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A multistep methodology for calculation of the tensile modulus in polymer/carbon nanotube nanocomposites above the percolation threshold based on the modified rule of mixtures

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A multistep model is proposed for calculating the tensile modulus values of polymer/carbon nanotube (CNT) nanocomposites (PCNTs) based on the modified rule of mixtures, assuming a percolated network of nanoparticles. In the first step, the network of nanoparticles is considered as a new phase with a novel volume fraction and Young's modulus. Then, the volume fraction of the filler network in the PCNTs is correlated to the density of the network. Also, the percolation of the nanoparticles is related to the aspect ratio of the nanoparticles. Finally, a new model is proposed based on the modified rule of mixtures (the Riley model) of the properties of the filler network. The predictions of the proposed model are compared with experimental results and the roles of the nanoparticles and network properties in the modulus values of nanocomposites are determined. The proposed model presents acceptable predictions when compared with the experimental data. Moreover, the density and modulus of the filler network, as well as the aspect ratio and diameter of the nanoparticles was found to directly affect the moduli of the nanocomposites.

1 Introduction

Carbon nanotubes (CNTs) are an ideal reinforcing material that are used to obtain a high-modulus and conductive polymer nanocomposite, because of their great mechanical properties, good conductivity, nanoscale diameter and high aspect ratio.[1–7] Previous studies have shown that CNTs have a Young's modulus of *ca.* 1000 GPa and a tensile strength of 10–50 GPa, which are much higher than those of conventional fibres, such as carbon fibers and other nanofillers such as clay and silica. To confirm an effective load transfer from a polymer matrix to CNTs, the interfacial bonding between the polymer matrix and the nanoparticles should be optimized to prevent the bonds breaking between the two materials. Also, the van der Waals attraction between the nanotubes causes the aggregation/agglomeration of CNTs in nanocomposites, which decreases the surface area of the nanofiller and disturbs the network structure. So, it is necessary to use some methods such as modification, functionalization and compatibilizing of CNTs to gain a uniform dispersion of nanoparticles in the polymer matrix and promote good interfacial interaction/adhesion between the polymer matrix and nanoparticles.[3,8–10]

The electrical conductivity of polymer/CNT nanocomposites (PCNTs) can be increased through the formation of a filler network above the filler concentration, known as the percolation threshold.[11–14] This means that significant conductivity of PCNTs is only obtained at or above the percolation threshold. Researchers have attempted to obtain a low percolation threshold in PCNTs by manipulating the material and processing parameters.[15–17] In addition to the electrical percolation threshold, a substantial improvement in the tensile modulus has also been observed in polymer nanocomposites, which is known as mechanical percolation.[18–21] In other words, a network of nanoparticles above a specific volume fraction (percolation) results in an important improvement in the mechanical properties. For example, the high shear modulus of reinforced films with cellulose whiskers has been explained by the percolation threshold of the filler.[22] Although the mechanisms of the electrical and mechanical percolations are different, they are consistent in PCNTs.

From a theoretical point of view, the models for the critical conductivity of polymer nanocomposites are in the form of a power-law,[23,24] as a function of the percolation volume fraction. Similarly, Ouali *et al.*[25] proposed a model for the tensile modulus of conventional composites above the percolation threshold. Although this model has been extensively used for polymer nanocomposites in the previous work, it does not produce accurate results for the tensile modulus. There have been very few studies on the mechanical percolation of polymer

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nanocomposites in the literature, even though the percolation threshold is quickly reached in nanocomposites containing CNTs, due to the high aspect ratio of nanofillers.[26–28] Therefore, it is necessary to study the effects of the filler network on the mechanical properties of PCNTs, such as the tensile modulus, because it directly affects the reinforcing role that CNTs play in polymer nanocomposites.

The modified rule of mixtures, first reported by Riley in 1976,[29] produces inaccurate predictions for the tensile modulus of PCNTs (as discussed in the following sections), because it incorrectly assumes the role of the nanofiller aspect ratio. Also, this model does not incorporate the effect of the filler network in nanocomposites such as PCNTs above the percolation threshold. In this study, this model is modified and developed assuming a percolating network in PCNTs and a new model is proposed for calculating the tensile modulus of PCNTs containing a filler network. In the first step, a network of nanoparticles is considered as a new phase with a novel volume fraction that depends on the density of the network. Also, the aspect ratio of the network phase is assumed to have a value of 1, because it is considered as a sphere. Finally, the proposed model for the tensile modulus is presented by modification of the Riley model and incorporation of the new properties of the filler network. The predictions of the proposed model were compared with experimental results. Furthermore, the effects of some parameters, such as the network modulus and density, the nanofiller aspect ratio, the percolation volume fraction and diameter and the number of nanotubes per unit volume on the modulus of PCNTs were determined using the proposed model.

