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Effect of Carbon Nanotube Geometries on Mechanical Properties of Nanocomposite Via Nanoscale Representative Volume Element

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ABSTRACT

Predicting the effective elastic properties of carbon nanotube-reinforced nanocomposites is of great interest to many structural designers and engineers for improving material and configuration design in recent years. In this paper, a finite element model of a CNT composite has been developed using the Representative volume element (RVE) to evaluate the effective material properties of nanocomposites. Based on this model, the effects of geometrical characteristics such as the aspect ratio, orientation and volume fraction of the CNTs in conjunction with the interphase behavior on the mechanical properties of the nanocomposites are elucidated and the elastic properties of a complex polymeric nanofibrous structure are determined.

Keywords : Carbon nanotube; Nanocomposite; Representative volume element; Geometrical characteristic.

1 INTRODUCTION

IN the past decades, composite structures have become one of the most interesting research areas. The structural performance offered by composite materials is much more versatile than can be realized with conventional materials [1]. Exceptional physical properties, high aspect ratio and large surface to volume ratio of nanostructures have made them ideal candidates for developing the next generation of composites which are called nanocomposites. Nanocomposites are composite materials that the matrix material is reinforced by one or more separate nanostructures in order to improve performance properties [2]. The most common nanostructures utilized as reinforce in the nanocomposites are nanoparticles [3], nanotubes [4] and nanolayers [5]. Among different nanocomposites, the carbon nanotube-reinforced composites have been widely and successfully synthesized and utilized. Due to significant properties of carbon nanotube-reinforced composites, evaluating the mechanical properties of nanocomposites filled with carbon nanotubes (CNTs) has become a subject of primary interest in recent studies. It should be noted that the mechanical properties of a nanocomposites are affected by the material properties of constituent materials, their geometric distribution and their interactions.

The effective mechanical properties of the CNT-based composites have been evaluated by Liu and Chen [6] based on a three-dimensional nanoscale representative volume element (RVE) and finite element method (FEM). Selmi et al. [7] investigated several micromechanical models and composite systems, with various properties of the matrix and the graphene, short or long nanotubes, fully aligned or randomly oriented in two- and three dimensions. They found that the two-level Mori–Tanaka homogenization model gives the best predictions in most cases. In

