

EFFECT OF INTERPHASE CHARACTERISTIC AND PROPERTY ON AXIAL MODULUS OF CARBON NANOTUBE BASED COMPOSITES

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Abstract: In carbon nanotube (CNT) based composite, due to the small (micrometer) size of reinforcements a large amount of interphases is developed during the time of production. It is important to assess whether the interphase is responsible for the poor mechanical properties of CNT-reinforced composite. In this research, the effect of interphase property and characteristics on effective mechanical properties of CNT based composites is evaluated using a 3-D nanoscale representative volume element (RVE). The effect of both soft and stiff interphases on the Tensile Elastic Modulus (TEM) of nanocomposites is investigated using the Finite Element Method (FEM) for the case of both long and short CNTs. With the increase of thickness of stiff interphase, the stiffness of the composite increases significantly for both the short and long CNT cases. On the other hand the increase of thickness of soft interphase reduces the stiffness of the overall composite in a considerable amount.

Key Words: Carbon nanotube, Interphase, Representative Volume Element, Finite Element method, Tensile Elastic Modulus.

INTRODUCTION

Carbon nanotube exhibits superior physical and mechanical properties which makes them excellent candidates for the reinforcement of high performance structural composites¹. Research work on carbon nanotubes and their related polymer based composites can be described into four particular scopes², such as: (i) production of highly pure and controllable CNTs in terms of their size, length and chiral arrangement; (ii) alignment of the nanotubes and their dispersion in the matrix; (iii) improvement of interfacial bonding strength between the nanotubes and their surrounding matrix and (iv) Real life applications of carbon nanotubes. Among these the third scope is very much important because poor interphase can degrade the overall quality of nanocomposites.

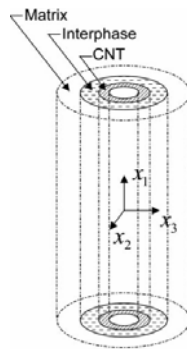


Figure 1: Interphase zone between the matrix and CNT for a cylindrical model⁴

Reinforcement effects of CNTs on polymer, depend not only on CNT properties and CNT weight fraction, but also on their dispersion in the matrix³. This dispersion zone is the critical region to predict known as interphase between the matrix and CNT. Figure 1 shows the interphase zone between the matrix and CNT for a cylindrical model.

Nomenclature

CNT	Carbon nanotube
E	Modulus of elasticity
FEM	Finite Element Method
K	Global stiffness matrix
MD	Molecular dynamics
r	Radial component of polar co-ordinate system
RVE	Representative Volume Element
R_{NT}	Radius of carbon nanotube
SWNT	Single-walled nanotube
MWNT	Multi-walled nanotube
TEM	Tensile elastic modulus
VF	Volume fraction
ν	Poisson' ratio
ϵ	Normal strain
γ	Shear strain
τ	Shear stress
σ	Normal stress

One of the main challenges in carbon nanotube based composite is to improve the dispersion of CNTs in a polymer matrix. There are several techniques to do this. Among these physical blending optimization, melt mixing, solution processing, chemical bonding and chemical functionalization of CNTs are the main. Thus four types of interactions possible between CNTs and the polymer matrix³, such as: (i) CNTs dispersed freely in the polymer; (ii) interfacial bonding between the matrix and CNTs; (iii) polymer absorption or assembly on the CNT surface in case of polymer-coated CNTs and (iv) covalent functionalization and polymer grafting on nanotubes.

Interphase is inevitable in the production of polymer matrix composites due to several reasons. Among these the presence of absorbed contaminants on the fiber surface, 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 are mainly responsible for interphase generation⁵. Interphase effect on the performance of composite is not well characterized, since its precise nature is difficult to predict⁶. Both Scanning Force microscopy (SFM) and Scanning probe microscopy (SPM)⁷⁻⁹ have been used to examine the mechanical and thermal properties of nanocomposites in the immediate vicinity of reinforcing carbon nanotubes. It is also established experimentally that an interphase can be both soft and stiff compared to the matrix¹⁰.

