Journal of Polymer Research (2021) 28: 131
https://doi.org/10.1007/s10965-021-02488-z

ORIGINAL PAPER

Development and simplification of a micromechanic model for conductivity of carbon nanotubes-reinforced nanocomposites

Yasser Zare[1] · **Kyong Yop Rhee**[2]

Received: 2 September 2020 / Accepted: 8 March 2021 / Published online: 17 March 2021

Abstract
In the current paper, Hui-Shia model for composite's modulus is simplified and advanced to evaluate the electrical conductivity of carbon nanotubes (CNT)-reinforced nanocomposites (PCNT) by CNT aspect ratio, CNT network part, interphase district and tunnels between adjacent CNT. CNT loading, CNT magnitude and interphase depth suggest the proportion of networked CNT. In addition, CNT and tunneling intrinsic resistances express the entire conductivity of included components in nanocomposites. The reasonable roles of all factors in the conductivity and the proper matching between trial data and predictions approve the established model. Both dense interphase and long CNT improve the conductivity; however, reedy interphase or short CNT induces an insulated sample. The polymer tunneling resistivity negatively affects the conductivity, while CNT conductivity is an unsuccessful term. Furthermore, both short and wide tunnels desirably control the nanocomposite's conductivity.

Keywords Reinforced polymer nanocomposites · Carbon nanotubes (CNT) · Electrical conductivity · Interphase district · Tunneling role

Introduction

The exceptional conductivity in addition to large aspect ratio of carbon nanotubes (CNT) considerably recovers the conductivity of polymer CNT nanocomposites (PCNT). The high aspect ratio of CNT decreases the percolation onset establishing the conductive nets at low filler loadings [1–3]. Therefore, a little content of CNT can grow the conductivity of samples facilitating their application in electronics, shielding and sensing [4–17]. The tunneling effect importantly manages the conductivity, because the adjacent CNT can conveyance the charges and improve the conductivity [18–20]. The tunnel qualifications such as length/distance, contact part and polymer resistivity mainly affect the tunneling conductivity [21, 22].

The significant surface area of CNT yields the interphase district amid polymer medium and reinforcing particles [23–25]. The reinforcing efficiency of interphase was stated in preceding articles [26, 27]. The interphase section also facilitates the construction of conductive networks in nanocomposites [28–30]. A dense interphase drops the percolation onset and extends the networks. Therefore, the interphase zone improves the extent of charge transferring and conductivity. The positive efficiency of interphase percolation in the mechanics of PCNT was studied [31, 32]; on the other hand the interphase role in the conductivity of nanocomposites has been disregarded in the earlier reports.

The earlier studies mostly focused on simple power expression for PCNT conductivity correlating the conductivity to CNT content, CNT conductivity and percolation onset [33–35]. However, this equation cannot reflect the effects of tunnels and interphase section on the conductivity. Some authors have advanced the models for conductivity by CNT specifications, conductive nets, interphase region and tunnels [35–37]. However, most authors have neglected the novel terms in nanocomposites such as interphase region and tunneling properties for conductivity. Also, some researchers have considered these terms, but complex equations and incorrect consideration of these terms baffle the readers and

✉ Kyong Yop Rhee
rheeky@khu.ac.kr

[1] Biomaterials and Tissue Engineering Research Group, Department of Interdisciplinary Technologies, Breast Cancer Research Center, Motamed Cancer Institute, ACECR, Tehran, Iran

[2] Department of Mechanical Engineering (BK21 Four), College of Engineering, Kyung Hee University, 1 Seocheon, Giheung, Yongin, Gyeonggi 449-701, Republic of Korea

cause the wrong estimations. Feng and Jiang [36] incorporated two electron hopping and conductive network mechanisms into the model by interphase layer and effective aspect ratio of CNT. However, the interphase layer around CNTs is a different phase than the tunneling region. Also, the complex equations in that work cannot provide a simple methodology for estimation of conductivity. Additionally, Jang and Yin [35] have given a complex equation for conductivity without interphase region. Clearly, this type of equation cannot give the accurate outputs, due to disregarding the interphase zone in nanocomposites. Moreover, Seidel and Lagoudas [37] presented a micromechanics model by the composite cylinders model as a nanoscale representative volume element where the effects of electron hopping are introduced in the interphase layer. This study also has complex equations, which are problematic for the estimation of conductivity. Our group has also developed many models for the conductivity of nanocomposites [38–40]. We should note that each paper has studied the conductivity by dissimilar approach, new model or novel parameters/terms. For example, we progressed the McLachlan model for electrical conductivity of nanocomposites assuming the networking of interphase region [38]. We also developed the expanded Takayanagi model for conductivity [39]. Furthermore, we presented the role of interfacial conductivity in the electrical conductivity of nanocomposites assuming the imperfect interfacial adhesion between polymer matrix and CNT [40]. Undoubtedly, all papers published by our group have studied the unique approach, novel model or new parameters/terms for conductivity of nanocomposites offering significant advances compared to other articles in this field.