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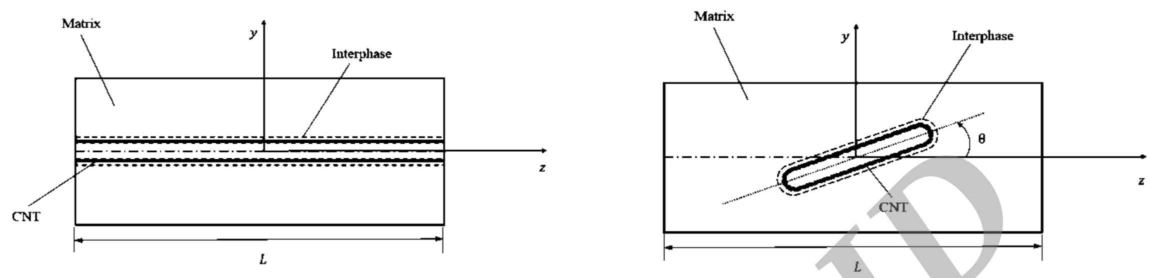
another study, classical molecular dynamics (MD) simulation has been employed to estimate the elastic moduli of the polymer/CNT composite systems [8]. Karimzadeh et al. [9] evaluated the effective mechanical properties of CNT-based by using two-dimensional nano-scale cylindrical RVE and three-dimensional square RVE. Giannopoulos et al. [10] described a micromechanical finite element approach for the estimation of the effective Young's modulus of aligned single-walled carbon nanotube (SWCNT) reinforced composites using a RVE. In this study, the effect of the interphase on the performance of the composite was investigated for various volume fractions. In another study, the effective fiber model has been utilized to calculate the elastic properties of nanocomposites with including the effect of the curvature of nanotubes [11]. The nanocomposites of Poly (ethylene terephthalate) (PET) reinforced with small quantities of the CNTs have been prepared by in situ polymerization and some important properties of SWCNT-reinforced PET composites have been studied [12]. Chatzigeorgiou et al. [13] investigated the mechanical behavior of carbon fiber composites, where the carbon fibers were coated with radially aligned carbon nanotubes. They studied the impact of the carbon nanotubes length and volume fraction in the overall composite properties. Tensile behavior of embedded short carbon nanotubes in polymer matrix in presence of van der Waals interactions in inter-phase region has been studdied by Shokrieh and Rafiee [14]. A nonlinear atomistic-based continuum model has been developed for the study of nano-reinforced polymers [15]. Kundalwal and Ray [16] analyzed a novel short fuzzy composite reinforced with aligned short carbon fibers and CNTs. It has been found that the transverse effective properties of these composites are significantly improved due to the radial growing of CNTs on the circumferential surface of the short carbon fibers. The influences of the inclinations of carbon nanotubes on the effective elastic moduli of the composites have been investigated by Joshi et al. [17] based on three-dimensional hexagonal RVE with short and straight CNTs. A three-dimensional FEM of single-walled carbon nanotube reinforced polymer has been constructed by Rafiee et al. [18] to study the effect of CNT on fracture properties of polymer. Based on the finite element modeling, Bhuiyan et al. [19] investigated the reinforcing efficiency of the CNT in the polymer matrices. They determined distribution functions of the CNT geometriesby use of scanning electron microscopy images [20]. In another study, Hu et al. [21] studied the effects of the CNT's volume fraction on the mechanical properties of the RVEs of the CNT reinforecd cpmposites. In order to reveal the effects of carbon nanotube waviness and the interaction with the matrix, parametric studies of the waviness and volume fraction of the CNTs have been caried out [22]. The effects of the SWCNT orientation and volume fraction on the mechanical properties of the nanocomposites have been investigated numerically by Huang et al. [23]. Recently, the mechanical behavior of CNT/poly methyl methacrylate composite materials subjected to tensile loading has been studied using MD simulations [24]. More recently, a micromechanical element-free method to study elastic properties of CNTs reinforced composites has been presented by Wang and Liew [25].

Since the CNT geometry and interphase region play an important role in characterizing the material properties of the CNT-reinforced nanocomposites, this paper presents the comprehensive prediction of the material properties of the CNT-reinforced nanocomposites. In this way, the effects of the aspect ratio, orientation and volume fraction of the CNTs in conjunction with the interphase behavior on the mechanical properties of the nanocomposites are elucidated and the elastic properties of a complex polymeric nanofibrous structure are determined. It should be noted that there is no existing model so far in the literature able to take into account the combined effects of interphase material and the inclination angle of the CNT. For this purpose, FEM of a CNT composite has been developed using the Representative volume element (RVE). FEM seems to be more computationally effective than other available numerical methods for the analysis of nanocomposite materials since it permits the continuum representation of the matrix phase, interphase and reinforcement. In addition, a method based on the elasticity theory is utilized to evaluate the effective material properties of CNT-based nanocomposites. It should be noted that the main parameters affecting the mechanical properties of nanocomposites are the volume fraction, aspect ratio and orientation of the CNTs and interphase materials. The cylindrical RVE is adopted in this study to predict these effects. To confirm the validity of the present research, the results are compared with those reported in the literature. The results of the present paper may be directly used in designing the nanocomposite structures.

2 MATHEMATICAL FORMULATIONS FOR EVALUATING THE EFFECTIVE MATERIAL PROPERTIES

Constructing a RVE that consists of a single CNT surrounded by matrix material is an effective method to estimate the macroscopic mechanical properties of nanocomposites. Generally, three types of the RVEs can be utilized in the literature: (1) cylindrical [26], (2) square [27] and (3) hexagonal [17] prismatic bars. In this study, a cylindrical RVE is considered for developing the formulation. It is assumed that the matrix, interphase and nanotube materials are linearly elastic, isotropic and homogeneous with specific Young's moduli and Poisson's ratios. In addition, the CNT, interphase and matrix are three separate elements which bonded to gather perfectly. Two types of the RVE with length *L* and consisting long or short CNTs are considered. To model the RVE with short CNT, hemispherical end caps are adopted for reducing the total surface energy. In the case of the RVE with long CNT, the CNT is aligned along the RVE length direction. It has been shown by many studies that the elastic properties of nanocomposite are remarkably dependent on the orientation of the dispersed CNTs. Therefore, for the case of the RVE with short CNT, we assume the CNT having inclined angle with respect to the RVE length direction. The geometries of two cases of the RVE are shown in Fig. 1.