The effect of interphase regions on the effective mechanical properties of composites has been investigated using a variety of techniques but mostly in the context of coated fiber inclusions. Recently, the interfacial bonding strength of CNT based composites has been calculated by Wagner¹¹, Lau¹², Haque and Ramasetty¹³, and Gao and Li¹⁴ using fundamental shear lag models. From their investigation it is found that the single walled zigzag nanotubes would induce higher interfacial bonding stress at both bonded end regions. Previously in most molecular dynamics model, nonbond interaction between carbon nanotube and polymer matrix was considered¹⁵. But Frankland et al.¹⁶ shows that even a relatively low density of cross-links between fiber and matrix can have a large influence on the properties of nanotube/polymer interface. Another possible ways to improve the bonding strength between the CNTs and the matrix is to make use of the nano-mechanical interlocking of the nanotubes². Recently, from the investigation of Lu et al.^{17,18} it was obtained that adding a small amount of coiled carbon nanotubes, instead of the straight ones, to the polymer-based materials can improve the thermal and mechanical properties of composite, as

well as the bonding strength at the interface. The effects of an interphase layer between the nanotubes and the polymer matrix as result of functionalization is also investigated using a multi-layer composite cylinders approach¹⁹.

Thus effective properties associated of CNT based composites can be greatly changed by the presence of an interphase region, particularly if that interphase region is a compliant interphase and therefore representative of poor load transfer from the matrix to the fiber. In this research, the effect of interphase property and characteristics on the effective mechanical properties of nanocomposites is investigated using a 3-D representative volume element based on continuum mechanics and using the FEM. To do this, interphase thickness is varied relative to matrix and CNT thickness, to examine the effect on the TEM of polymer composites in case of both soft and stiff interphases. Both long and short CNT models are examined to observe this effect.

Various previous researches has explored interphase thickness, volume fraction, poisson's ratio and stiffness effect on the axial modulus, transverse modulus, axial shear modulus and transverse shear modulus of carbon nanotube based composites^{4, 5, 19, 20}. But these are not adequate. In previous studies, in order to investigate interphase thickness effect, thickness is widely varied between 0.2 nm to 3 nm²⁰. But in most of the cases, interphase thickness varies from 0.5 times to 4 times the CNT thickness^{2,4}. In the present study interphase thickness is varied from 0.5 times to 2.0 times CNT thickness (which is a thin interphase) in order to investigate whether thin interphase thickness variation significantly affect the TEM of nanocomposites and the range considered here is also reasonable^{2, 4}.

In previous studies, in order to investigate interphase thickness effect, high TEM of the matrix has been considered that corresponds to the thermosetting plastic²⁰. In this work the matrix material and interphase stiffness is varied within a narrow range and is taken around 2.5 to 15 GPa which is corresponding to thermoplastic polymer (thermoplastic ethylene, polypropylene etc.). Here comparatively low value of TEM of matrix and interphase stiffness is considered; as a result interphase thickness effect will be conspicuous. Previously research has been conducted by considering the interface thickness effect on TEM of CNT based nanocomposites²⁰. But in this research, the effect of interphase relative stiffness to matrix (E_i/E_m) is considered instead of interphase stiffness in order to understand interphase-matrix interaction more clearly. Finally a deductive comparison is made between soft and stiff interphase to show how they are affected by interphase thickness variation. Finite element modeling and analysis has been done in ANSYS

10.0 and FEM results are verified using an extended rule of mixture theory based on mechanics of elasticity. 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 and traction.

ANALYSIS

A comprehensive approach combining analytical, experimental and computational methods need to be adopted to tackle the multiscale, multiphysics problems in the development of nanocomposites²¹. At the nanoscale, analytical models are difficult to establish and experiments are expensive to conduct but modeling can be done effectively and efficiently^{22, 23}. Molecular Dynamic (MD) simulations generally take very long computational time and require powerful computer facilities, which inevitably create a barrier for adopting this technique for practical applications. In the past few years other models such as continuum models, constitutive models, equivalent-truss models and MD associated with FE models²⁴ have appeared gradually²⁵. Equivalent-continuum models are based on the equilibrium molecular structure obtained from the MD simulations and are used to predict the bulk mechanical behavior of nano-structured materials. The current results using the continuum approaches have indicated that continuum mechanics can be applied to models with dimensions of a few hundred nanometers and larger, where averaging of material properties can be done properly for CNTs. Liu and Chen²⁴ have applied the concept of representative volume elements (RVEs) to extract the mechanical properties of the nanotube/ polymer composites based on 3D elasticity theory and FEM. In the RVE approach, a single walled nanotube with surrounding polymer matrix is modeled, with properly applied boundary and interface conditions to account for the effects of the surrounding matrix (Fig. 1). As this RVE model is very much effective in the study of interactions of the nanotubes with the matrix and for the evaluation of the effective materials properties of the nanocomposites; in this paper the RVE model is taken into consideration. Three nanoscale RVEs based on the 3-D elasticity theory have been proposed by Liu and Chen²⁶ for the study of CNT-based composites are shown in Fig. 2.

