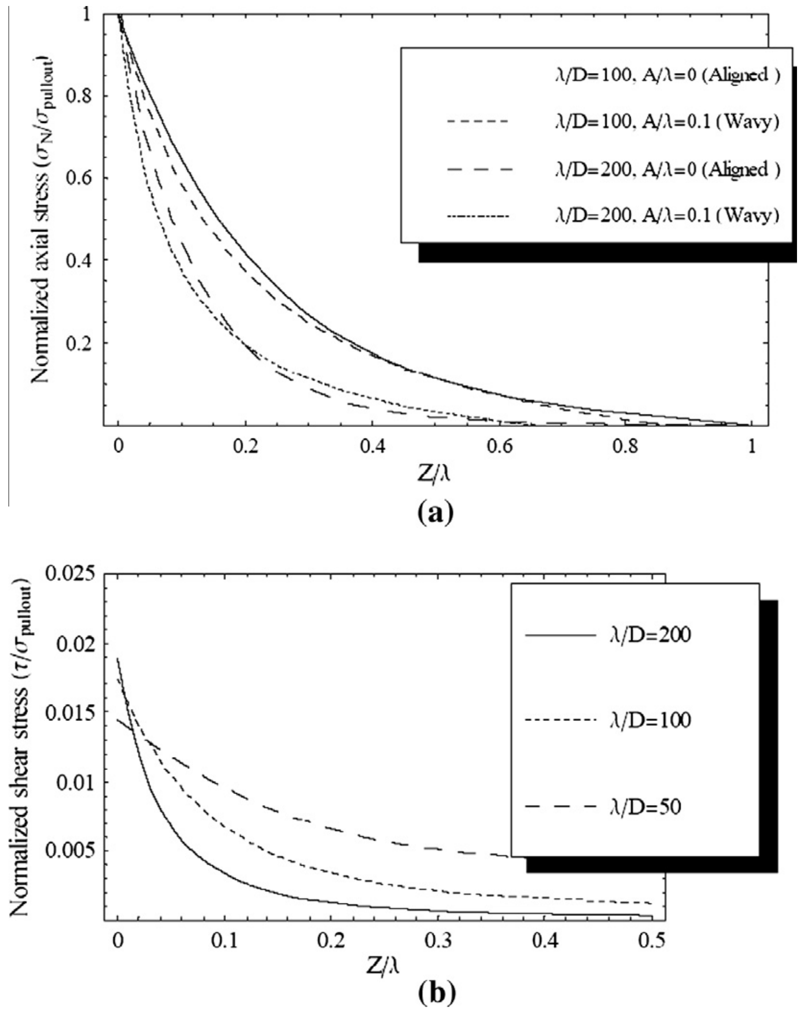

Fig. 4. Plot of the (a) axial and (b) interfacial stresses, against the normalized length of CNT, Z/λ, for different values of wavelength, λ/D. In Fig. 4b, $A/\lambda = 0.1$.

where D and B are the diameter of the CNT and matrix, respectively. The equilibrium equations between the matrix axial stress, CNT interfacial shear stress and axial can be expressed as (illustrated in Fig. 2):

$$\sigma^z_{CNT}\cdot\left(\frac{\pi D^2}{4}\right)-(\sigma^z_{CNT}+d\sigma^z_{CNT})\cdot\left(\frac{\pi D^2}{4}\right)-\tau\cdot(\pi D ds)=0\Rightarrow\frac{d\sigma^z_{CNT}}{ds}=-\frac{4}{D}.\tau$$
$$\sigma^z_m\cdot\left(\frac{\pi(B^2-D^2)}{4}\right)-(\sigma^z_m+d\sigma^z_m)\cdot\left(\frac{\pi(B^2-D^2)}{4}\right)+\tau\cdot\gamma\cdot(\pi D ds)=0\Rightarrow\frac{d\sigma^z_m}{ds}=\frac{4\gamma}{D}\cdot\tau \tag{5}$$

where the axial stresses σ^z_{CNT} and σ^z_m are assumed z-functions only and $\sigma_{pullout} = \sigma^z_{CNT} + \sigma^z_m/\gamma$. Since the CNT radius is small, the stresses in the CNT can satisfy all boundary and equilibrium conditions approximately.