(a) (b)

Fig.1
Cylindrical RVE. (a) RVE with long CNT and (b) RVE with short CNT.

when the RVE containing CNT with different orientations, the material properties of the RVE are anisotropic. However, the numerical results of the previous studies showed that the variation of the material properties in lateral directions due to change of the CNT orientation is not significant [17]. Hence, in this study, it is assumed that the material properties are transversely isotropic. Therefore, the stress-strain relations which are linked the normal stresses ($\sigma_x, \sigma_y, \sigma_z$) and the normal strains ($\varepsilon_x, \varepsilon_y, \varepsilon_z$) can be written as follow:

$$\varepsilon_x = \frac{1}{E_x}\sigma_x - \frac{\nu_{xy}}{E_x}\sigma_y - \frac{\nu_{zx}}{E_z}\sigma_z \tag{1}$$

$$\varepsilon_y = -\frac{\nu_{xy}}{E_x}\sigma_x + \frac{1}{E_x}\sigma_y - \frac{\nu_{zx}}{E_z}\sigma_z \tag{2}$$

$$\varepsilon_z = -\frac{\nu_{zx}}{E_x}\sigma_x - \frac{\nu_{zx}}{E_x}\sigma_y + \frac{1}{E_z}\sigma_z \tag{3}$$

where $E_z, E_x, (=E_y), \nu_{xy}$ and $\nu_{zx} (=\nu_{zy})$ are four effective material constants. It should be noted that these relations are also valid for the stress and strain components in the cylindrical coordinate system. In order to determine four material constants, the RVE is subjected to three types of loading: axial tension, uniform pressure and torsional load.

2.1 Axial tension

First of all, the cylindrical RVE undergoes a uniform stretch ΔL in axial direction at its free end and the other end is constrained. Since there is no loading on the lateral surface, there is traction free condition. Under this assumption and using Eqs. (1) and (3), the Young's modulus in axial direction and Poisson's ratio, ν_{zy} , can be obtained by:

$$E_z = \frac{\sigma_z}{\varepsilon_z} = \frac{L}{\Delta L}\bar{\sigma}_z \tag{4}$$

$$\nu_{zx} = -\frac{\Delta R_a}{R} \bigg/ \frac{\Delta L}{L} \tag{5}$$

wherein $\bar{\sigma}_z$ is average stress in *z* direction and ΔR_a is the mean radial contraction. $\Delta L, \bar{\sigma}_z$ and ΔR_a can be obtained from finite element results.

2.2 Lateral loading under uniform pressure

In this case, the RVE is constrained in *z* direction and subjected to the uniform negative pressure *p* in the radial direction. It can be shown that tangential and radial stresses exist whose magnitudes are [28]

$$\sigma_r = \frac{R^2 p}{R^2 - r_i^2}\left(1 - \frac{r_i^2}{r}\right) \qquad \sigma_\theta = \frac{R^2 p}{R^2 - r_i^2}\left(1 + \frac{r_i^2}{r}\right) \qquad r_i \le r \le R \tag{6}$$

For $r = R$, the stresses and tangential strain can be obtained as follow:

$$\sigma_r = p \qquad \sigma_\theta = \frac{R^2 + r_i^2}{R^2 - r_i^2} p \qquad \varepsilon_\theta = \frac{\Delta R_p}{R} \tag{7}$$

where ΔR_p is the radial displacement and can be determined from FEM results. In addition, r_i and *R* are inner and outer radii of the RVE, respectively. According to the plane-strain assumption, the following relations are obtained.

