EFFECTS OF INTERPHASE AND MATRIX PROPERTIES ON EFFECTIVE TENSILE ELASTIC MODULUS OF CARBON NANOTUBE-BASED COMPOSITE

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Abstract: *The aim of this research is to assess the effects of interphase property and matrix property on the tensile elastic modulus (TEM) of the carbon nanotube (CNT) using a 3-D nanoscale representative volume element (RVE) based on continuum mechanics and using the finite element method (FEM). Formulas to extract the effective material constants from solutions for the RVE is derived based on the elasticity theory. Based on the strength of materials theory, an extended rule of mixtures, for estimating the effective Young's modulus, is applied for comparisons with the numerical solutions based on the elasticity theory. Both long and short CNT embedded in matrix at a volume fraction of 2% and 5% respectively is considered for investigating the effects of interphase and matrix property variation. The results demonstrate that in both the cases, matrix property and interphase property significantly influence the TEM of the CNT based composite. These results suggest that a coating of harder polymer on the CNT or a surface treatment can significantly increase the TEM of CNT based composite.*

Keywords: *Carbon nanotube, Nanocomposites, Interphase, Tensile elastic modulus, FEM.*

INTRODUCTION

Carbon nanotubes (CNT) are carbon fibers with diameters on the nanometer scale. Carbon nanotubes have been shown to possess exceptional stiffness and strength, extraordinary resilience, remarkable thermal and electrical properties[1]. There has been intense interest in carbon nanotubes (CNTs) since their discovery by Ijima in 1991[2], because they possess unique structural and electronic properties which conceivably, could have a far reaching impact on the next generation of advanced products ranging from aerospace vehicles, to surgical implants, to micro and nanoelectronic components[3]. Carbon nanotubes occur as single walled carbon nanotubes (SWCNTs) and multi-walled carbon nanotubes (MWCNTs)[4]. A SWCNT can be viewed as a single sheet of graphite (i.e. graphene), which has been rolled into the shape of a tube[5]. SWCNTs have radii on the order of nanometers and lengths on the order of micrometers resulting in large aspect ratios beneficial to their use in composites[5-6]. Elastic modulus of SWNTs are of the order of 1 TPa, with a breaking strength of about 37 GPa, making SWNT the strongest and stiffest material known[7]. Many believe that CNTs may provide the ultimate reinforcing materials for the development of a new class of nanocomposites[8-9]. Experimental measurement of the effective properties of CNT reinforced polymer matrix composites have indicated substantial increases in the composite modulus over the matrix modulus. Schadler et al. found a 40% increase in the effective stiffness of CNT reinforced epoxy as compared to the matrix value with 5% weight CNTs[10].Qian et al. also found an increase in the effective modulus of CNT reinforced polystyrene to be of the order of 40% for just 1% weight CNTs[11]. A wide variety of composites containing CNTs have been manufactured. Peigney have fabricated composites of CNTs embedded in ceramic powders[12] while Milo has embedded CNTs in poly vinyl alcohol[13].

Nomenclature

TEM	Tensile elastic modulus
FEM	Finite Element Method
RVE	Representative Volume Element
CNT	Carbon nanotube
SWNT	Single-walled carbon nanotube
MWNT	Multi-walled carbon nanotube
r_i	Inner radius of carbon nanotube
r_o	Outer radius of carbon nanotube
E_z	Effective tensile elastic modulus in the axial direction
E^t	Elastic modulus of carbon nanotube
E^m	Elastic modulus of matrix
R	Radius of the matrix
ε	Poisson's ratio

Meltmixing has been used by Potschke to introduce CNTs into a polyethylene matrix[14]. Such efforts have identified several key challenges in the fabrication of CNT composites. Adequate dispersion of CNTs within the matrix has been a key issue given the tendency of CNTs to form bundles due to inter atomic forces[15-16]. Adhesion of the CNTs to the surrounding matrix has been another key issue[17] as the orientation of CNTs and bundles of CNTs within the matrix[18]. Efforts to address the adhesion and dispersion issues in particular have lead to the use of polymer wrapped CNTs and functionalized CNTs producing distinct interphase regions between matrix and CNTs[17-20].

An interphase is unavoidable in the production of polymer matrix composites and can form in a number of ways[21], for example:

- The presence of absorbed contaminants on the fiber surface, which are not absorbed during impregnation and cure;
- Diffusion of chemical species to the interface between fiber and matrix;
- Acceleration or retardation of polymerization at the interface;
- The deliberate inclusion of sizing resin at the time of fiber manufacture;
- To introduce a thin coating on the CNTs which act as a third phase between CNT and matrix[1].