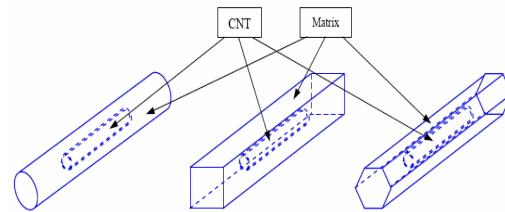


Figure 2: Three possible RVEs for the analysis of CNT-based composites.

The three types of RVEs are cylindrical RVE, square RVE and hexagonal RVE. The square RVE models are applied when the CNTs are arranged evenly in a square array, while the hexagonal RVE models are applied when CNTs are in a hexagonal array, in the transverse direction. In this paper the cylindrical RVE is applied to model the CNTs which have different diameters²⁷ and CNTs embedded in a regular matrix material. For any case of axisymmetric or antisymmetric loading, a 2-D axisymmetric model for the cylindrical RVE can reduce the computational work significantly²⁴. The geometry of the elasticity based model can be compared to a hollow cylindrical RVE model with length L, inner radius r_i and outer radius R. This geometry represents the case when the CNT is relatively long and thus all the way through the length of the RVE. In the case that the CNT is relatively short and thus fully inside the RVE, a solid cylindrical RVE (r_i=0) is considered for extracting the material constant, because elasticity solutions are difficult to find in this case²². The model of long fiber through the length of RVE and short fiber inside the RVE is shown in Fig. 3.

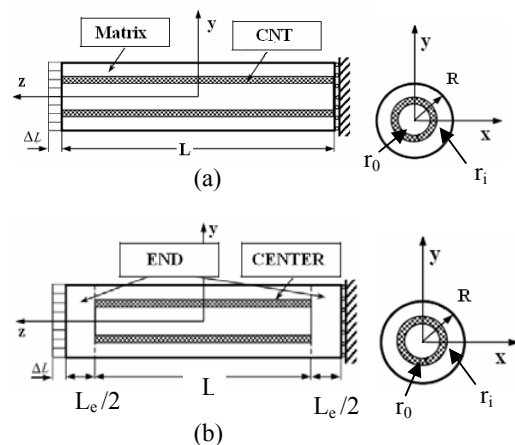


Figure 3: Cylindrical RVE with long and short CNT

For axial loading, the stress and strain components at any point on the lateral surface are²⁴,

$$\sigma_r = \sigma_\theta = 0$$

$$\varepsilon_z = \frac{\Delta L}{L} \quad (1)$$

$$\varepsilon_\theta = \frac{\Delta R_a}{R} \quad (2)$$

$$E_z = \frac{\sigma_z}{\varepsilon_z} = \frac{L}{\Delta L} \sigma_{avg} \quad (3)$$

where the average stress is given by

$$\sigma_{avg} = \frac{1}{A} \int_A \sigma_z(x, y, L/2) dx dy \quad (4)$$

Here A is the area of end surface. Equation (4) can be evaluated using the finite element results from ANSYS 10.0. The rule of mixture theory can be applied to verify the numerical results for the effective Young's moduli in the CNT axial direction. The volume fraction of the CNT (V_f) is given by following equation²³.

$$V_f = \frac{V_c - V_i}{V_c - V_i} \quad (5)$$

The longitudinal tensile modulus of composite for the case of long CNT (Fig. 3(a)),

$$E_z = E_f V_f + E_m (1 - V_f) \quad (6)$$

In short fiber case (Fig. 3(b)), RVE model can be considered as two segments, one accounting for the center portion of the matrix considering CNT throughout the length (neglecting the end caps) for calculating E_z of the center part, and the other segment accounting for the two ends of the RVE without CNT with modulus E_m . From the Fig. 3(b), the center part of the RVE is assumed to be a long CNT case neglecting the hemispherical end caps, and the effective modulus E_c of this segment is found to be²⁴

$$E_c = E_f V_f + E_m (1 - V_f) \quad (7)$$

Effective modulus (E_z) for the short fiber reinforced composite using the extended rule of mixtures²⁶ can be obtained from following expression²⁴

$$E_z = \frac{1}{\frac{1}{E_m} \left(\frac{L_c}{L} \right) + \frac{1}{E_c} \left(\frac{L_c}{L} \right) \left(\frac{A}{A_c} \right)} \quad (8)$$

which was derived from the strength of materials theory, in which, $A = \pi R^2$ and $A_c = \pi (R^2 - r^2)$