$$\varepsilon_r = \frac{1}{E_x}\sigma_r - \frac{\nu_{xy}}{E_x}\sigma_\theta - \frac{\nu_{zx}}{E_z}\sigma_z \tag{8}$$

$$\varepsilon_\theta = \frac{1}{E_x}\sigma_\theta - \frac{\nu_{xy}}{E_x}\sigma_r - \frac{\nu_{zx}}{E_z}\sigma_z \tag{9}$$

$$\sigma_z = \nu_{xz}(\sigma_r + \sigma_\theta) \tag{10}$$

Substitution of Eq. (10) into Eq. (9), the tangential strain is developed as:

$$\varepsilon_\theta = -\left(\frac{\nu_{xy}}{E_x} + \frac{\nu_{zx}^2}{E_z}\right)\sigma_r + \left(\frac{1}{E_x} - \frac{\nu_{zx}^2}{E_z}\right)\sigma_\theta \tag{11}$$

Using Eq. (7), we have

$$\frac{\Delta R_p}{pR} = -\left(\frac{\nu_{xy}}{E_x} + \frac{\nu_{zx}^2}{E_z}\right) + \left(\frac{1}{E_x} - \frac{\nu_{zx}^2}{E_z}\right)\frac{R^2 + r_i^2}{R^2 - r_i^2} \tag{12}$$

2.3 Torsional load

In the third loading case, the RVE subjected to a torsional load applied by a torque *T* at one end whereas the other end is fixed. Under applying torque *T*, angle of twist (α) on the end surface of the RVE is obtained as:

$$\alpha = \frac{TL}{G_{xy} J} \tag{13}$$

in which $J = \pi(R^4 - r_i^4)/2$ is the polar moment of inertia of the cross sectional area and G_{xy} is the shear modulus which is defined as:

$$G_{xy} = \frac{E_x}{2(1+\nu_{xy})} \tag{14}$$

It is noted that the angle of twist is calculated by the measurement of the end surface rotation angle in the FEM. Therefore, the magnitude of the shear modulus of elasticity can be predicted by Eq. (13). Finally, the effective Young's modulus and Poisson's ratio in the transverse direction can be calculated by solving Eqs. (12) and (14) simultaneity:

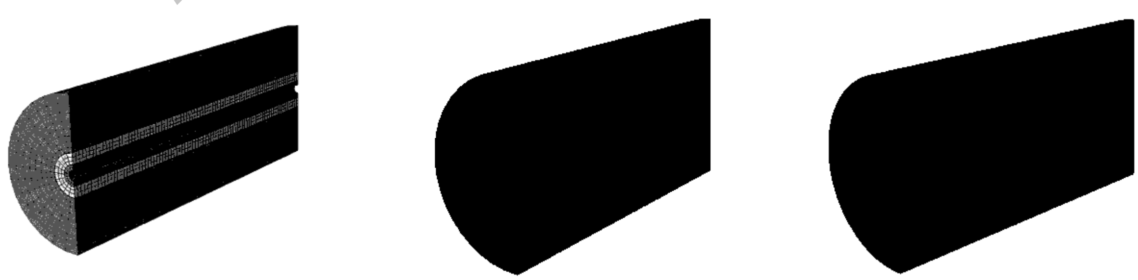
$$E_x = E_y = 2(1+\nu_{xy})G_{xy} = \frac{1}{\frac{1}{4G_{xy}}\left(1-\frac{r_i^2}{R^2}\right)+\frac{\Delta R_p}{2pR}\left(1-\frac{r_i^2}{R^2}\right)+\frac{\nu_{zx}^2}{E_z}} \tag{15}$$

$$\nu_{xy} = -1+\frac{1}{\frac{1}{2}\left(1-\frac{r_i^2}{R^2}\right)+\frac{G_{xy}\Delta R_p}{pR}\left(1-\frac{r_i^2}{R^2}\right)+\frac{2\nu_{zx}^2 G_{xy}}{E_z}} \tag{16}$$

3 MODELING OF THE RVE

To evaluate the elastic properties of the CNT-reinforced composite, the FEM and the above continuum approach under three mentioned loading conditions are utilized. The cylindrical RVE surrounded by epoxy matrix is considered. The geometrical parameters such as $\Delta L, \Delta R_a, \Delta R_p$ and α are calculated to obtain the effective Young's moduli and Poisson's ratios. The analyses have been carried out using three-dimensional finite element models of the RVE developed by the ABAQUS commercial software. The models for different RVEs are shown in Fig. 2.