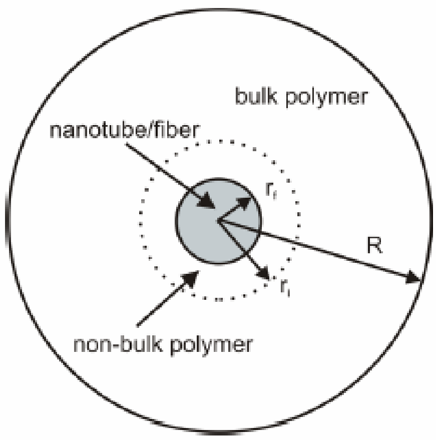

Figure 1. Schematic of bulk polymer and the non-bulk polymer (interphase) regions surrounding an inclusion. Thickness of interphase is $t = r_i - r_f$.[22]

It is anticipated that the CNTs will alter the local polymer morphology in the region directly surrounding the carbon nanotube[22]. This change in local structure will result in a material with mechanical behavior different from that of the bulk polymer. We refer to the material displaying this non-bulk behavior as the interphase, a term used in the composites community that refers to the region separating the fiber and matrix phases[22]. While in traditional composites research, the interface region is generally attributed to a host of factors (such as the use of fiber sizing, mechanical imperfections, and unreacted polymer components), in this research study, discussion will be limited specifically to the change in the TEM of nanotube based composite due to the presence of thin coated layer and interactions with the nanotube inclusions.

Carbon nanotubes (CNT) are an excellent candidate of reinforcements for nanocomposites[23-24]. The CNT-reinforced composites, however, never reach their expected mechanical properties[7]. This has triggered significant research efforts to understand this shortfall and to improve the mechanical properties of CNT-reinforced nanocomposites[25-29].

Nanocomposites possess a large amount of interfaces due to the small (nanometer) size of reinforcements. The interface behavior can significantly affect the mechanical properties of nanocomposites. For example, carbon nanotubes in general do not bond well to polymers[30-31], and their interactions result mainly from the weak Van Der Waals forces[32-36]. Consequently CNTs may slide inside the matrix and may not provide much reinforcing effect. It is, however, important to assess whether the poor interface behavior is indeed responsible for the short fall of CNT-reinforced composites in order to reach their expected properties. One of the significant differences between micron-sized carbon fiber-filled polymers and nanotube-filled polymers the large interfacial area of the nanotubes[38]. This interfacial area provides an opportunity for altering the mobility and properties of a significant volume of polymer near the interface (i.e., the interphase region). The interphase region will play key role in optimizing load transfer between the nanotube and the polymer matrix. While for traditional composites a variety of experimental techniques have been developed in an effort to quantify the fiber-matrix interface[37], for nanotube-polymer composites these tests are exceedingly difficult because of the small size of the nanotubes. The nature of the interphase is becoming more widely studied in nanotube composites[39].

The current study is to evaluate the effects of interphase and matrix property on the effective tensile elastic modulus (TEM) of CNT based composites using a 3-D nanoscale representative volume element based on continuum mechanics and using finite element method. Finite element analysis software ANSYS 10.0 has been used to examine the result in the presence of an interphase. This includes a consideration of the interphase thickness, as well as the isotropic elastic linear nature of the materials used.

RVEs FOR EVALUATION OF THE EFFECTIVE MATERIAL PROPERTIES

CNTs are in different sizes and forms when they are dispersed in a matrix to make a nanocomposite[1]. They can be single-walled or multi-walled with length of a few nanometers or a few micrometers, and can be straight, twisted and curled, or in the form of ropes[8-11]. Their distribution and orientation in the matrix can be uniform and unidirectional or random. All these factors make the simulations of CNT-based composites extremely difficult[1]. For this reason, the concept of unit cells or representative volume elements, which have been applied successfully in the studies of conventional fiber-reinforced composites at the microscale[40-41], has been extended to study the CNT-based composites at the nanoscale. In this RVE approach, a single (or multiple) nanotube(s) with surrounding matrix material can be modeled, with properly applied boundary and interface conditions to account for the effects of the surrounding materials[1]. Numerical methods can be applied to analyze the mechanical responses of these RVEs under different loading conditions.

In this paper the cylindrical RVE is employed to evaluate the effective Young's modulus of the CNT based composites under axial loading condition.