The relationships between the stresses, strains and displacements are

$$\varepsilon^r_j = \frac{\partial u^r_j}{\partial r} = \frac{1}{E_j}\left[\sigma^r_j - \upsilon_j\left(\sigma^\theta_j + \sigma^z_j\right)\right] \tag{6}$$

$$\varepsilon^\theta_j = \frac{u^r_j}{r} = \frac{1}{E_j}\left[\sigma^\theta_j - \upsilon_j\left(\sigma^r_j + \sigma^z_j\right)\right] \tag{7}$$

$$\varepsilon^z_j = \frac{\partial u^z_j}{\partial z} = \frac{1}{E_j}\left[\sigma^z_j - \upsilon_j\left(\sigma^r_j + \sigma^\theta_j\right)\right] \tag{8}$$

where E and υ denotes the Young's modulus and Poisson's ratio and the subscript 'j' can be 'CNT' (nanotube/fiber) or 'm' (matrix), respectively. Based on previous works [34–36], the hoop and radial stresses of the CNT are:

$$\sigma^r_{CNT}(z) = \sigma^\theta_{CNT}(z) = q(z) \tag{9}$$

where $q(z)$ is the interfacial radial stress caused by the fabrication-induced residual stress and Poisson's contraction. Using Eqs. (6)–(9) the displacements of the CNT and matrix at $r = D/2$ are

$$u^r_{CNT} = \frac{D}{2E_{CNT}}\left[(1-\upsilon_{CNT})q(z) - \upsilon_{CNT}\sigma^z_{CNT}(z)\right],$$
$$\frac{\partial u^z_{CNT}}{\partial z} = \frac{-1}{E_{CNT}}\left[2\upsilon_{CNT}q(z) - \sigma^z_{CNT}(z)\right] \tag{10}$$

$$u^r_m = \frac{-D}{2E_m}\left\{(1+2\gamma+\upsilon_m)q(z) + \gamma\upsilon_m(\sigma_{pullout} - \sigma^z_{CNT}(z)) + U_1\frac{d^2\sigma^z_{CNT}}{dz^2}\right\},$$
$$\frac{\partial u^z_m}{\partial z} = \frac{1}{E_m}\left\{\gamma(\sigma_{pullout} - \sigma^z_{CNT}(z)) + 2\upsilon_m\gamma q(z) - U_2\frac{d^2\sigma^z_{CNT}}{dz^2}\right\} \tag{11}$$