2 The proposed model

The high aspect ratio (the length per diameter) of CNTs, which produces a low percolation threshold, i.e. the networking of CNTs at a very low concentration in nanocomposites, is the main advantage of using CNT nanoparticles. The current study focuses on this specification of polymer CNT nanocomposites with nanoparticles/polymer nanocomposites. A network of nanoparticles can be considered as a new phase in PCNTs, which includes CNT nanoparticles and the polymer matrix among the nanotubes. In this condition, the volume fraction of the network can be expressed as:

$$\phi_N = A\phi_f \tag{1}$$

where "ϕ_f" is the volume fraction of the CNTs and "A" is a constant. Clearly, the volume fraction of the network depends on its density in the PCNTs. So, "ϕ_N" can be defined as:

$$\phi_N = P\hat{\rho}\phi_f \tag{2}$$

where "P" is a constant parameter, which depends on the fraction of CNTs in the network, because some CNTs in nanocomposites do not participate in the networks and "$\hat{\rho}$" is the relative density of the 3D CNT network. Certainly, the exact value of the "P" parameter can be calculated using some

techniques, such as morphological analysis. The "$\hat{\rho}$" parameter can be defined[30] as:

$$\hat{\rho} = \frac{\rho_N}{\rho_{CNT}} = \frac{\pi N l d^2}{4L_1 L_2 L_3} \tag{3}$$

where "ρ_N" and "ρ_{CNT}" are the density of the network and the CNTs, respectively, "N" is the total number of CNTs in the unit cell and $L_1 L_2 L_3$ is the unit volume of the network. Assuming that the aspect ratio of the CNTs is $\alpha = l/d$ (where "l" and "d" are the length and diameter of the filler, respectively) and $L_1 = L_2 = L_3 = 10$ μm, "$\hat{\rho}$" can be expressed as:

$$\hat{\rho} = \frac{10^{-9}\pi\alpha n d^3}{4000} = \frac{10^{-12}\pi\alpha n d^3}{4} \tag{4}$$

where "n" is the number of CNTs in a unit volume of the network and "d" is in units of nm. By substituting eqn (4) into eqn (2), "ϕ_N" can be represented as:

$$\phi_N = P\frac{10^{-12}\pi\alpha n d^3}{4}\phi_f \tag{5}$$

Moreover, the percolation threshold of CNTs can be related to the aspect ratio of the nanoparticles[30] as:

$$\phi_p = \frac{2.2}{\alpha} \tag{6}$$

By substituting the above equation into eqn (5), "ϕ_N" can be defined as:

$$\phi_N = 2.2P\frac{10^{-12}\pi n d^3}{4\phi_p}\phi_f \tag{7}$$

Riley modified the rule of mixtures for the tensile modulus of polymer nanocomposites assuming a modulus reduction factor (MRF) by including the aspect ratio of the nanofiller and the shear modulus of the polymer matrix[29] as:

$$E = E_m\phi_m + \text{MRF}\phi_f E_f \tag{8}$$

$$\text{MRF} = 1 - \frac{\ln(u+1)}{u} \tag{9}$$

$$u = \frac{1}{\alpha}\sqrt{\frac{\phi_f G}{E_f \phi_m}} \tag{10}$$

where "E_m" and "E_f" are the Young's moduli of the polymer matrix and filler, respectively, "ϕ_m" is the volume fraction of the polymer matrix ($\phi_f = 1 - \phi_m$) and "G" is the shear modulus of the matrix.

"ϕ_f" can be calculated by the weight fraction of the nanofiller in the nanocomposite (m_f) as:

$$\phi_f = \frac{d_c}{d_f}m_f \tag{11}$$

$$d_c = \frac{d_m d_f}{(1 - m_f)d_f + m_f d_m} \tag{12}$$

where "d_c", "d_f" and "d_m" are the densities of the nanocomposite, nanofiller and polymer matrix, respectively.