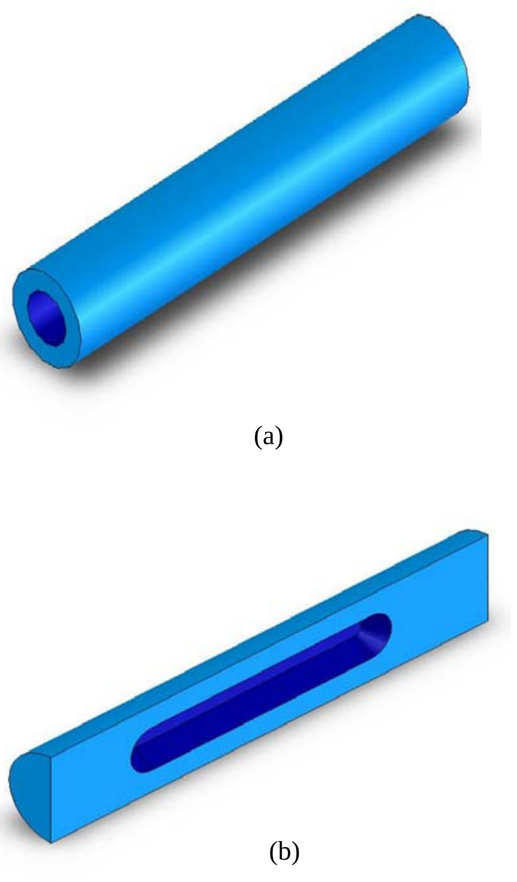

Figure 2. The cylindrical RVE with (a) long CNT; (b) short CNT (cut through view).

FORMULAS FOR EVALUATIONS OF THE EFFECTIVE MATERIAL CONSTANTS

The material of the elasticity model is isotropic and homogenous. According to Hooke's law the stress strain relationship for isotropic material in compliance matrix form is given by,

$$\begin{pmatrix} \varepsilon_x \\ \varepsilon_y \\ \varepsilon_z \end{pmatrix} = \begin{pmatrix} 1 & -v & -v \\ -v & 1 & -v \\ -v & -v & 1 \end{pmatrix} \begin{pmatrix} \sigma_x \\ \sigma_y \\ \sigma_z \end{pmatrix}$$

In this load case the stress and strain components at any point on the lateral surface are:

$$\sigma_x = \sigma_y = 0 \quad \text{and} \quad \varepsilon_z = \frac{L}{\Delta L}$$

$$E_z = \frac{\sigma_z}{\varepsilon_z} = \frac{L}{\Delta L}\sigma_{ave} \tag{1}$$

Where the averaged stress is given by[1],

$$\sigma_{ave} = \frac{1}{A}\int_A \sigma_z\left(x, y, L/2\right) dx\, dy \tag{2}$$

with A being the area of the end surface. σ_{ave} can be evaluated for the RVE using the FEM results. Once the stress is obtained above equation can be applied to estimate effective Young's modulus E_z[1].

ANALYTICAL RESULTS BASED ON THE STRENGTH OF MATERIAL THEORY

CNT through the Length of the RVE

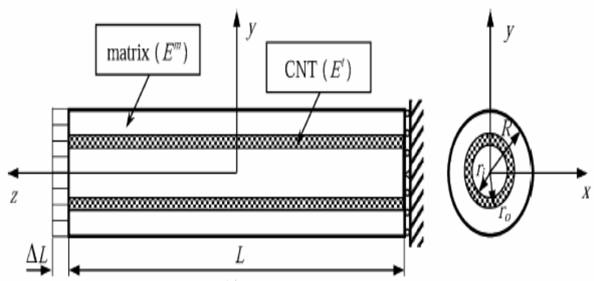

Figure 3. The cylindrical RVE with long CNT

This is the case when the CNT is relatively long (with large aspect ratio) and therefore a segment can be modeled using an RVE. The volume fraction of the CNT is[1]:

$$V^t = \frac{\pi\left(r_o^2 - r_i^2\right)}{\pi\left(R^2 - r_i^2\right)} = \frac{\left(r_o^2 - r_i^2\right)}{\left(R^2 - r_i^2\right)} \tag{3}$$

It is assumed that the matrix and CNT deform independently of each other under the stretch ΔL (Fig. 3). By considering the compatibility of strains and equilibrium of stresses, one obtains the following expression for the effective Young's modulus in the axial direction[1]:

$$E_z = E_t V_t + E^m \left(1 - V^t\right) \quad (4)$$

where E^m and E^t are the Young's modulus of the matrix and CNT, respectively.