MODELING

In this paper the cylindrical RVE (Fig.1, 2) for a single-walled carbon nanotube in a matrix material is studied using the finite element method. The deformations and stresses are computed for the axial loading cases (Fig. 3). Two cases are studied,

one on a RVE with a long CNT and the other on a RVE of the same size but with a short CNT. In all the cases, axisymmetric FEM models are used since the RVEs have an axisymmetric geometry and the loading case to be analyzed is also axisymmetric. Quadratic 8-node ring elements (known as PLANE 82 element in ANSYS 10.0.) for axisymmetric problem is employed, which are second order elements and offer better accuracy in stress analysis. Mapped meshing using solid elements has been used as it offers good results compared to the tetrahedral and triangular elements in the FEM analysis. In the present study one end is made fixed and the other end is given to a fixed displacement value. The FEM results are then processed and Eqs. (4) and (5) are applied to extract the effective Young's modulus for the CNT-based composite. Maximum first principal stress is also found from the finite element solution software.

Modeling for Interphase Thickness Effect

To investigate the effect of interphase thickness on CNT-based composites first long CNT through the length of RVE similar to the one shown in Fig. 3(a) is studied. The length (L) of matrix, CNT and interphase is 100 nm and the effective thickness (t) of CNT is 0.4 nm in this case²³. In short CNT RVE model, CNT is not through out the matrix. Its length is less than the RVE model (Fig. 3(b)). In this case the RVE length is 100 nm and CNT length is 50 nm (including two hemispherical end caps). In order to make 5 % (approximately) CNT volume fraction inner radii (r_i), outer radii (r_o) and the radii of matrix (R) are considered as 4.60 nm, 5.00 nm and 10.00 nm respectively. Effective thickness of the interphase is varied between 0 to 0.8 nm. As a result volume fraction of interphase inside the matrix varies from 0% to 10 % (approximately). Here the zero interphase is the perfect bonding case and the CNT with interphase is the imperfect bonding case. The layer of interphase region is between matrix and CNT. The Young's moduli and Poisson's ratios used for the CNT, matrix and interphase are:

$$\begin{aligned} \text{CNT: } E_t &= 1000 \text{ nN/nm}^2 \text{ (GPa); } \nu^t = 0.3 \\ \text{Matrix: } E_m &= 10 \text{ nN/nm}^2 \text{ (GPa); } \nu^t = 0.3 \\ \text{Interphase: } E_i &= 5 \text{ nN/nm}^2 \text{ (GPa); } \nu^t = 0.3 \end{aligned}$$

Here the value of the Young's Modulus for the matrix is representative of a polymer. The interphase region young's modulus is less than the matrix region which represent the soft interphase case. For stiff interphase case Young's moduli and poisson's ratios are:

$$\begin{aligned} \text{Matrix: } E_m &= 5 \text{ nN/nm}^2 \text{ (GPa); } \nu^t = 0.3 \\ \text{Interphase: } E_i &= 10 \text{ nN/nm}^2 \text{ (GPa); } \nu^t = 0.3 \end{aligned}$$

The dimensions and material constants used here are chosen within the wide ranges of those CNTs as reported in various previous literature^{22, 25, 27, 28}. Moreover it can be modified or fine-tuned readily for any specific case in future simulations. To deliver converged FEM result two layers of elements are used for the CNT in the mesh. Small elements compare to CNT are used for the matrix to ensure the connectivity and to avoid elements with large aspect ratios. Finite element mesh for the case of long and short CNT is shown in Fig. 4 & 5.

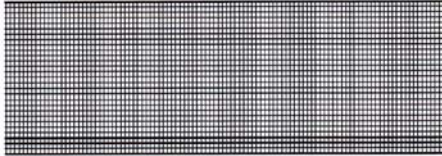


Figure 4: Partial view of the finite element mesh of the long CNT-based model with 0.4 nm CNT thickness and 0.2 nm Interphase thickness.

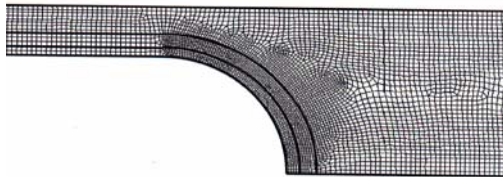


Figure 5: Partial view of the finite Element mesh of the short CNT based model with 0.4 nm CNT thickness and 0.8 nm thick Interphase.