where

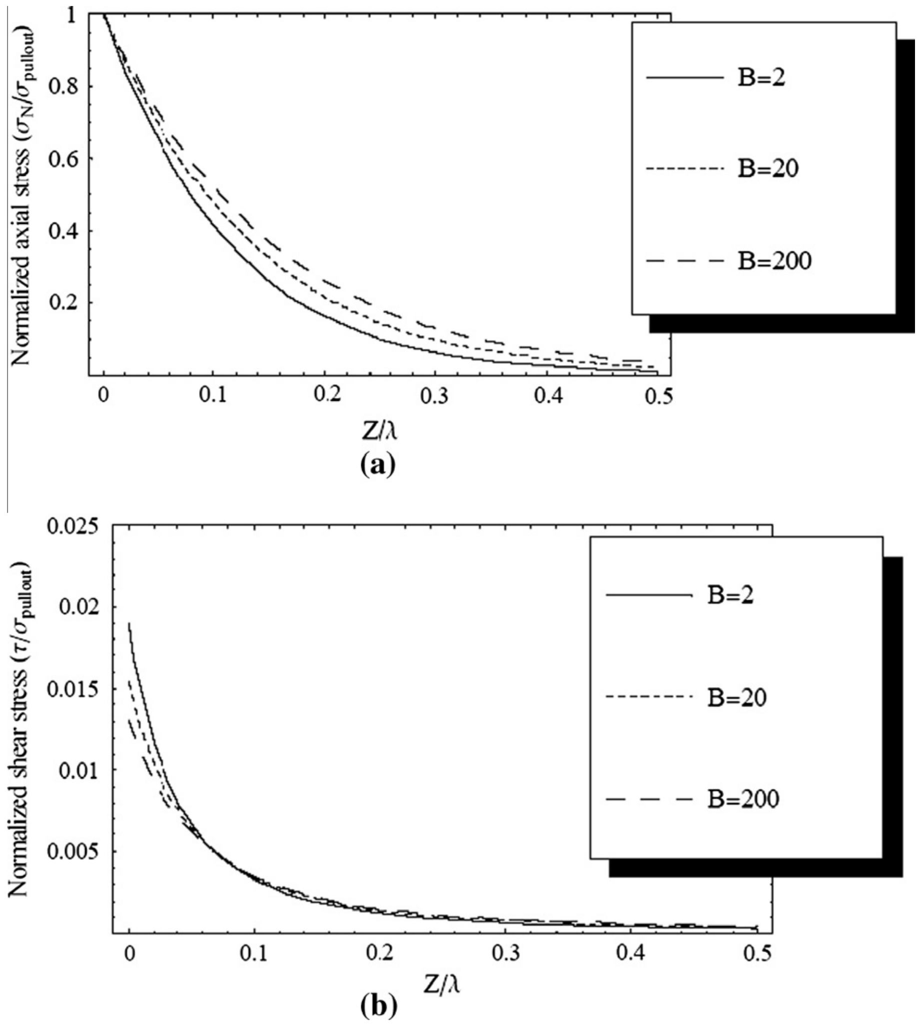

Fig. 5. Effects of matrix diameter on the (a) axial (b) interfacial shear stresses of NRPCs.

$$U_1 = \frac{\gamma^2}{8}\left\{\frac{\eta_1}{2}B^2 Ln\left(\frac{B}{D}\right)\left[1+\gamma\left(\frac{B^2}{D^2}+1\right)\right] - \frac{\eta_2}{2}(B^2+D^2) + B^2 - \frac{\eta_1}{2}(B^2-D^2)\right\}, U_2 = \frac{\gamma \upsilon_m}{4}\left\{\frac{\eta_1}{2}B^2 Ln\left(\frac{B}{D}\right)[1+\gamma] - \frac{\eta_2}{4}(B^2+D^2) + \frac{B^2}{2} - \frac{\eta_1}{4}(B^2-D^2)\right\} \quad (12)$$

It should be noted that when the interfacial shear stresses vary rapidly with the axial distance, in solid cylinder modeling, we can not neglect the effects of the radial and circumferential matrix stresses. Note that the matrix and CNT are fully bonded at $z = 0$ and therefore their respective $\partial u^z/\partial z$ must be identical for all $z > 0$ to satisfy the continuity requirement for u^z. Combining Eqs. (10)–(12) and the continuity of axial and radial deformation at the any bonded interfaces, $u^z_m(D/2, z) = u^z_{CNT}(D/2, z)$ and $u^r_m(D/2, z) = u^r_{CNT}(D/2, z)$, the axial stress in the CNT can be obtained from the following differential equation:

$$\frac{d^2\sigma^z_{CNT}(z)}{ds^2} - A_1\sigma^z_{CNT}(z) - A_2\sigma_{pullout} = 0 \quad (13)$$

in which has been solved and plotted numerically. The terms of A_1, A_2 are the functions of the mechanical properties and geometrical factors of the CNT and matrix, and are given as follows:

$$A_1 = \frac{\alpha(1-2K\upsilon_{CNT}) + \gamma(1-2K\upsilon_m)}{U_2 - 2KU_1}, \quad A_2 = \frac{-\gamma(1-2K\upsilon_m)}{U_2 - 2KU_1} \quad (14)$$

where:

$$K = \frac{\alpha\upsilon_{CNT} + \gamma\upsilon_m}{\alpha(1-\upsilon_{CNT}) + 1 + 2\gamma + \upsilon_m}, \quad \alpha = \frac{E_{CNT}}{E_m}, \quad \eta_1 = \frac{2(1+\upsilon_m)}{\upsilon_m}, \quad \eta_2 = \frac{(1+2\upsilon_m)}{\upsilon_m} \quad (15)$$