CNT inside the RVE

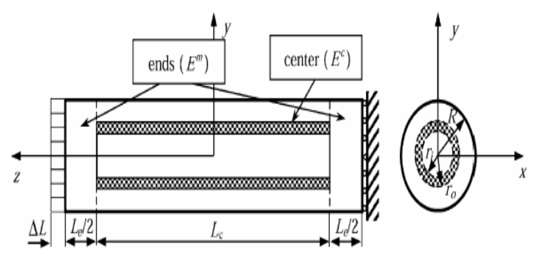

Figure 4. The cylindrical RVE with short CNT

In this case (Fig. 4), the RVE can be divided into two segments: one segment accounting for the two ends with total length L_e and Young's modulus E^m and another segment accounting for the center part with length L_c and an effective Young's modulus E^c [1]. The two hemispherical end caps of the CNT have been ignored in this derivation. Since the center part is a special case of Fig. 3, its effective Young's modulus is found to be[1]

$$E^c = E^t V^t + E^m \left(1 - V^t\right)$$

using Eq. (4), in which the volume fraction of the CNT, V_i^t given by Eq. (3) is computed based on the center part of the RVE (with length L_c) only. Again, by considering the compatibility of strains and equilibrium of stresses, one obtains the following expression for the effective Young's modulus in the axial direction[1]:

$$E_z = \left[\frac{1}{E^m}\left(\frac{L_e}{L}\right) + \frac{1}{E^c}\left(\frac{L_c}{L}\right)\left(\frac{A}{A_c}\right) \right]^{-1} \quad (5)$$

in which the areas $A = \pi R^2$ and $A_c = \pi (R^2 - r^2)$.

FINITE ELEMENT MODELING IN ANSYS

The RVEs are analyzed using the finite element software ANSYS 10.0. In all cases, axi-symmetric FEM models are used since the RVEs have an axi-symmetric geometry and the loading case to be analyzed is axi-symmetric. First the geometry of the model is drawn by generating the keypoints, lines and areas. The respective material is assigned to the model. Then it is meshed by a suitable element. The models in this paper are meshed using plane 82 element, which is a quadratic (8 node) ring element. This element offer better accuracy for axi-symmetric model with axi-symmetric loading. Then proper boundary conditions are applied to them. As the present study involves in the evaluation of effective tensile elastic modulus, one end of the models is made fixed and the other end is given to a fixed displacement value.

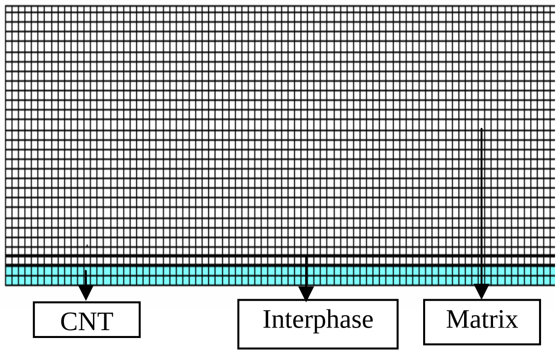

Figure 5. Partial enlarged view of long CNT based composite

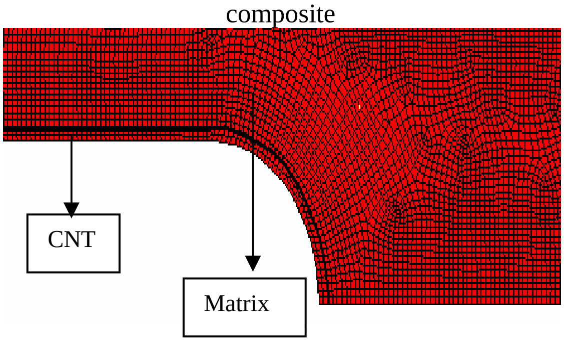

Figure 6. Partial enlarged view of short CNT based composite

After applying the boundary conditions, the solution phases of the finite element analysis are executed. For the present work, the Sparse Direct Solver is used for the solution of the finite element models.

RESULTS AND DISCUSSION

Effect of Interphase Property (Young's Modulus) on the TEM of Nanocomposites for Imperfect Bonding of Long CNT

In the paper, armchair type CNT is taken into consideration, which is specified by chiral indices (m,m). The volume fraction of CNT in the composite is taken to be 5%. All materials, matrix, CNTs, and any interphase region, are assumed to be isotropic linear elastic, subject to small deformations. All boundaries between materials are assumed to have continuous displacements.