An armchair single-walled carbon nanotube (10, 10) is chosen for the analysis. An interphase is considered to be between the CNT and the matrix. In the present paper, a homogeneous isotropic interphase medium of the same shape as the CNT is inserted between the CNT and matrix. The interphase thickness is considered to be twice of CNT thickness, according to the literature [29]. Here, both "hard" and "soft" interphases are studied. The Young's modulus of the hard interphases is larger than the Young's modulus of the matrix. In contract, the Young's modulus of the soft interphases is lower than the Young's modulus of the matrix. Two types of three-dimensional elements are used for modeling the RVE: (1) 10-node tetrahedron elements and (2) 20-node hexahedron elements. Although it is not show here, based on the convergence study, the element size, element density distribution, number of total elements, and number total nodes are finalized and so the results are mesh-independent. Effective elastic properties of CNT-based nanocomposites are predicted for different CNT volume fraction with different lengths of short CNTs. Moreover, the effect of hard and soft interphase materials is investigated.



(a) (b) (c)

Fig. 2
Finite element models of the RVE. (a) RVE with long CNT and (b) RVE with short CNT(ϑ = 0) and (c) RVE with short CNT(ϑ = 45).

4 NUMERICAL RESULTS AND DISCUSSION

To confirm the validity of this technique, the present results are compared with those reported in the literature. First, let us discuss the case of aligned CNT along the RVE length direction and without interphase. Table 1. compares the present results for both long and short CNTs and those obtained earlier based on FEM [6], atomistic-based continuum model [14], element-free moving least-squares (MLS) method [25] and rule of mixtures.

For validation, the following parameters are used:

Matrix: $L = 100nm, R = 10nm, E = 5GPa, v = 0.3$

Long CNT: $L = 100nm, R_{inner} = 4.6nm, t = 0.4nm, E = 1000GPa, v = 0.3$

Short CNT: $L = 50nm, R_{inner} = 4.6nm, t = 0.4nm, E = 1000GPa, v = 0.3$

Reasonable agreement between the present solutions and those given in the literature [6, 14 and 25] is achieved. The differences between the present and previous results for the short CNT are seen due to different averaging methods.

Table 1
Comparison between material properties of the RVE obtained from present simulations and previous results.

RVE types	Reference	E_z/E_m	$\nu_{zx} = \nu_{yz}$	E_x/E_m	ν_{xy}
Long CNT	Present work	10.6910	0.3000	3.7461	0.044
	FEM [6]	10.6925	0.3000	3.7654	0.0431
	Atomistic-based continuum model [14]	10.93	---	---	---
	Rule of mixtures	10.6925	---	---	---
Short CNT	Present work	1.5415	0.3038	1.7301	0.5064
	FEM [6]	1.6920	0.2758	1.2878	0.3043
	Element-free MLS method [25]	1.7356	0.2759	---	---
	Rule of mixtures	1.7879	---	---	---

In addition, Table 2. presents the comparison of axial Young's modulus from the model developed in this study and those found in literature [9, 13, 18 and 25] along with a theoretical relation known as continuum rule of mixture. A good agreement can be seen with the published data.

Table 2
Comparative study on the axial Young's moduli of RVEs with armchair (10,10) reinforcement.

Reference	V_f	E_z GPa
Karimzadeh et al. [9]	3	53.46
Rafiee et al. [18]	5	56.47
Zuberi and Esat [30]	5	54.47
Shokrieh and Rafiee [13]	5	58.59
Rule of mixtures	5	53.65
Present work (without interphasse)	5	57.05
Present work (hard interphasse)	5	66.26
Present work (soft interphasse)	5	56.33

After verifying the accuracy and reliability of the present technique, we now proceed to the application of this method to various conditions. To determine the numerical results, basic quantities which have to be defined appropriately are the geometrical and mechanical properties of the components of the RVEs. The parameters used in this work are given in Tables 3. and 4.

Table 3
Geometrical parameters of the RVE components.

$L_{mat} = L_{CNT-long}$ nm	$R_{inner-CNT}$ nm	t_{CNT} nm	$t_{interphas.}$ nm
25.6	0.32	0.34	0.68

Table 4
Material properties of the RVE components.