The magnitudes of the axial and shear stresses are highly affected by the inherent properties and geometrical factors of the nanotubes and matrices. It should be mentioned that for aligned CNTs ($A = 0$), Eq. (13) reduced to the differential equation that was reported before [24,31]. After determining the axial stress in CNT from Eq. (13), with replacing it to Eq. (5), the interfacial shear stress and axial stress in matrix can be easily obtained.

3. Numerical results

In this section, numerical examples are given for a CNT/polymer composite system to demonstrate the axial and interfacial shear stresses distribution of the wavy CNTs in a fully bonded region. The material properties and geometrical characteristics of the CNTs and matrix are [24]:

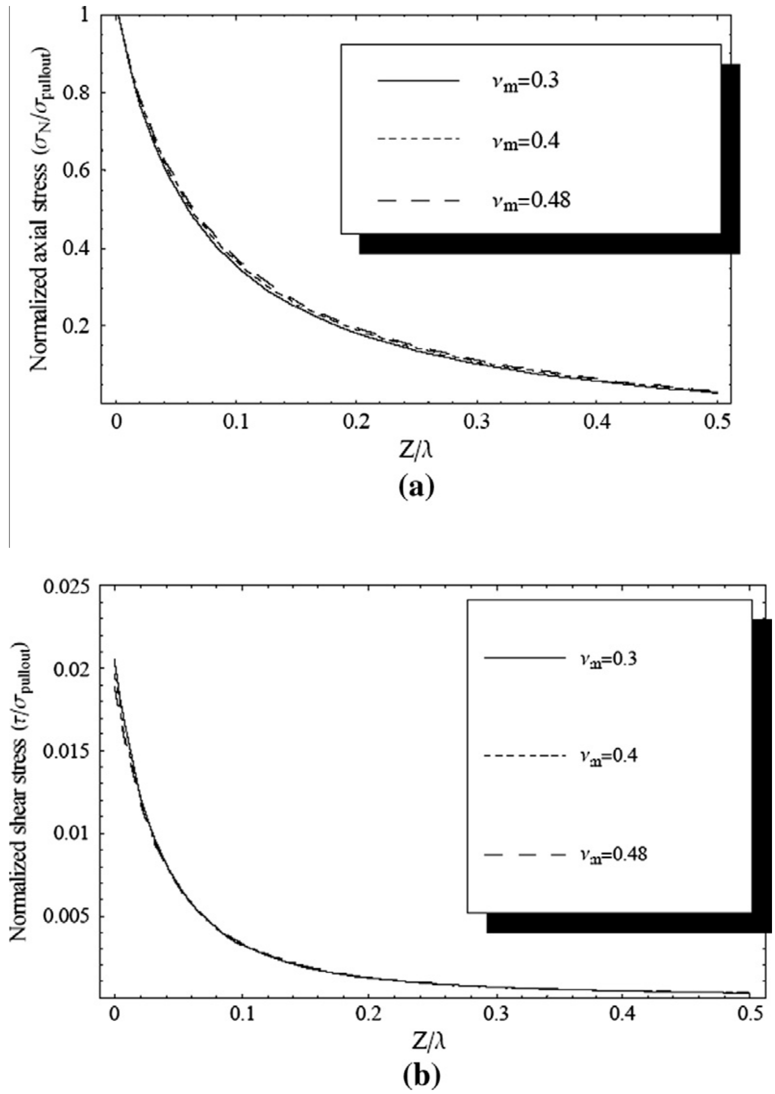

Fig. 6. Effects of Poisson's ratio on the (a) axial and (b) interfacial shear stresses of NRPCs.