Now, the Riley model is modified and developed for PCNTs containing a filler network by assuming that the network of nanoparticles is a filler phase in the PCNTs. So, the characteristics of the whole filler network are considered as filler properties. The "ϕ_f" and "E_f" parameters in the Riley model are replaced by "ϕ_N" and "E_N" parameters in the proposed model. Moreover, the aspect ratio of the filler network is considered to have a value of 1, because the network is assumed as being a sphere in the PCNTs. Also, the shear modulus of isotropic materials, such as the polymer matrix, can be defined as:

$$G = \frac{E_m}{2(1 + \nu_m)} \qquad (13)$$

where "ν_m" is the Poisson ratio of the polymer matrix. Since the tensile modulus values of thermoplastic polymers are commonly between 2 and 4 GPa and "ν_m" has values between 0.33–0.5, "G" can be considered as 1 GPa in the proposed model.

Assuming the properties of the filler network as the filler phase in PCNTs and $\alpha = 1$ and $G = 1$ GPa in the Riley model, a new model can be proposed for calculating the tensile modulus of PCNTs above the percolation threshold as:

$$E = E_m(1 - \phi_N) + \mathrm{MRF}\phi_N E_N \qquad (14)$$

$$\mathrm{MRF} = 1 - \frac{\ln(u + 1)}{u} \qquad (15)$$

$$u = \sqrt{\frac{\phi_N}{E_N(1 - \phi_N)}} \qquad (16)$$

If the "ϕ_N" parameter in the above equations is substituted by eqn (5), the proposed model represents the properties of both the filler network and the nanoparticles. Furthermore, when "ϕ_N" is expressed by eqn (7), the proposed model shows the dependence of the modulus on the percolation threshold of the nanoparticles.

The relative modulus of PCNTs can also be expressed by:

$$E_R = (1 - \phi_N) + \frac{\mathrm{MRF}\phi_N E_N}{E_m} \qquad (17)$$

3 Results and discussion

The calculations made using the original Riley model and the proposed model were evaluated by comparing them against the experimentally determined moduli of some previously prepared samples. Fig. 1 shows the experimental data of the relative modulus and the model calculations for polyamide 6 (PA6)/CNTs from ref. 31, chitosan/multi-walled CNTs (MWCNTs) from ref. 32, PA6/MWCNTs from ref. 33, ultra-high molecular weight polyethylene (UWPE)/MWCNTs from ref. 34 and polyacrylonitrile (PAN)/MWCNTs from ref. 35. The calculations made using the original model gave the average values: $\nu_m =$

0.4, $E_f = 1000$ GPa and $\alpha = 200$. It is obvious that the calculated values from the Riley model are below those of the experimental values. As a result, the original model is not suitable for the calculation of the modulus value in PCNTs and the application of the model proposed in this study is thus justified.

Good agreement is observed between the experimental values and the calculated values of the developed model for all samples at all filler concentrations, which validates the proposed model. In other words, although some deviation is observed between the experimental and theoretical data, the agreement between the calculations and experimental measurements is acceptable. Accordingly, the proposed model can be applied to predict the tensile modulus of PCNTs. Also, the good predicting power of the proposed model indicates that the reported samples contain CNT networks above the percolation threshold. It should be noted that the calculations of the developed model show high deviations from the experimental data at high CNT concentrations, because the CNT agglomerates at a high filler concentration weaken the modulus. However, the developed model cannot consider the agglomeration of nanoparticles. Therefore, it is suggested to apply the developed model at low CNT concentrations, which excludes the agglomerations. In addition, the proposed model is suitable for nanocomposites containing CNT networks above the percolation threshold. Since a low percolation threshold arises as a result of the high aspect ratio of nanoparticles, this model can properly predict the modulus in the samples containing high aspect ratio CNTs.

The best "A" and "E_N" values can be calculated by fitting the proposed model to the experimental results. The best "A" obtained were 3, 16, 17, 5 and 10 for PA6/CNTs, chitosan/MWCNTs, PA6/MWCNTs, UWPE/MWCNTs and PAN/MWCNTs, respectively. Moreover, the calculated "E_N" values were 4000, 15 000, 14 500, 5500 and 9000 GPa for these samples, in that order. These calculations demonstrate that the best network is formed in the PA6/MWCNT sample, while PA6/CNTs has a poorly formed network. The predicted results are reasonable, because a strong network leads to a strong nanocomposite, while a poor network decreases the modulus of the nanocomposite sample. For example, the strong network of CNTs in the PA6/MWCNT sample results in an improvement in the modulus of 90% for 1 wt% of MWCNTs, but the weak network of nanoparticles only improves the modulus of the PA6/CNT sample by 50% for 6 wt% of MWCNTs. Therefore, the predicted values of the network are consistent with the improvement ranges of the modulus in the samples. Additionally, if the value of "P" is determined by looking at the material morphology in recorded images, it is possible to estimate the levels of "n", "α" and "ϕ_p" using the proposed model. However, it is possible to evaluate the general properties of a network in different samples, such as the density and modulus, using the proposed model.