	Young's Modulus (GPa)	Poisson's Ratio	Reference
CNT	1054	0.28	[21]
Matrix	5	0.4	[21]
Interphase	50 (Hard) 1.5 (Soft)	0.4	[31]

4.1 Effects of interphase and volume fraction

In this part, the influences of the interphase materials and volume fraction of the CNT on the material properties of the RVE are investigated. The mechanical properties for the long CNT RVEs predicted from modeling are listed in Table 5.

Table 5
Effective material properties of long CNT RVEs extracted from modeling.

Volume fraction	Material Properties			
	E_z GPa	$\nu_{zx} = \nu_{yz}$	$E_x = E_y$ GPa	ν_{xy}
Without Interphase				
1%(R_{mat} =5.7812)	15.409	0.3968	5.7182	0.5733
2%(R_{mat}=4.0942)	25.8175	0.395	6.2969	0.5935
3%(R_{mat} =3.3480)	36.2309	0.3932	7.0939	0.5777
4%(R_{mat} = 2.9039)	46.6375	0.3914	8.1579	0.5405
5%(R_{mat}=2.6012)	57.0522	0.3896	9.5379	0.4839
Hard Interphase				
1%(R_{mat} =5.7812)	17.2445	0.3968	5.9059	0.5861
2%(R_{mat}=4.0942)	29.4934	0.3949	6.8567	0.599
3%(R_{mat}=3.3480)	41.7474	0.393	8.2382	0.5734
4%(R_{mat}= 2.9039)	53.9998	0.3912	10.0785	0.5279
5%(R_{mat}=2.6012)	66.2554	0.3893	12.1111	0.4827
Soft Interphase				
1%(R_{mat} =5.7812)	15.261	0.3969	5.654	0.5523
2%(R_{mat}=4.0942)	25.5267	0.3953	6.106	0.5516
3%(R_{mat}=3.3480)	35.7969	0.3937	6.7302	0.5104
4%(R_{mat}= 2.9039)	46.0603	0.3921	7.5314	0.4434
5%(R_{mat}=2.6012)	56.3322	0.3905	8.2748	0.3718

As expected, the axial Young's modulus of the RVEs, E_z, increases with a constant rate when the volume fraction of the long CNTs increases. Similarly, the transverse Young's modulus, E_x, tends to increase with adding the CNT. In addition, Poisson's ratio, v_{zx}, slightly decreases as the CNT volume fraction increases whereas the Poisson's ratio, v_{xy}, for all interphase materials increases at the beginning of adding CNT and reaches the maximum value and then decreases. The present results are quite consistent with the literature results. Moreover, the interphase materials generally have a considerable influence on the value of the mechanical properties of the nanocomposites properties. It is cleared that the Young's moduli of the nanocomposites increase in the case of hard interphase and decrease for the soft interphase.

As stated before, the interphase strength plays a significant role in determining the elastic properties of the nanocomposite. Fig. 3 shows the variation of four effective material constants with respect to nanotube length of short CNT for different interphase materials. In this figure, the radius of matrix (R_{mat}) is fixed at 6.4 nm. As can be seen, with increasing the nanotube length four effective material constants increase for the cases of hard interphase and without interphase. The results show that hard interphase can help the stiffening effect of the CNT in the composite. This indicates that the material properties of the nanocomposite are sensitive to the existence of interphase and its mechanical properties.