Many models have projected the nanocomposite's modulus/strength by the properties of polymer medium, nanoparticles and interphase region [24, 41–43]. These models can be advanced to forecast the nanocomposite's conductivity, since both modulus and conductivity regularly depend on the CNT and interphase properties. Hui and Shia [44] proposed a simple model for tensile modulus of nanocomposites supposing CNT aspect ratio, CNT concentration and moduli of polymer matrix and interphase area. This equation can be modified to estimate the conductivity for PCNT. The present article simplifies and develops the Hui-Shia model for conductivity by the portion of conductive networks containing CNT and interphase region and the tunneling region between near CNT. The current paper assumes the effective parameters/terms on the conductivity of PCNT by a novel approach based on Hui-Shia model. CNT content, CNT extent and interphase depth formulate the percolation onset and operative filler concentration to express the network volume portion. In addition, the intrinsic resistances of CNT and tunneling region propose the total conductivity of these components. Accordingly, the established model implies the dependencies of conductivity on CNT, interphase, tunnels and nets. All parameters' roles in the conductivity are stated spending the advanced model. Furthermore, the calculations of developed model for several examples are linked to the empirical outcomes. The equitable impacts of all factors on the conductivity and good agreements among trial data and forecasts justify the established model. The current study suggests the significant advances compared to other articles in this field, which can guide the researchers to predict and optimize the conductivity of PCNT by simple and accurate equations.

Modeling technique

Hui and Shia [44] derived a simple model for composites tensile modulus as:

$$E = \frac{E_m}{1 - \frac{\phi_f}{4}(\frac{1}{A} + \frac{3}{A+B})} \tag{1}$$

$$A = \phi_f + \frac{E_m}{E_f - E_m} + 3(1 - \phi_f)[\frac{(1-g)\alpha^2 - \frac{g}{2}}{\alpha^2 - 1}] \tag{2}$$

$$B = (1 - \phi_f)[\frac{3(\alpha^2 + 0.25)g - 2\alpha^2}{\alpha^2 - 1}] \tag{3}$$

$$g = \frac{\pi}{2}\alpha = \frac{\pi R}{l} \tag{4}$$

where "E_m" and "E_f" denote the moduli of polymer medium and nanofiller, in that order. "ϕ_f", "R" and "l" are the volume portion, radius and length of CNT, in that order. "α" is inverse aspect ratio of nanoparticles as the relation of diameter to length.

However, very large and thin CNT extremely decrease the "α" to about 0. Also, "ϕ_f" is very low and CNT show very high modulus about 1000 GPa. These remarks simplify the equations for "A" and "B" as:

$$A = \phi_f + 3[\frac{-\frac{g}{2}}{-1}] = \phi_f + 3\frac{g}{2} \tag{5}$$

$$B = [\frac{3(0.25)g}{-1}] = -0.75g \tag{6}$$

Hui-Shia model can predict the nanocomposite's conductivity once the modulus is substituted by conductivity as:

$$\sigma = \frac{\sigma_m}{1 - \frac{\phi_f}{4}(\frac{1}{A} + \frac{3}{A+B})} \tag{7}$$

where "σ" and "σ_m" denote the conductivity of nanocomposite and polymer matrix, correspondingly.

This equation mainly under-estimates the conductivity of PCNT owing to too deprived conductivity of polymer medium (about 10^{-15} S/m) [45]. Therefore, "σ_m" can be detached from this equation. Furthermore, this equation cannot reflect the volume portion of attached CNT afterward percolation onset (ϕ_N), which effectively manipulates the conductivity [46]. Also, it was reported that the aspect ratio and conductivity of nanofillers directly affect the conductivity of nanocomposites [36, 47]. Moreover, this equation lacks the tunneling resistance in the samples. The intrinsic resistances of CNT and tunnels can suggest the total conductivity of these components (σ_{tot}) in PCNT.