To validate the FEM results, strength of material solutions based on extended rules of mixture theory are extracted from Eqn. (6) and (8). The strength of material solutions are very close to those using the FEM because the geometry and loading condition used here is simple comparatively. In case of long fiber and a soft interphase thickness of 0.4 nm the ratio of E_z/E_m obtained from FEM solution is 5.7966. The strength of material solution obtained from Eqn. (6) is 5.7949 which is very close to the FEM result. Similarly for all other cases FEM results are checked with strength of material solution in order to ensure validity of the results.

Modeling for Interphase Stiffness Effect

To investigate the effect of interphase relative stiffness to matrix (E_i/E_m) on the effective material constants of CNT-based composites, the length (L) of matrix, CNT and interphase is taken as 100 nm for the case of long CNT²³. For short fiber the length (L) of CNT and interphase is considered 50 nm along with two end caps. The effective thickness (t) of CNT is taken as 0.4 nm and the effective thickness of interphase is also taken as 0.4 nm in order to ensure a volume fraction of CNT of 5% (approximately). Interphase

volume fraction is also 5% in this case. The dimensions are:

Matrix: Length, L=100 nm; Radius, R = 10 nm

CNT: Length, L = 100 nm; Outer radius, $r_0 = 5$ nm; Inner radius, $r_i = 4.6$ nm (Effective thickness = 0.4 nm)

Interphase: Length, L = 100 nm; Outer radius, $r_1 = 5.4$ nm; Inner radius, $r_i = 5$ nm (Effective thickness = 0.4 nm)

The Young's moduli and Poisson's ratios used are:

CNT: $E_t = 1000$ nN/nm² (GPa); $\nu^t = 0.3$

Matrix: $E_m = 5$ nN/nm² (GPa); $\nu^t = 0.3$

The relative stiffness of interphase than matrix (E_i/E_m) is taken as 0.5, 1.0, 1.5, 2.0 and 2.5. The FEM results obtained in this way is then compared with the results of strength of material theory. In case of long fiber with a relative stiffness of interphase 2.5, the ratio of E_z/E_m obtained from FEM solution is 10.78. The strength of material solution obtained from Eqn. (6) is 11.025 which is close to the FEM result. Similarly for all other cases FEM results are checked with strength of material solution in order to ensure validity of the results.

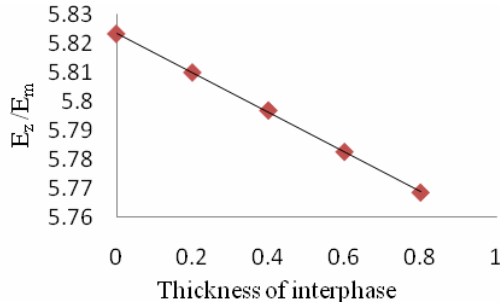
RESULTS AND DISCUSSION

Effect of Soft Interphase Thickness

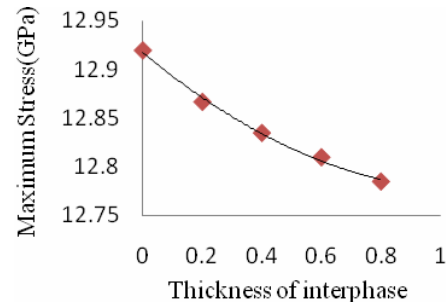
From the investigation and simulation results obtained from FEM it is found that for both long and short CNT based polymer composite effective tensile modulus (TEM) of elasticity decreases with increase in the thickness of the soft interphase (Fig. 6(a, c)).

With the increase of interphase thickness, volume fraction of interphase also increases from 0 to 10.95% in the composite. Stiffness (TEM) of this interphase is less than the matrix (soft interphase). The increased percentage of soft interphase decreases the stiffness of composite in this case. For long fiber the TEM of nanocomposites decreases almost 1% with the increase of only 0.8 nm interphase thickness. But for the short CNT, the TEM of composite decreases to 3.86% with the increase of 0.8 nm interphase thickness. It seems interphase thickness affects short CNT more than the long CNT.

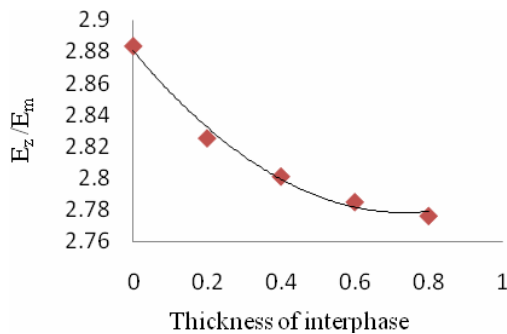
The maximum first principal stresses developed in the polymer based CNT composite also decreases with the increase of the thickness of the soft interphase for both the case of long and short CNT. The Figure 6(b, d) shows the variation of the maximum first principal stresses with the thickness of the soft interphase. Soft interphase load carrying and transfer capability is less than the matrix. As a result maximum first principal stresses of nanocomposites decreases in this case.