Next, the roles of different parameters on the predicted modulus of PCNTs were plotted in the form of 3D and contour designs according to the proposed model. Fig. 2 shows the effects of the "A" and "E_N" parameters on the predicted modulus with values of $\phi_f = 0.02$ and $E_m = 2$ GPa. The best
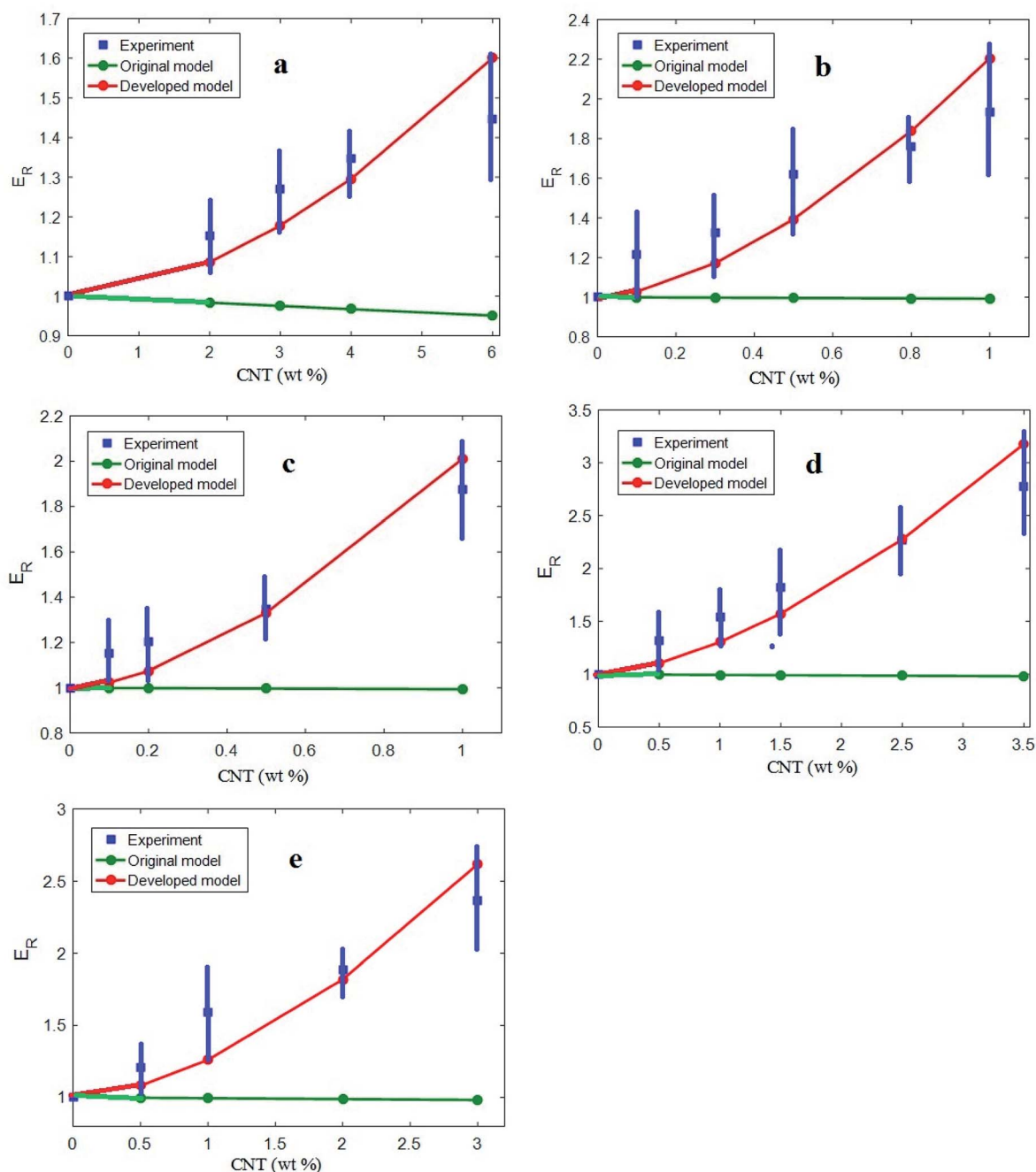
Fig. 1 The experimental data and the calculations of the relative modulus using the original and proposed model for (a) PA6/CNTs,[31] (b) chitosan/MWCNTs,[32] (c) PA6/MWCNTs,[33] (d) UWPE/MWCNTs[34] and (e) PAN/MWCNTs.[35]