$$D = 5\text{ nm},\quad B = 1\ \mu\text{m},\quad \upsilon_m = 0.48,\quad \upsilon_{CNT} = 0.3,\quad E_m = 3\text{ GPa},\quad E_{CNT} = 900\text{ GPa},\quad (A/\lambda) = 0.1,\quad P = 20\text{ nN},\quad \lambda = 1\ \mu\text{m}, \tag{16}$$

3.1. Effects of CNT waviness (A/λ)

Fig. 3 shows the variation of normalized axial and interfacial shear stress in NRPCs along dimensionless axial distance (z/λ) for different values of waviness. From Fig. 3a, it is observed that the average axial stress of the wavy CNT is lower than that of aligned ones and decreases by increasing the waviness. This reduction in axial stress between CNTs and polymer matrix will reduce the stiffening effect of the CNTs. In Fig. 3b it is seen that the maximum interfacial shear stress occurs at $(z/\lambda) = 0$ and gradually increases as the magnitude of the waviness increases and has negligible effect at $z/\lambda > 0.1$. The influence of waviness of CNTs is more pronounced on the axial stress rather than the interfacial shear stress. The results for aligned CNTs i.e.$(A/\lambda) = 0$, are in good agreement with previous analytical published data obtained from the uses of local density approximation, elastic shells and conventional fiber pullout models [24].

3.2. Effects of CNT wavelength (λ/D)

In Fig. 4 the variation of axial and interfacial stresses with the normalized length of CNT, z/λ, at different values of wavelength, λ/D, is shown. With increasing the wavelength of the CNT, the normalized axial stress reduces and also drops more sharply from 1 to zero (see Fig. 4a). These results show that higher values of wavelength of the CNTs will result in lower overall stiffness of NRPCs. The maximum interfacial shear stress increases by increasing the wavelength however the average interfacial shear stress decreases, see Fig. 4b. This high interfacial shear stress could critically result in debonding phenomena and implies that the interface debonding due to the shear stress easily arises from the pullout end of CNTs.

3.3. Effects of matrix diameter

As intuition suggests, by increasing the diameter of matrix (B), the volume fraction of the embedded nanotube will decrease. The results obtained from Fig. 5 show that with increasing the diameter of matrix, the maximum shear stress decreases. The effect of matrix diameter on interfacial shear stress at $z/\lambda > 0.1$ is