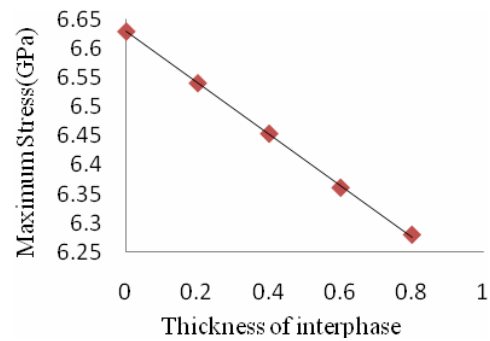
(a) Effect of Interphase thickness on the TEM of nanocomposites in case of long fiber



(b) Effect of Interphase thickness on the maximum first principal stresses of nanocomposites in case of long fiber



(c) Effect of Interphase thickness on the TEM of nanocomposites in case of short fiber



(d) Effect of Interphase thickness on the maximum first principal stresses of nanocomposites in case of long fiber

Figure 6: Effect of Soft interphase on TEM and maximum first principal stress of nanocomposites

Effect of Stiff Interphase Thickness

For long CNT based polymer composite effective tensile modulus (TEM) of elasticity increases with increase in the thickness of the stiff interphase (Fig. 7(a)). As the stiff interphase elastic modulus is higher than matrix, with the increase of its volume fraction (0% to 10.95%) in nanocomposites the TEM of composite increases in this case.

For short CNT based composite the TEM of composite also increases with the increase of interphase thickness (Fig. 7(c)). Increase of the stiffness of the composite is significant in the axial direction though the rate of increment of the interphase thickness is not rapid. While the increase in TEM is only 1.1 % for long CNT it is 4.03 % for short CNT. It seems interphase thickness affects short CNT composites more than long CNT based composites. The maximum first principal stresses developed in nanocomposites increases with the increase of stiff interphase

thickness for the case of long CNT. The rate of increase of the maximum first principal stresses decline with the increase of the thickness of stiff interphase (Fig. 7(b)).

But the maximum first principal stresses developed in the short CNT based composite decreases with the increase of the thickness of the stiff interphase. As the cross sectional area of interface between CNT and matrix increases with the increase in interphase thickness, it results in larger end caps. These larger end cap areas take more transverse load than the smaller end caps though the load in the RVE is axial. This may lead to lower maximum first principal stresses of the composite with the increase in interphase thickness. The Figure 7(d) shows the variation of the maximum first principal stresses with the thickness of the stiff interphase.