According to these explanations, the simplified Hui-Shia model for conductivity is developed as:

$$\sigma = \frac{\phi_N^2 \sigma_{tot}}{20\alpha[1 - \frac{\phi_N}{4}(\frac{1}{A} + \frac{3}{A+B})]} \tag{8}$$

$$A = \phi_N + 3\frac{g}{2} \tag{9}$$

$$B = -0.75g \tag{10}$$

$$g = \frac{\pi R}{l} \tag{11}$$

The interphase district raises the operative content of CNT in PCNT, because they add to the nets. The volume portion of interphase zone in specimens is recommended [48] by:

$$\phi_i = \phi_f(1 + \frac{t}{R})^2 - \phi_f \tag{12}$$

where "t" is interphase depth. The operative volume portion of nanofiller includes CNT and interphase as:

$$\phi_{eff} = \phi_f + \phi_i = \phi_f(1 + \frac{t}{R})^2 \tag{13}$$

Besides, the percolation onset is expressed [49] by:

$$\phi_p = \frac{\pi R^2 l}{\frac{32}{3}\pi(R+t)^3[1 + \frac{3}{4}(\frac{l/u}{R+t}) + \frac{3}{32}(\frac{l/u}{R+t})^2]} \tag{14}$$

The substantial length of CNT causes the waviness dwindling the nanocomposite's conductivity [50]. The operative length of CNT (l_{eff}) is the least expanse amongst two tops of curled CNT suggesting the curliness term as:

$$u = \frac{l}{l_{eff}} \tag{15}$$

Also, the curliness falls the conductivity of CNT (σ_f) [51] as:

$$\sigma_{CNT} = \frac{\sigma_f}{u} \tag{16}$$

Equation 14 can calculate the percolation onset by CNT and interphase extents.

The proportion of networked CNT after percolation onset is expressed [36] by:

$$f = \frac{\phi_f^{1/3} - \phi_p^{1/3}}{1 - \phi_p^{1/3}} \tag{17}$$

The operative filler fraction (Eq. 13) and percolation onset (Eq. 14) change the "f" to:

$$f = \frac{\phi_{eff}^{1/3} - \phi_p^{1/3}}{1 - \phi_p^{1/3}} \tag{18}$$

Also, "φ_N" is predicted by:

$$\phi_N = f\phi_{eff} \tag{19}$$

Exchanging of "f" (Eq. 18) and "φ_{eff}" (Eq. 13) into the latter equation expresses "φ_N" as:

$$\phi_N = (\frac{\phi_{eff}^{1/3} - \phi_p^{1/3}}{1 - \phi_p^{1/3}})\phi_f(1 + \frac{t}{R})^2 \tag{20}$$

Furthermore, the entire resistance of CNT and tunnels is suggested by:

$$R_{tot} = R_f + R_{tun} \tag{21}$$

where "R_f" and "R_{tun}" denote the inherent resistances of CNT and tunnels, correspondingly.

"R_f" is given [45] by:

$$R_f = \frac{l}{\pi R^2 \sigma_f} \tag{22}$$

The waviness cannot change "R_f", because the waviness decreases the CNT length and conductivity, simultaneously.

The tunneling resistance involves the resistances of CNT (R_1) and polymer sheet (R_2) between adjacent CNT as:

$$R_{tun} = R_1 + R_2 \tag{23}$$

"R_1" is expressed [45] by:

$$R_1 = \frac{1}{d\sigma_f} \tag{24}$$

where "d" is tunneling diameter/width. The waviness strengthens "R_1" as:

$$R_1 = \frac{u}{d\sigma_f} \tag{25}$$

Also, "R_2" is defined [45] as:

$$R_2 = \frac{\rho\lambda}{S} \cong \frac{\rho\lambda}{d^2} \tag{26}$$

where "ρ" is polymer tunneling resistivity (ohm.m), "λ" is tunneling length/distance and "S" is contact part.