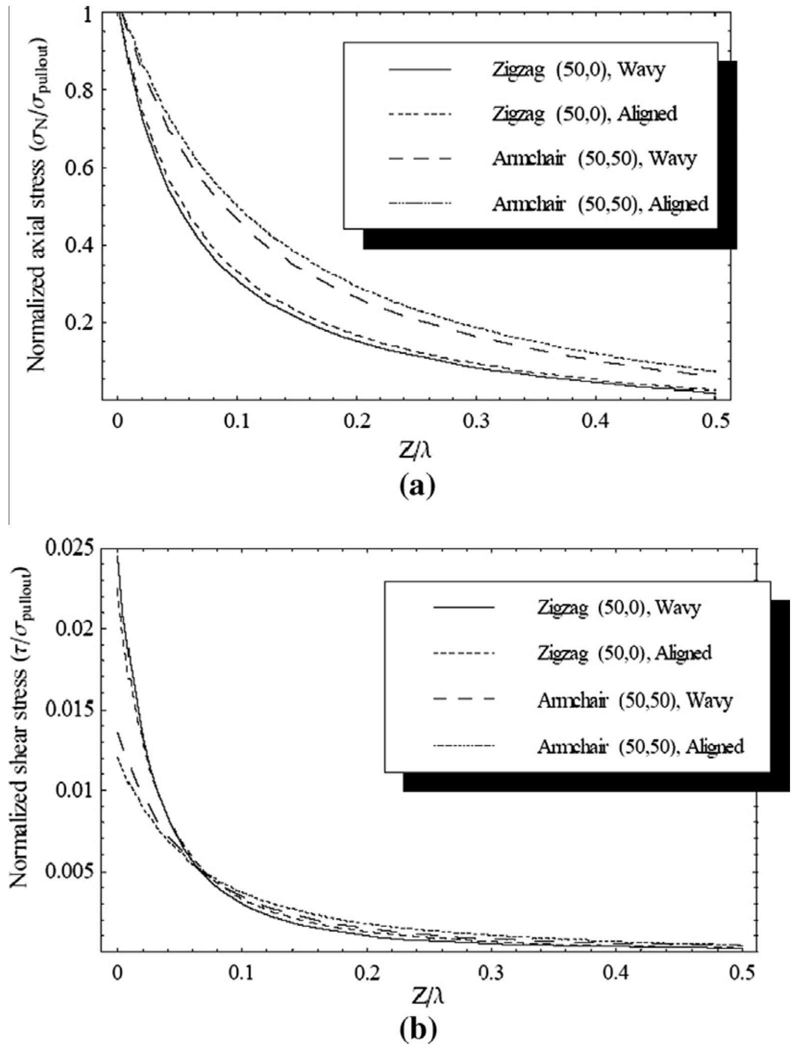

Fig. 7. Variation of normalized (a) axial and (b) interfacial shear stress of Armchair and Zigzag SWCNTs with normalized length (the effect of CNT diameter).

negligible and it has the maximum influence on axial stress at $z/\lambda \cong 0.2$.

3.4. Effects of Poisson's ratio

From Fig. 6, it is observed that Poisson's ratio has negligible effect of interfacial shear stress for both straight and wavy CNTs.

3.5. Effects of CNT diameter

In the proposed analytical model we have neglected the effect of arrangement of carbon atoms in the CNTs on the interfacial stress transfer and therefore in this subsection we have investigated the effect of CNT diameter/area, being due to the type of CNT, on the stress transfer characteristics of CNT/polymer composites. Representing the CNTs by a pair of indices (n, m) called the chiral vector, the diameter of CNTs is calculated as [2]:

$$D = \frac{0.246}{\pi}\sqrt{n^2 + nm + m^2}[\mathrm{nm}] \tag{17}$$

where for the Zigzag ($m = 0$) and Armchair ($n = m$) nanotubes it reduces to:

$$D_{\mathrm{Zigzag}} = \frac{0.246}{\pi}n[\mathrm{nm}], \quad D_{\mathrm{Armchair}} = \frac{0.246\sqrt{3}}{\pi}n[\mathrm{nm}] \tag{18}$$

In Fig. 7, a plot of the interfacial stresses of wavy and straight SWCNTs with different chiralities is shown. The maximum shear stress of a Zigzag (50, 0) CNT, $D = 3.9152$ nm, is comparatively higher than that of Armchair (50, 50), $D = 6.7813$ nm, nanotubes. Because of the small cross sectional area of the Zigzag nanotubes, the total surface contact area at the bond interface of the Zigzag nanotube is therefore comparatively smaller than that of the Armchair, and hence, higher interface shear stress is generated.

3.6. Effects of matrix modulus

Fig. 8 provides a comparison of the wavy and straight CNTs with different matrix modulus. Fig. 8a shows that by increasing the matrix modulus, E_m, the axial stress will decrease more sharply. By increasing the matrix modulus the maximum interfacial shear stress will increase dramatically, i.e., making the matrix modulus three times larger will cause the maximum shear stress become almost doubled (see Fig. 8b). These observations can be used in manufacturing process of NRPCs in order to prevent debonding due to high interfacial shear stresses. The wavy CNT is found to be more sensitive to the matrix modulus than the aligned ones.