modulus values were obtained using the highest "A" and "E_N" values, as expected. As observed, the relative modulus increased to 2.8 at $A = 11$ and $E_N = 5000$ GPa, while the relative modulus of 1 related to the modulus of the polymer matrix was observed at $A < 4$ and $E_N < 3000$ GPa. So, high values of the "A" and "E_N" parameters are required to achieve a high modulus value in PCNTs. The "A" parameter is a function of the relative density of the CNT network in the PCNT (eqn (1) and (2)). Obviously, a high network density results in a high level of reinforcement in the nanocomposites, because a denser network can bear

a higher level of external stress. Also, a high "E_N" value shows the formation of a strong network in the nanocomposite, which results in a high modulus.

The percolation of nanoparticles in nanocomposites depends on the interaction between the nanoparticles and a percolated network with a strongly connected network has more desirable properties compared one with a poorly connected network. Moreover, a strong network can significantly reinforce the nanocomposites, because it has a much higher modulus than that of a polymer matrix. As a result, high values
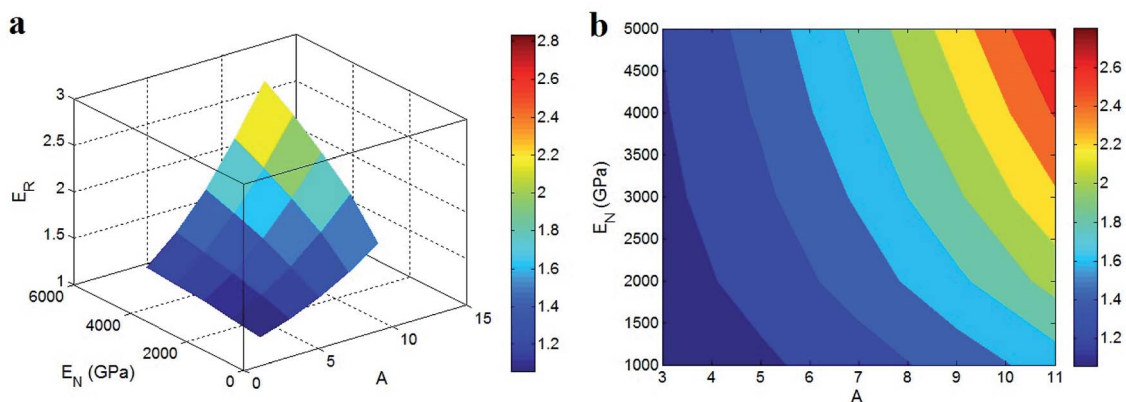
Fig. 2 (a) 3D and (b) contour plots showing the relative modulus as a function of the "A" and "E_N" parameters at $\phi_f = 0.02$ and $E_m = 2$ GPa.

for both the "A" and "E_N" parameters have been shown to logically improve the modulus of PCNTs, based on the proposed model.

Fig. 3 also depicts the effects of the "n" and "α" parameters on the relative modulus of PCNTs using the proposed model with values of: $\phi_f = 0.02$, $E_m = 2$ GPa, $E_f = 3000$ GPa, $d = 20$ nm and $P = 1000$. Low modulus values were observed at low "n" and "α" values. In other words, the modulus value of a nanocomposite cannot be improve if $n < 1500$ and $\alpha < 700$, under these conditions. However, the relative modulus value of the nanocomposite increases to 3.5 (a 250% improvement) at $n = 2500$ and $\alpha = 1000$. These explanations demonstrate the important effects of the "n" and "α" parameters on the modulus value of PCNTs. When high "n" and "α" values are achieved by tuning the material and processing parameters, the modulus significantly improves to good values, while the poor "n" and "α" values result in an undesirable modulus value in nanocomposites.