Presumptuous the last equations in Eq. 23 proposes the intrinsic tunneling resistance as:

$$R_{tun} = \frac{u}{d\sigma_f} + \frac{\rho\lambda}{d^2} \tag{27}$$

In addition, replacing of "R_f" and "R_{tun}" into Eq. 21 expresses the whole confrontations of components as:

$$R_{tot} = \frac{l}{\pi R^2 \sigma_f} + \frac{u}{d\sigma_f} + \frac{\rho\lambda}{d^2} \tag{28}$$

The total conductivity of components (S/m) can be calculated by:

$$\sigma_{tot} = \frac{l}{\pi R^2 R_{tot}} \tag{29}$$

Changing of "R_{tot}" from Eq. 28 into the last equation presents:

$$\sigma_{tot} = \frac{l}{\pi R^2 \left(\frac{l}{\pi R^2 \sigma_f} + \frac{u}{d\sigma_f} + \frac{\rho\lambda}{d^2}\right)} = \frac{l}{\frac{l}{\sigma_f} + \frac{\pi R^2 u}{d\sigma_f} + \frac{\pi R^2 \rho\lambda}{d^2}} \tag{30}$$

Replacing of "φ_N" (Eq. 16) and "σ_{tot}" (Eq. 26) into Eqs. 8–11 advances a conductivity model for PCNT as:

$$\sigma = \frac{\left[\left(\frac{\phi_{eff}^{1/3} - \phi_p^{1/3}}{1 - \phi_p^{1/3}}\right)\phi_f\left(1 + \frac{t}{R}\right)^2\right]^2 \left(\frac{l}{\frac{l}{\sigma_f} + \frac{\pi R^2 u}{d\sigma_f} + \frac{\pi R^2 \rho\lambda}{d^2}}\right)}{20\alpha\left[1 - \frac{\left(\frac{\phi_{eff}^{1/3} - \phi_p^{1/3}}{1 - \phi_p^{1/3}}\right)\phi_f\left(1 + \frac{t}{R}\right)^2}{4}\left(\frac{1}{A} + \frac{3}{A+B}\right)\right]} \tag{31}$$

$$A = \left(\frac{\phi_{eff}^{1/3} - \phi_p^{1/3}}{1 - \phi_p^{1/3}}\right)\phi_f\left(1 + \frac{t}{R}\right)^2 + 3\frac{g}{2} \tag{32}$$

$$B = -0.75g \tag{33}$$

$$g = \frac{\pi R}{l} \tag{34}$$

correlating the PCNT conductivity to CNT content, CNT size, network portion, interphase depth and tunnel parameters.

Results and discussion

Parametric analyses

The industrialized model can define the parameters' roles in the conductivity. The logical effects of whole parameters on the conductivity confirm the established equations. Three-dimensional (3D) and contour plans exhibit the effects of two independent factors on the conductivity at medium $R = 10$ nm, $\phi_f = 0.01$, $\sigma_f = 10^5$ S/m, $l = 10$ µm, $u = 1.2$, $t = 10$ nm, $d = 15$ nm, $\rho = 500$ Ω.m and $\lambda = 5$ nm. These plots can lead the investigators to optimize the nanocomposite's conductivity.

Figure 1 exhibits the influences of "R" and "u" factors on the conductivity using the advanced model. $R = 5$ nm and $u = 1$ make the highest conductivity to 20 S/m, but $R > 7.5$ nm produce an insulated nanocomposite. Clearly, reedy CNT and poor waviness develop the conductivity, although only dense CNT produce an insulated sample. The high range of conductivity at dissimilar ranks of "R" and "u" demonstrate that these parameters significantly affect the conductivity. The CNT radius role in the conductivity is further important than waviness, because thick CNT mainly weaken the conductivity.

Thin CNT enlarge the interphase region and decline the percolation onset. Accordingly, thin CNT develop the nets developing the extent of charge transporting in nanocomposites. Moreover, small radius of CNT increases the total conductivity of CNT and tunneling zone, which effectively improves the conductivity. Also, thin CNT decrease the inverse aspect ratio positively governing the conductivity. It can be stated that thin CNT grow the scope and conductivity of nets promoting the electron transportation. Nevertheless, dense CNT significantly deteriorate the dimensions and conductivity of networks weakening the electron transportation. Accordingly, thick CNT dominantly decline the conductivity, but thin CNT advantageously control it.