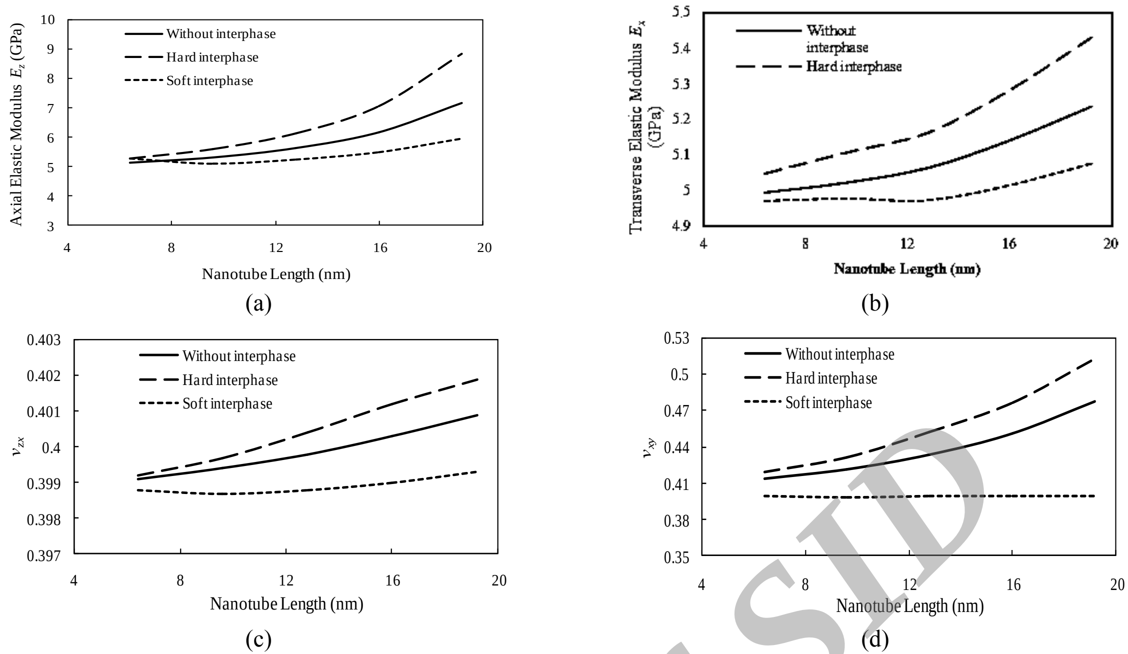


Fig. 3
Predicted variation of (a) E_z, (b) E_x, (c) ν_{zx} and (d) ν_{xy} with regard to nanotube length for different interphase materials.

The volume fraction of the nanotube is one of the most parameter affecting the elastic properties of nanocomposite. It is obvious that large value of volume fraction leads to large values for the elastic moduli. However, it is not always easy, from the practical viewpoint, to implement large values of volume fraction in nanocomposites due to problems associated with treatment and processing of the composite material.

In Fig. 4, the elastic moduli of RVE with short CNT without interphase material are plotted with respect to the volume fraction for different nanotube length. It is observed from these curves that the elastic moduli of the nanocomposite increase with increasing volume fraction of the CNTs as expected. Moreover, for a particular value of the CNT volume fraction, the modulus of the nanocomposite increases with increasing the nanotube length.

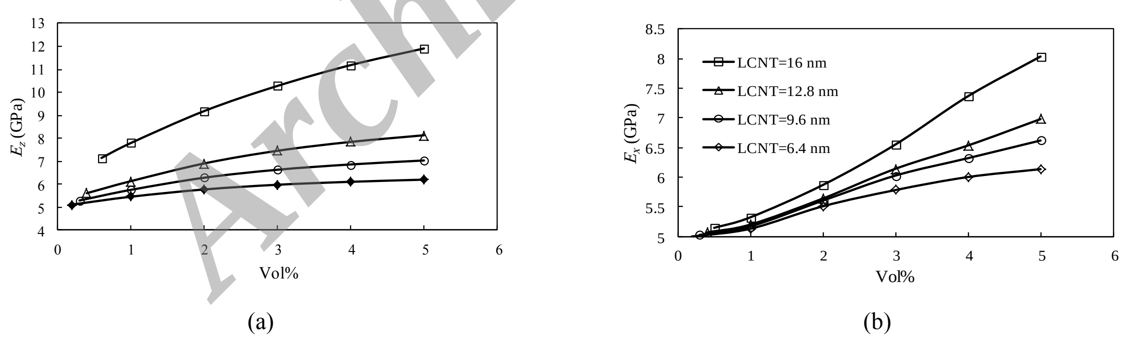


Fig. 4
Predicted variation of (a) E_z and (b) E_x with respect to volume fraction for different nanotube length.

4.2 Effects of inclination angle of CNT

To illustrate the effect of the inclination angle of the CNT, the mechanical properties of the RVE with short CNT are displayed in Fig. 5 for different interphase materials. It this figure, the radius of matrix (R_{mat})is fixed at 6.4 nm and the nanotube length is 6.4 nm.