These outputs from the proposed model are reasonable, because the "n" and "α" parameters directly affect the modulus of the nanocomposites. The "n" parameter representing the number of nanotubes in a unit volume of the network obviously affects the modulus value, because the number of nanotubes in the network determines the level of reinforcement. A high number of nanotubes in the network significantly reinforces the polymer matrix, whereas a poor "n" value has a negligible effect on the modulus of the nanocomposite. So, there is a direct influence of "n" on the modulus of PCNTs. Moreover, the high aspect ratio of nanotubes results in good reinforcement in the nanocomposites. As is known, long and thin nanotubes provide more interfacial area between the polymer matrix and nanoparticles, which results in a strong interfacial interaction.[36–38] Therefore, the high aspect ratio of nanoparticles results in a large interfacial interaction between the polymer matrix and nanofiller, which facilitates the transfer of stress from the polymer matrix to the nanoparticles. Also, a high "α" value results in the breaking of the aggregates/agglomerates of the CNTs (or achievement of nanoscale in most cases) and good dispersion of the nanoparticles in the polymer matrix, which in turn results in an increase in the modulus. Therefore, the role of "α" in the modulus of nanocomposites in the proposed model is justified according to the values calculated for the interface/interphase between the polymer matrix and CNTs and the properties of the nanoparticles.[39,40]
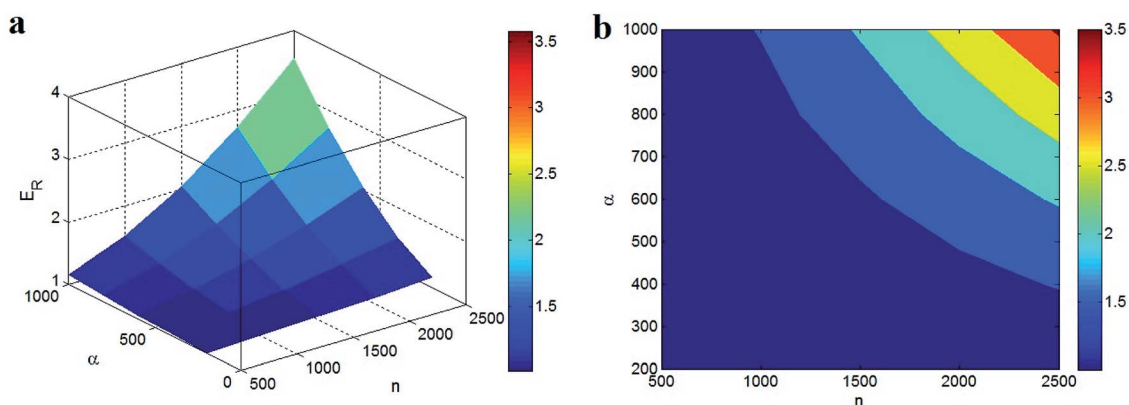

Fig. 3 The effects of the "n" and "α" parameters on the relative modulus values of PCNTs using the proposed model at $\phi_f = 0.02$, $E_m = 2$ GPa, $E_f = 3000$ GPa, $d = 20$ nm and $P = 1000$: (a) 3D and (b) contour plots.
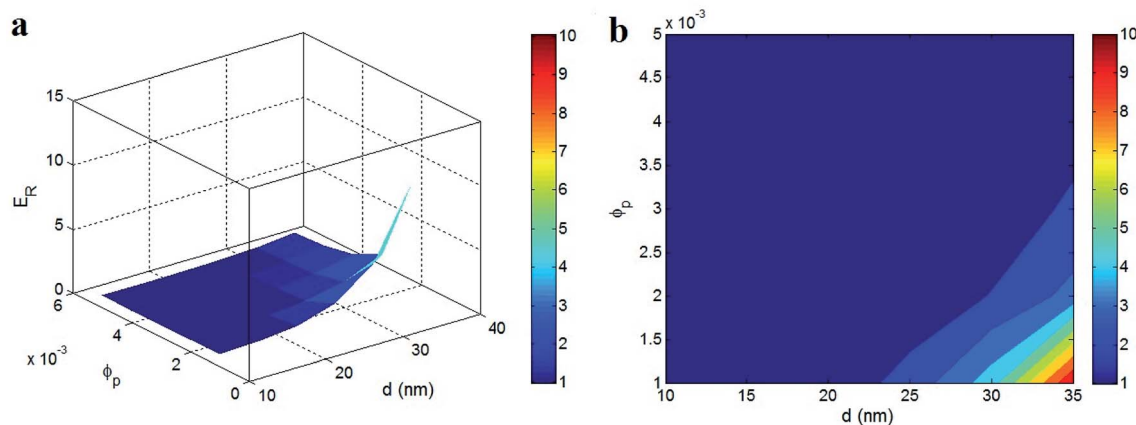
Fig. 4 "E_R" as a function of the "d" and "ϕ_p" parameters using the proposed model at values of $\phi_f = 0.02$, $E_m = 2$ GPa, $E_f = 3000$ GPa, $N = 400$ and $P = 1000$: (a) 3D and (b) contour plots.