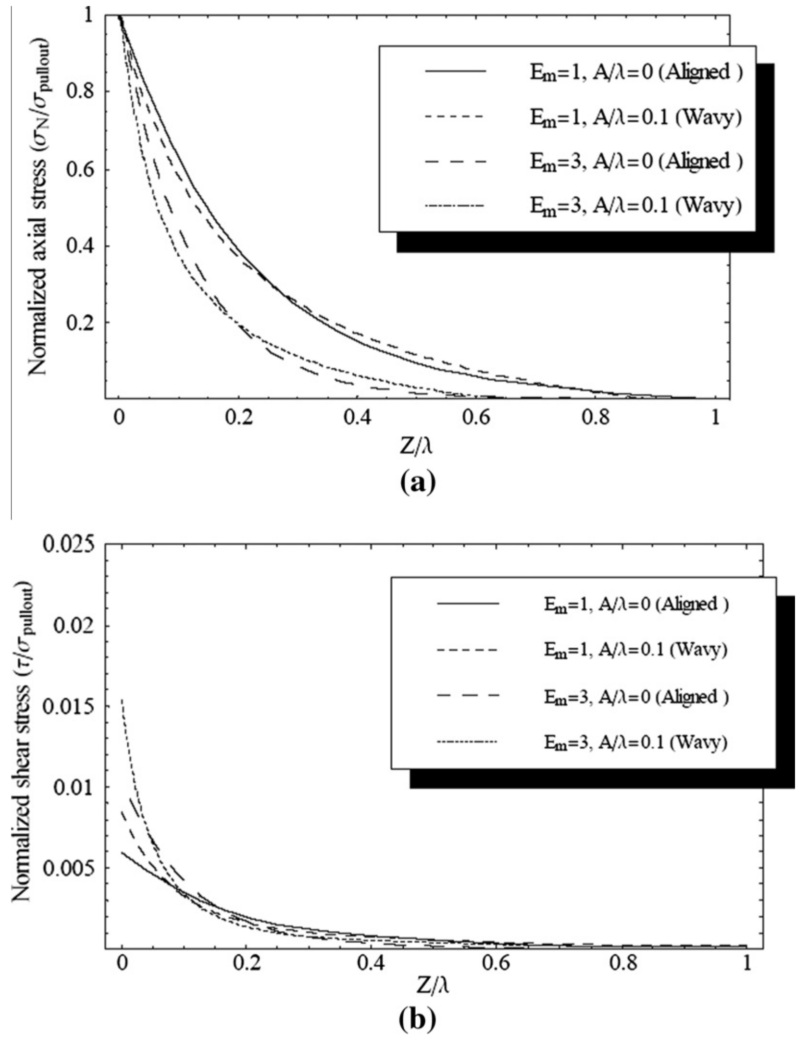

Fig. 8. Effects of matrix modulus on the (a) axial (b) interfacial shear stresses of NRPCs.

4. Concluding remarks

Based on the pullout modeling technique, a new simplified 3-D Representative Volume Element (RVE) of a wavy CNT has been developed to study the effects of waviness, aspect ratio, CNT diameter, volume fraction, Poisson's ratio and matrix modulus on axial and interfacial shear stresses of NRPCs. The computational results have lead to the following conclusions:

- The maximum interfacial shear stress of a wavy CNT is higher than that of straight ones and increases with increasing the waviness; i.e. in constant value of λ, the value of A increases, but the variation trend of the average axial stress is reverse.
- The results show that with increasing the wavelength of the CNT; i.e. increasing the value of λ, the maximum interfacial shear stress increases.
- Maximum interfacial shear stress increased sharply with increasing the volume fraction of the embedded wavy nanotube.
- Like the straight CNTs, the effect of Poisson's ratio on the interfacial stresses can be ignored.
- The wavy CNT is found to be more sensitive to the matrix modulus than the straight ones. By increasing the matrix modulus, E_m, the maximum interfacial shear stress increases, however, the axial stress decreases. These observations can be used in manufacturing of NRPCs.
- If the maximum value of the interfacial shear stress is used to assess debonding, the longer and/or wavy CNT may result in faster debonding. Also Zigzag CNTs are more critical than the Armchair ones.

The model proposed in this research can also be utilized in the analysis of other problems involving nanotube-reinforced polymers, including viscoelastic response and in the determination of thermal and electrical conductivity. The correlations of the present results from the analytical model with those of the straight CNTs are very satisfactory, the comparison is not shown here.

The effect of anisotropic properties of CNTs, end caps, defects and any possible relative motion between individual shells or tubes in a MWNT and Nanotube (NT) bundles will be studied in the future.

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