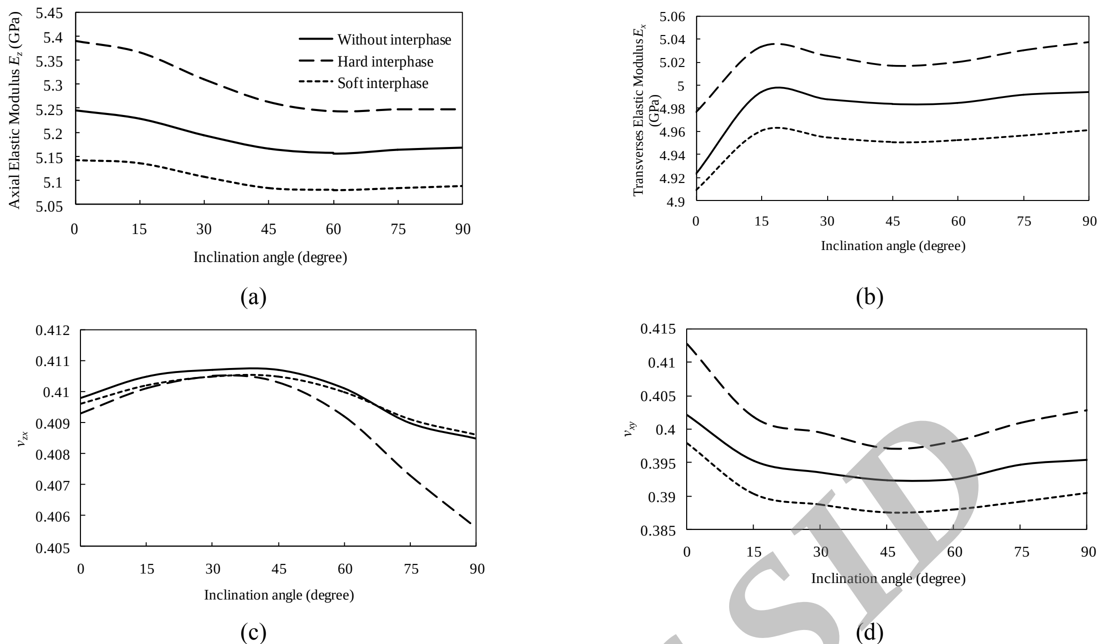


Fig. 5
Variation of (a) E_z, (b) E_x, (c) ν_{zx} and (d) ν_{xy} with regard to nanotube inclination angle for different interphase materials.

It is obvious that with increase in CNT inclination angle, the axial Young's modulus of nanocomposite decreases until it reaches to inclination angle of 60°. For $\vartheta > 60°$, the axial Young's modulus is not sensitive to the inclination angle. The Poisson's ratio, ν_{zx} , for all interphase materials, increases until it reaches to inclination angle of 30° and then decreases. In addition, it is cleared that, for $\vartheta > 15°$, the transverse elastic modulus is not very sensitive to the inclination angle.

5 CONCLUSIONS

In this paper, the effects of carbon nanotube geometries on mechanical properties of nanocomposite have been investigated. In spite of some achievement in predicting the mechanical properties of the nanocomposites, to the best authors' knowledge, there has been no attempt to tackle the problem described in the present investigation. Considering the interphase material and including the inclination angle of the CNT are the main contributions of the present paper. In order to avoid complex and expensive physical testing to determine the effective elastic properties, a numerical technique was explored. The advantage of the present approach is that it uses macroscopic properties to describe the CNTs and matrix's behaviors. Detailed representation of the molecular nanostructure of the CNTs and matrix is avoided and significant reductions in both computational cost and complexity are achieved. Using this model, the effects of main parameters such as volume fraction, aspect ratio and orientation of the CNTs and interphase materials on the effective elastic properties of the nanocomposites were also examined. The results showed that with increase of inclination angle of CNTs, the axial stiffness decreases. In addition, we found that the interphase material mostly affects the elastic properties in the nanocomposites. As expected, there is a significant effect of nanotube volume fraction on all elastic properties. Finally, it is hoped that this work would be helpful and provided useful information to the modeling, analysis and design of carbon nanotube-reinforced composites.

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