Fig. 4 shows the effects of the "d" and "ϕ_p" parameters on the predicted relative modulus using the proposed model with values of: $\phi_f = 0.02$, $E_m = 2$ GPa, $E_f = 3000$ GPa, $N = 400$ and $P = 1000$. It is suggested that the relative modulus of nanocomposites is highly dependent on the "d" and "ϕ_p" parameters, because it changes from 1 to 10 for different values of these parameters. The modulus values of nanocomposites do not improve for the polymer matrix over wide ranges of the "d" and "ϕ_p" parameters, including low "d" and high "ϕ_p" values. So, low "d" and high "ϕ_p" values produce undesirable modulus values for polymer nanocomposites. However, an increase in the "d" value from about 25 nm and a decrease in the "ϕ_p" value from 0.003 results in some improvement in the modulus of nanocomposites. The best relative modulus of 10 is observed at $d = 35$ nm and $\phi_p = 0.001$. So, a good nanocomposite modulus can be obtained with a thick layer of nanoparticles and a low percolation threshold. The thickness of the nanoparticles increases the density of the CNT network in the nanocomposites (eqn (4)), which reinforces the nanocomposites, as previously mentioned. In addition, a low percolation threshold accelerates the formation of the filler network in the nanocomposites for only a slight amount of nanofiller. On the other hand, a high percolation does not permit the nanoparticles to form a network at a low concentration of nanofiller and the network forms at a high nanoparticle content.

Since the modulus of nanocomposites considerably improves by the percolation of the nanoparticles, the inverse relationship between the modulus and the percolation is correct. Therefore, the observed effects of the "d" and "ϕ_p" parameters on the PCNT modulus are correct, which validates the proposed model for calculating the modulus of PCNTs containing a filler network. Now, the use of the proposed model to predict the values of the relative density of the CNT network and other parameters in PCNTs and replicate experimental tensile modulus values is promising. Therefore, the present modeling method can be helpful in the absence of accurate and practical methods for the determination of these parameters. Also, there is the potential to compare the

network properties in different samples using the proposed model without undertaking morphological analysis.

4   Conclusions

The conventional model proposed by Riley for calculating the tensile modulus of polymer nanocomposites was modified and developed in various steps, assuming a percolated network of CNTs in PCNTs. A network of nanoparticles was considered as a filler phase with a new volume fraction and Young's modulus. Also, the volume fraction of the filler network and the percolation of nanoparticles were found to be dependent on the density of the network and the aspect ratio of the nanoparticles, respectively. The proposed model can be used to predict the modulus of PCNTs as a function of the properties of the nanoparticles and filler network. The predictions of the proposed model show an acceptable level of agreement with the experimental results of several samples from literature, while the original model cannot be used to calculate logical results for the modulus values of PCNTs. The relative modulus value was found to increase to 2.8 at $A = 11$ and $E_N = 5000$ GPa, while it did not improve at $A < 4$ and $E_N < 3000$ GPa. Thus, the "A" and "E_N" parameters were found to have a positive effect on the modulus values of PCNTs. Moreover, the modulus of a nanocomposite did not increase at $n < 1500$ and $\alpha < 700$, while the relative modulus improved by 250% at $n = 2500$ and $\alpha = 1000$. These findings demonstrate the direct effect of the "n" and "α" parameters on the modulus values of PCNTs. The proposed model also shows that a thin layer of nanoparticles and high percolation threshold decreases the modulus values of polymer nanocomposites. Nevertheless, the best relative modulus value of 10 was observed at $d = 35$ nm and $\phi_p = 0.001$. Therefore, a good modulus can be obtained with a thick layer of nanoparticles and a low percolation threshold.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

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