To investigate the effect of interphase property (Young's Modulus) variation for long CNT, the cylindrical RVE, as shown in Fig. (3), is studied which shows the CNT all the way through its length. The length *(L)* of both matrix and CNT are 100 nm and the effective thickness (t) of CNT is taken as 0.4 nm. The radius (R) of matrix is 10 nm, the inner radius of CNT is (r_i) 4.6 nm, the outer radius (r_0) of CNT is 5 nm. The length of interphase is 100 nm, inner radius of interphase region is 5 nm and outer radius is 5.2 nm and effective thickness of the interphase is 0.2 nm. The Young's moduli and Poisson's ratios used for the CNT and matrix are:

CNT: $E_t = 1000$ nN/nm^2 (GPa); $\nu^t = 0.3$

Matrix: $E_m = 5$ nN/nm^2 (GPa); $\nu^m = 0.3$

where, the value of the Young's Modulus for the matrix is representative of a polymer. The Young's Modulus of interphase region is varied from 1 to 9 GPa. The interphase region with Young's Modulus less than 5 GPa represent soft polymer, whereas, the interphase with Young's Modulus more than 5 GPa represent harder polymer. The quadratic 8 node ring element is used to mesh the model. One end is made fixed and the other end is given a strain of 1 %. The stress contour plot of first principal stresses under axial stretch in finite element software ANSYS is shown in Fig. 7.

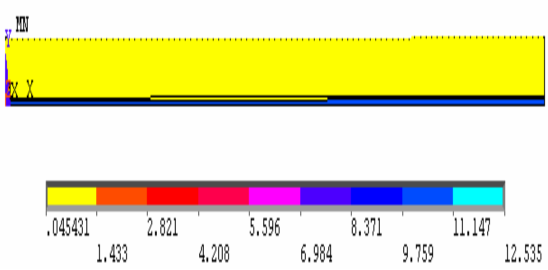

Figure 7. Plot of the first principal stresses for the RVE (long CNT) with 0.2 nm thick interphase under axial stretch.

It is observed that for long CNT based polymer composite, effective tensile elastic modulus (TEM) increases linearly with the increase in Young's Modulus of interphase at a particular volume fraction of the CNT (5%). The results also show that increase of the stiffness of the composite is significant in the axial direction though the rate of increment is not rapid. With the volume fraction of the CNT being at only about 5%, the stiffness of the composite in the axial direction can increase by more than ten times compared with that of the polymer matrix. The presence of a harder interphase (harder than polymer matrix) can result in higher effective properties than the presence of a soft interphase (softer than polymer matrix) of the polymer based CNT composite.

The maximum first principal stresses developed in the polymer based CNT composite also increases with increase of Young's Modulus of interphase (shown in Fig. 9). In this case, the rate of increase is not linear. At first the increase rate is high; the increase rate slowly decreases with the increase of Young's Modulus of the interphase region.

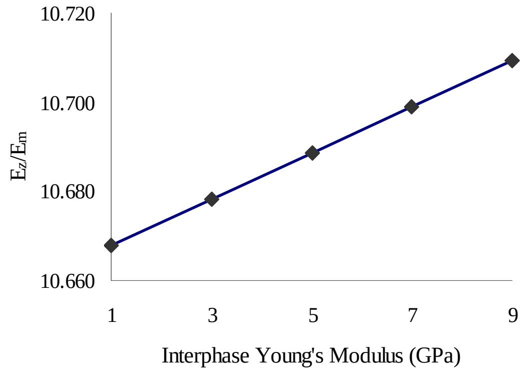

Figure 8. Effect of interphase Young's Modulus variation on the TEM of nanocomposites in case of long fiber (CNT through the length of RVE)

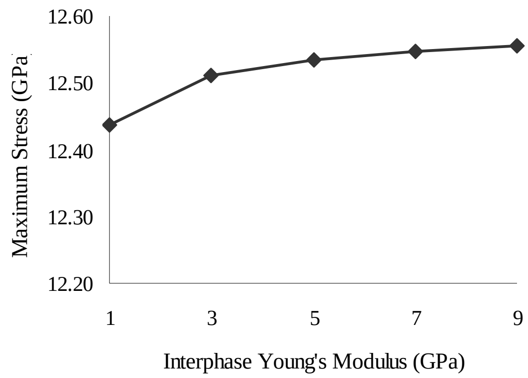

Figure 9. Effect of interphase Young's Modulus variation on the maximum first principal stress of nanocomposites in case of long fiber (CNT through the length of RVE)

Effect of Interphase Property (Young's Modulus) on the TEM of Nanocomposites for Imperfect Bonding (Presence of Interphase) of Short CNT

Next short fiber (CNT inside the RVE) based polymer composite is studied to observe the effect of interphase property (Young's Modulus) variation on the TEM of nanocomposites. The length of the matrix is same as 100 nm but the length of CNT and interphase is changed, which is reduced to 50 nm (with the two hemispherical end caps). The volume fraction is taken as 2%. The dimensions of short fiber based composites are given below:

Matrix: Length, L = 100 Nm; Radius, R = 10 Nm.