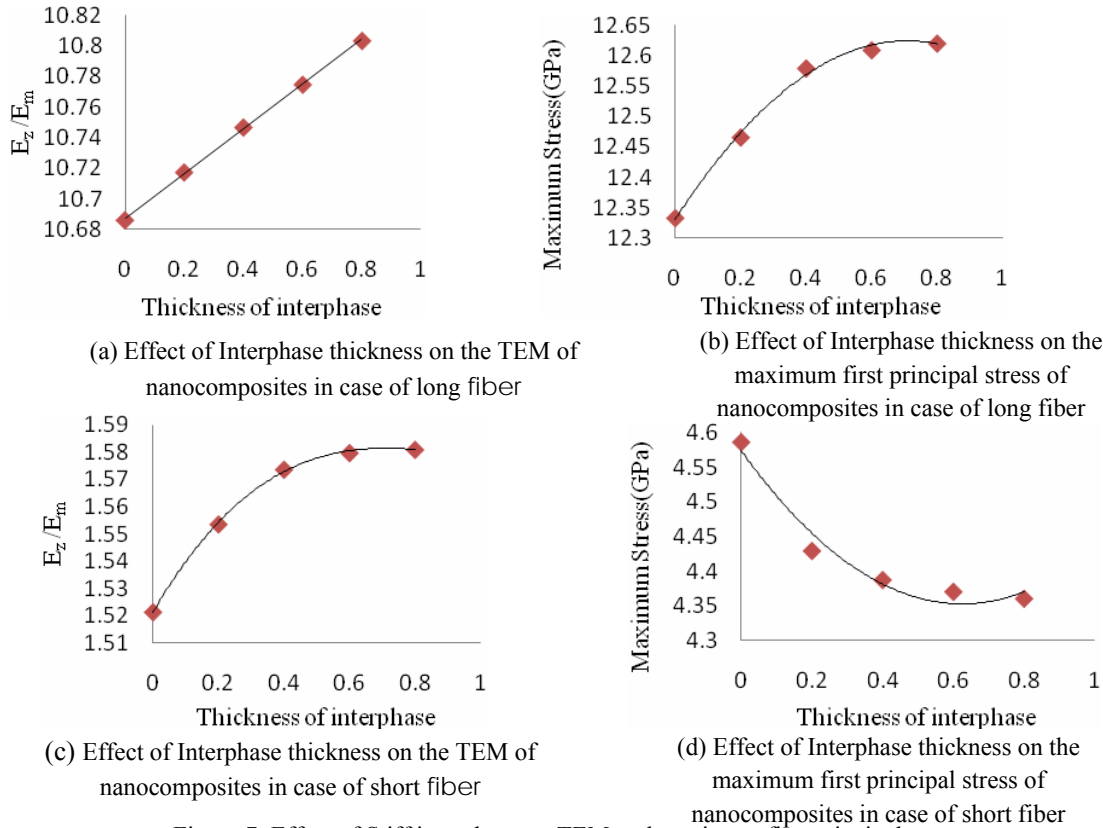
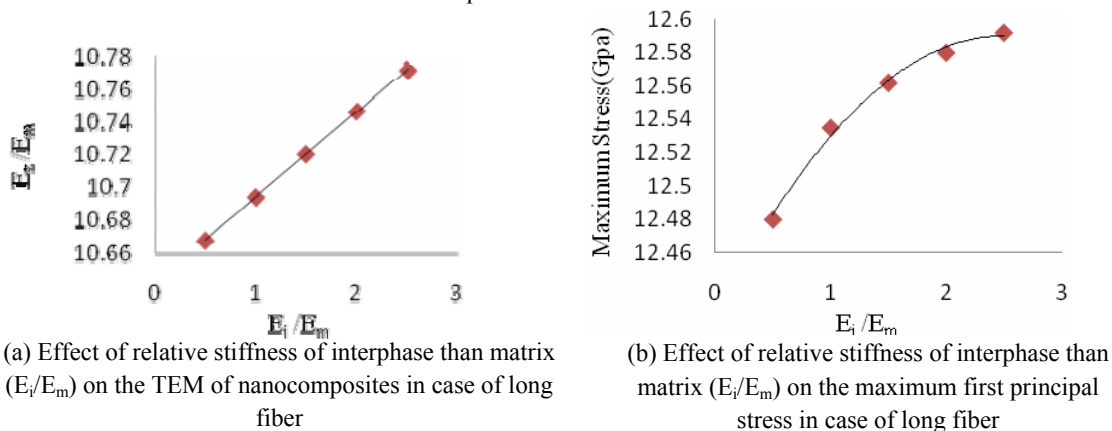


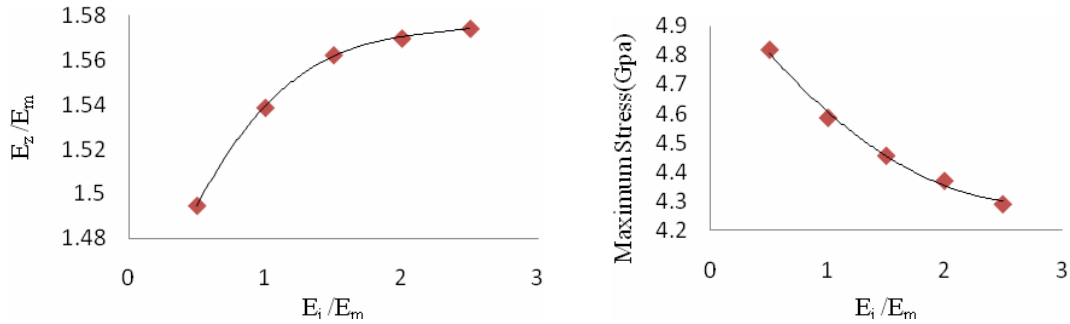
Figure 7: Effect of Stiff interphase on TEM and maximum first principal stress

Effect of Interphase Relative Stiffness to Matrix (E_i/E_m) on Nanocomposites

The effective tensile modulus (TEM) of elasticity of nanocomposites increases with increase in the relative stiffness of the interphase with the matrix for both the case of long and short CNT (Fig. 8 (a, c)). The results show that increase of the stiffness of the composite for long fiber is 1 % and for short fiber it is 5.33 % for the increase of interphase relative stiffness 0.5 to 2.5. The maximum first principal stresses developed in the polymer based CNT composite increases with increase in the relative stiffness of the interphase

with the matrix for long fiber but decreases for short fiber (Fig. 8(b, d)). The rate of decrease of the maximum first principal stresses declines with the increase in E_i/E_m for short CNT. The reason behind it, short fiber is not efficient load carrier like long CNT. Increase in interphase thickness results larger contact surface with matrix and larger end caps. As a result load carrying capacity of the composite decreases. Figure 8(b, d)) shows the variation of the maximum first principal stresses with increase in the relative stiffness of the interphase.