Small waviness positively affects the operative CNT length and conductivity, as stated. A poor curliness grows the operative specifications of CNT increasing the electron transportation. Indeed, low curliness encourages the size and conductivity of nets for efficient transferring of electrons. On the other hand, high waviness shortens the CNT and diminishes its conductivity, which minimize the size and conductivity of networks. Undoubtedly, the small networks cannot intensify the charge transporting in the samples. Therefore, the waviness inversely handles the conductivity legalizing the calculations of the advanced model.

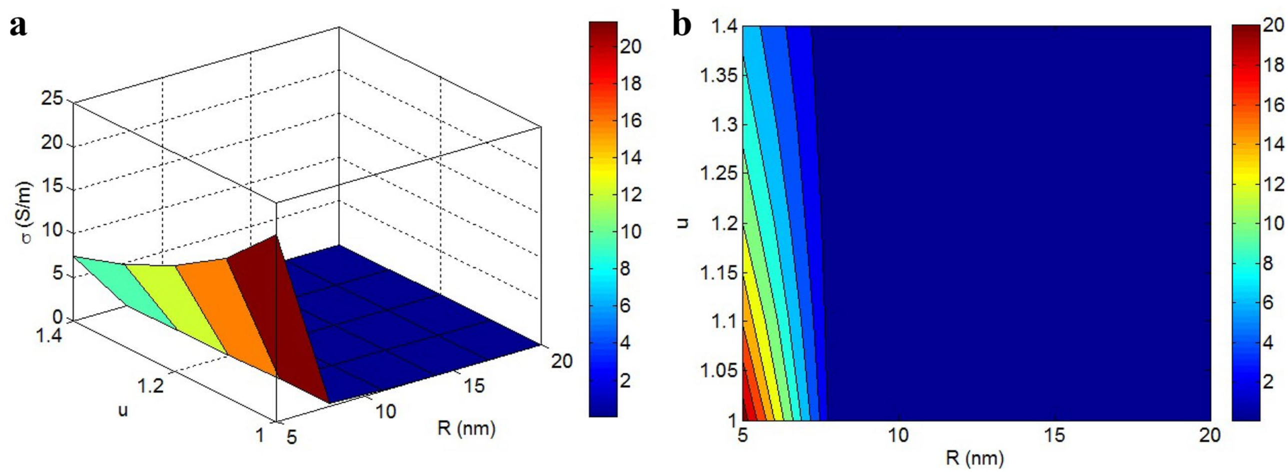

Fig. 1 Significances of "R" and "u" terms on the conductivity by the developed model: **a** 3D and **b** contour patterns

Figure 2 clarifies the conductivity as a function of "t" and "*l*" factors utilizing the advanced model. The maximum conductivity of 6 S/m is observed at $t=20$ nm and $l=20$ μm, whereas an insulated specimen is induced by $t<14$ nm or $l<11.5$ μm. These outputs indicate that the interphase depth and CNT length straightly control the conductivity. Dense interphase and large CNT improve the conductivity, but thin interphase or short CNT cannot change the polymer medium conductivity. As a result, interphase and CNT dimensions significantly manage the conductivity.

Thick interphase decreases the percolation onset and enhances the operative filler concentration. So, a dense interphase broadens the conductive nets. Since big networks largely transfer the electors in nanocomposites, thick interphase positively affects the conductivity. However, thin interphase negligibly expands the networks, which cannot change the charge transferring through networks producing an insulated nanocomposite. Therefore, providing a dense interphase is essential to develop the wide networks and charge transportation in nanocomposites. These remarks establish that the advanced model correctly shows the role of interphase depth in the conductivity.

Large CNT change the percolation onset to low filler loadings [47]. Therefore, long CNT can participate a big quantity of nanoparticles in attached nets enhancing the network volume portion. Moreover, large CNT grow the entire conductivity of CNT and tunnels increasing the electron transportation in nanocomposites. In fact, large CNT positively handle the size and condition of nets intensifying the conductivity. In addition, long CNT mainly decrease the inverse aspect ratio, which desirably affects the conductivity. These observations confirm the advanced model associating the conductivity to CNT size.