CNT: Length, L = 50 nm; Outer radius, r_o = 5 nm; Inner radius, r_i = 4.6 nm (effective thickness = 0.4 nm).

Interphase: Length = 100 nm; Outer radius, r_o = 5 nm; Inner radius, r_i = 5.2 nm (effective thickness = 0.2 nm).
The Young's moduli and Poisson's ratios used are:
CNT: E_t = 1000 nN/nm^2 (GPa); v^t = 0.3
Matrix: E_m = 5 nN/nm^2 (GPa); v^m = 0.3

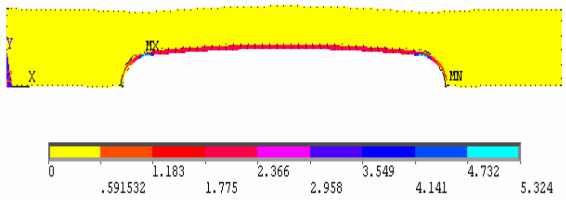

Figure 10. Plot of the first principal stresses for the RVE (short CNT) with 0.2 nm thick interphase under axial stretch

The stress contour plot of first principal stresses under axial stretch in finite element software ANSYS is shown in Fig. 10.

It is observed that for short CNT based polymer composite, effective tensile elastic modulus (TEM) increases with the increase in Young's Modulus of interphase. The rate of increase seems to decline with the increase of interphase Young's Modulus. With the volume fraction of the CNT being at about 2%, the stiffness of the composite in the axial direction can increase by more than 1.7 times compared with that of the polymer matrix. The presence of a soft interphase (softer than polymer matrix) results in lower effective property than the presence of a harder interphase (harder than polymer matrix). Hard interphase can improve the effective property of the polymer based CNT composite slightly.

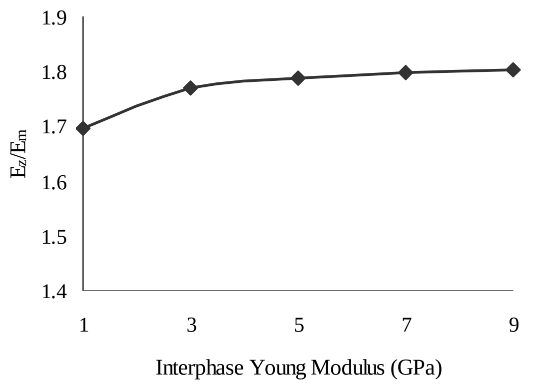

Figure 11. Effect of interphase Young's Modulus variation on the TEM of nanocomposites in case of short fiber (CNT inside the RVE)

The maximum first principal stresses developed in the polymer based CNT composite decreases with the increase of Young's Modulus of interphase. The Fig. 12 shows the variation of the maximum first principal stress with the interphase property. In this case, the rate of decrease is not linear. The decrease rate is high when the Young's Modulus is changed from 1 to 3 GPa; the decrease rate slowly lessens with the increase of Young's Modulus of the interphase region.

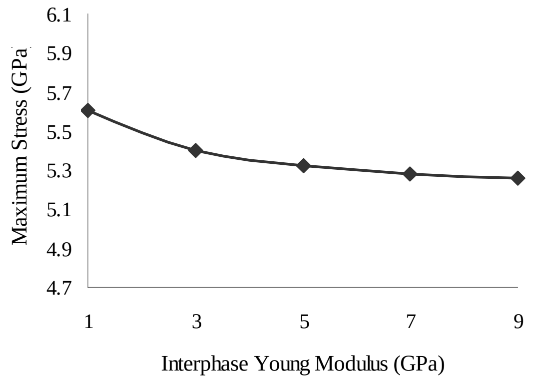

Figure 12. Effect of interphase Young's Modulus variation on the maximum first principal stress of nanocomposites in case of short fiber (CNT inside the RVE)

Effect of Matrix Material on the TEM of Nanocomposites for Perfect Bonding (No Interphase) of Long CNT

To investigate the effect of matrix material variation for long CNT, the cylindrical RVE, as shown in Fig. 2(a), is studied which shows the CNT all the way through its length. No interphase is considered between matrix and CNT i.e. the bonding is perfect. The dimensions of the cylindrical RVE are:
Matrix: Length, L = 100 nm; Radius, R = 10.4 nm
CNT: Length, L =100 nm; Outer radius, r_o = 5.4 nm; Inner radius, r_i = 5 nm (effective thickness = 0.4 nm).
The Young's Moduli and Poisson's ratio used are:
CNT: E_t =1000 nN/nm^2 (GPa); v^t =0.3
Matrix: E_m = variable; v^m = 0.3

Due to axi-symmetric of the model, one half of the model is considered to perform the finite element analysis.