(c) Effect of relative stiffness of interphase than matrix (E_i/E_m) on the TEM of nanocomposites in case of short fiber

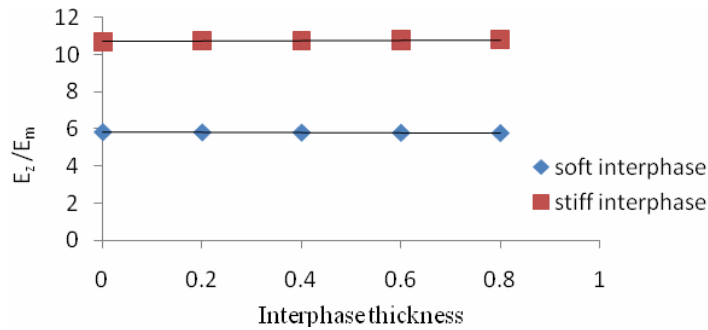
(d) Effect of relative stiffness of interphase than matrix (E_i/E_m) on the maximum first principal stress in case of short fiber

Figure 8: Effect of relative stiffness of interphase than matrix (E_i/E_m) on the TEM and maximum first principal stress.

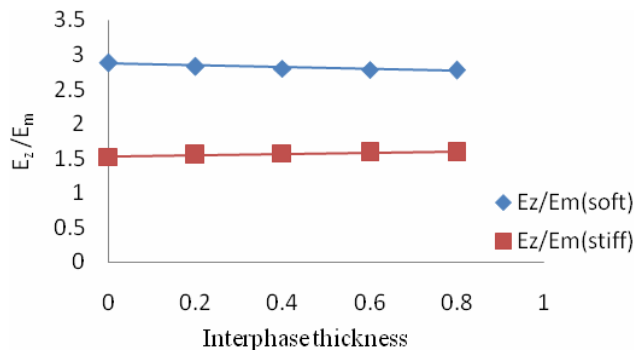
Comparison Between Soft and Stiff Interphase

From the results it is found that, the TEM of composite increases in case of stiff interphase but decreases for soft interphase with the increase of interphase thickness. The results are

similar for both the long and short CNT based composite. It is also observed that short fiber is more sensible than long fiber. The results are shown in Fig. 9.



(a) Comparison Between soft and stiff interphase CNT Model (E_z/E_m Vs Thickness of interphase) for long CNT



(b) Comparison Between soft and stiff interphase CNT Model (E_z/E_m Vs Thickness of interphase) for short CNT

Figure 9: Comparison between soft and stiff interphase CNT Model

All these results suggest that interphase thickness and volume fraction has significant effect on nanocomposites but short CNT based composite is induced more by the thickness variation.

Moreover increase in interphase stiffness can increase the stiffness of overall composite.

CONCLUSION

In this study the effective mechanical properties of carbon nanotube based composites, presented in several numerical examples, demonstrate that interphase thickness and stiffness has significant impact on the stiffness of composite. With only about 0 to 0.8 nm of the interphase thickness variation in a matrix, the stiffness of the composite in the CNT axial direction can increase as many as 1.1 and 4.3 times for the cases of long and short CNT fibers, respectively. The conclusions that can be summarized are presented below:

1. Thin interphase can affect considerably on the TEM of nanocomposites even if the change of thickness variation is comparatively less.
2. In case of soft interphase, effective tensile elastic modulus of elasticity decreases with the increase of the thickness for both long and short CNT based polymer composite.
3. In case of stiff interphase, effective tensile elastic modulus of elasticity increases with the increase of the thickness for both long and short CNT based polymer composite.
4. The effective tensile modulus (TEM) of elasticity of nanocomposites increases linearly with increase in the relative stiffness of the interphase with the matrix for both the case of long and short CNT.
5. Short CNT is affected more than long CNT with the variation of interphase thickness and stiffness.
6. When interphase thickness is increased then stiff interphase is better than soft interphase because it exhibits much better mechanical properties of nanocomposites.

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