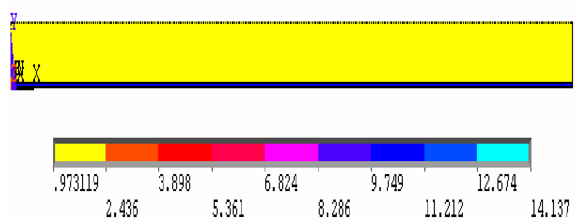

Figure 13. Plot of the first principal stresses for the RVE (long CNT) with no interphase under axial stretch

The quadratic 8 node ring element is used to mesh the model. One end is made fixed and the other end is given a strain of 1 %. The stress contour plot of first principal stresses under axial stretch in finite element software ANSYS is shown in the Fig. 13. Calculated E_z/E_m using FEM and Eq. (4) for respective value of E_t/E_m are shown in Table 1. The strength of materials solutions for the effective Young's modulus E_z, using Eq. (4), are quite close to the FEM solutions which are based on 3-D elasticity, with the difference less than 0.06%.

Table 1. Computed effective material constant for CNT through RVE

E_t/E_m	E_z/E_m	
	FEM	***Equation (4)***
5.263	1.214	1.2133
8.333	1.368	1.3668
9.091	1.406	1.4045
11.111	1.507	1.5055
14.286	1.665	1.6643
16.667	1.784	1.7833

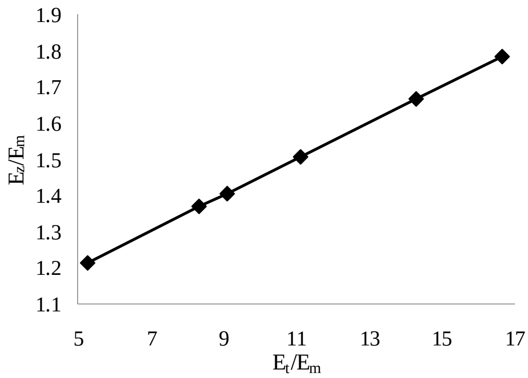

Figure 14. Effect of matrix material variation on the TEM of nanocomposites in case of long fiber (CNT through the length of RVE)

It is observed that, effective tensile elastic modulus (TEM) increases linearly with the increase in Young's Modulus of matrix at a particular volume fraction of the CNT (5%).

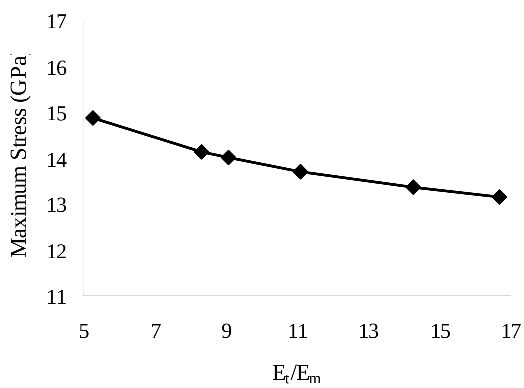

Figure 15. Effect of matrix material variation on maximum first principal stress of nanocomposites in case of long fiber (CNT through the length of RVE)

The results also show that change of the stiffness of the composite is significant in the axial direction. The ratio E_z/E_m changes from 1.2 to 1.8 as the Young's Modulus varies from 60 to 190 GPa.

The maximum first principal stresses developed in the long CNT composite decreases almost linearly with increase in Young's Modulus of matrix. The Fig. 15 shows the variation of the maximum first principal stress with the matrix property.

Effect of Matrix Material on the TEM of Nanocomposites for Perfect Bonding (No Interphase) of Short CNT

A perfect bonding is assumed between CNT and matrix i.e. there is no interphase region. The volume fraction is taken as 2%. The dimensions of the RVE are:

Matrix: Length, L = 100 nm; Radius,R = 10.4 nm

CNT: Length, L = 50 nm; Outer radius, r_o = 5.4 nm; Inner radius, r_i = 5 nm (effective thickness = 0.4 nm).

The Young's Moduli and Poisson's ratio used are:

CNT: E_t =1000 nN/nm^2 (GPa); ν^t =0.3

Matrix: E_m = variable; ν^m = 0.3

The stress contour plot of first principal stresses under axial stretch in finite element software ANSYS is shown in Fig. 16.

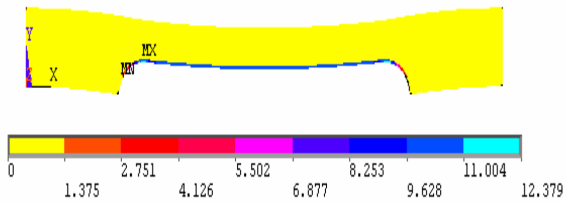

Figure 16. Plot of the first principal stresses for the RVE (short CNT) with no interphase under axial stretch

Table 2 shown below represents that, the strength of materials solutions for the effective Young's modulus E_z, using the extended rules of mixtures (Eq. (5)), are quite close to the FEM solutions which are based on 3-D elasticity, with the difference less than 3.4%.

Table 2. Computed effective material constant for CNT inside the RVE

E_t/E_m	E_z/E_m	
	FEM	***Equation (5)***
5.263	0.933099	0.96520
8.333	1.007356	1.02475
9.091	1.02405	1.03841
11.111	1.06585	1.07302
14.286	1.124234	1.12265
16.667	1.163656	1.15652

It is observed that for short CNT based composite, effective tensile elastic modulus (TEM) increases with the increase in Young's Modulus of matrix at a particular volume fraction of the CNT (2%).

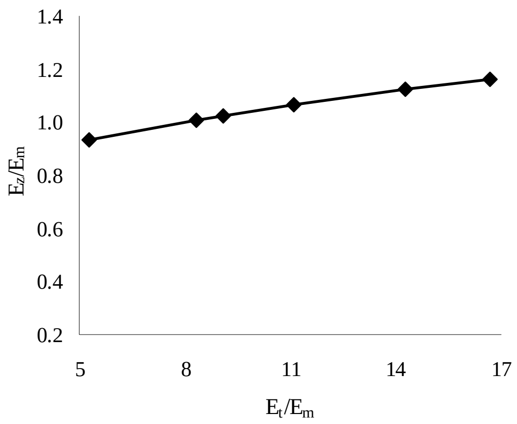

Figure 17. Effect of matrix material variation on the TEM of nanocomposites in case of short fiber (CNT inside the RVE)

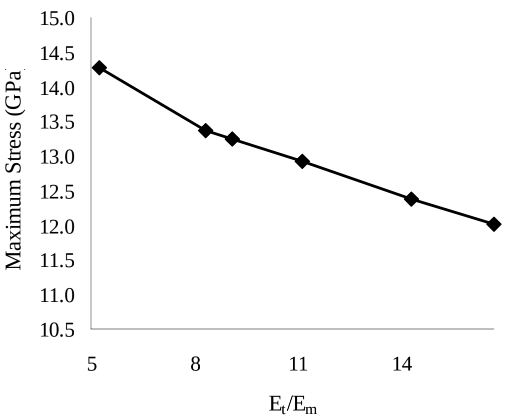

Figure 18. Effect of matrix material variation on the maximum first principal stress of nanocomposites in case of short fiber (CNT inside the RVE)

The results also show change of the stiffness of the composite is noteworthy in the axial direction. The ratio E_z/E_m changes from 0.9 to 1.2 as the Young's Modulus varies from 60 to 190 GPa.

The maximum first principal stresses developed in the long CNT composite decreases almost linearly with increase Young's Modulus of matrix, shown in Fig. 18.

It is observed that, for both long and short CNT based composite, a harder matrix results in higher effective property than softer matrix.

CONCLUSIONS

From the above results obtained from theoretical interpretation and performed simulation, it can be concluded that:

- For long CNT based polymer composite, effective tensile elastic modulus increases linearly with the increase in Young's modulus of interphase for imperfect bonding between CNT and matrix.
- For short CNT based polymer composite (CNT inside the RVE), effective tensile elastic modulus increases with the increase in Young's modulus of interphase for imperfect bonding between CNT and matrix. The rate of increase is not constant in this case.
- For long CNT based composite, effective tensile elastic modulus increases linearly with increased Young's Modulus of matrix for perfect bonding between CNT and matrix.
- For short CNT based composite, effective tensile elastic modulus increases linearly with increased Young's Modulus of matrix for perfect bonding between CNT and matrix.

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