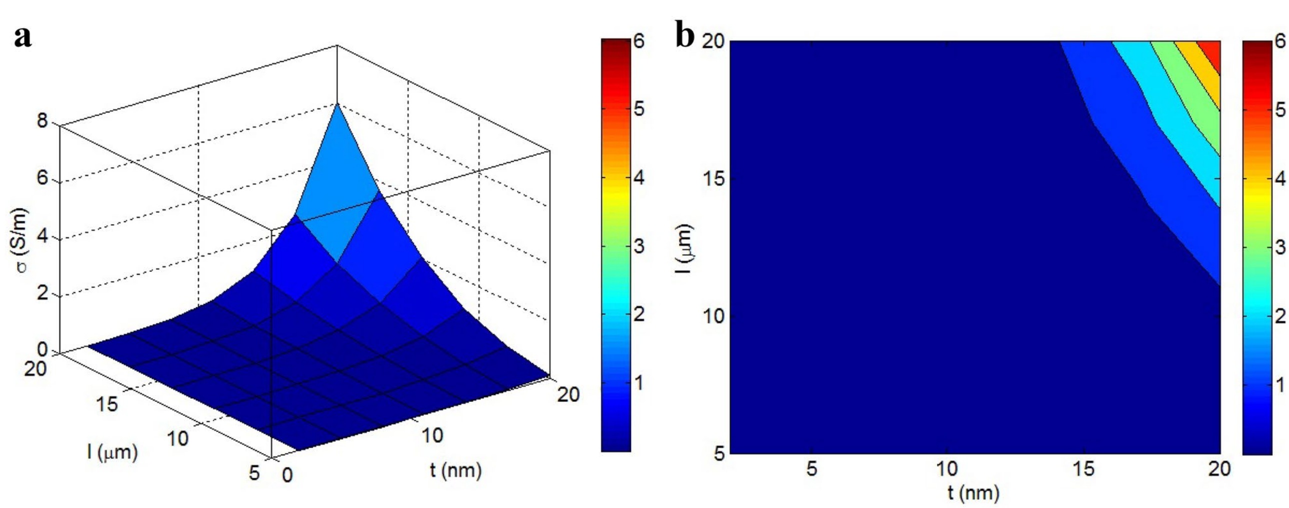

Fig. 2 Alteration of conductivity at dissimilar ranges of "t" and "*l*" using the present model: **a** 3D and **b** contour plots

Figure 3 demonstrates the predictions of conductivity at the changed ranges of "ϕ_f" and "ϕ_p" using the advanced model. The maximum conductivity of 0.4 S/m is obtained at $\phi_f = 0.02$ and $\phi_p = 0.001$, but the conductivity decreases to about 0 at $\phi_f < 0.011$. Both extraordinary CNTcontent and low percolation onset improve the conductivity, but low filler concentration causes an insulated example. The results prove the more significant role of filler concentration than that of percolation threshold, although both filler loading and percolation onset control the conductivity.

High CNT content reasonably raises the conductivity, since the big quantity of conductive nanoparticles effectively encourages the charge transferring. A more content of CNT produces the bigger networks facilitating the transportation of electrons. So, high concentration of CNT positively handles the conductivity. However, very low CNT concentration causes a small number of nanoparticles with large separation distance. Therefore, poor CNT concentration fails the interacting of nanoparticles deteriorating the conductivity. Actually, since the distributed CNT with large separation distance cannot transfer the electrons, very little attentiveness of CNT cannot develop the conductivity. It should be noted that CNT concentration after percolation threshold can establish the conductive networks for charge transferring, while too little CNT content under percolation cannot provide the network structures. Accordingly, the conductivity directly associates to the CNT concentration, as expressed by the advanced model.

A little percolation onset enlarges the conductive nets, because a big amount of CNT can join to the linkages. Though, a big percolation level limits the formation of large nets. Therefore, the percolation threshold inversely affects the conductivity. In fact, a big percolation onset decreases the percentage of CNT nets and worsens the charge transferring. Some prototypes such as power-law model explicitly indicated the inverse role of high percolation threshold in the conductivity [21, 52], but the developed models in recent studies highlighted the effect of percolation threshold on the conductivity by its influence on the network part [36, 47]. Thus, the advanced model sensibly exhibits the inverse connection between conductivity and percolation onset.

Figure 4 presents the effects of "σ_f" and "ρ" terms on the conductivity rendering the advanced model. The supreme conductivity of 0.12 S/m is observed at $\rho = 100$ Ω.m and different levels of "σ_f", while $\rho > 650$ Ω.m significantly diminish the conductivity to about 0.015 S/m. So, the polymer tunneling resistivity negatively manages the conductivity, but filler conductivity is an unsuccessful parameter. High tunneling resistivity weakens the conductivity, but the CNT conductivity cannot change it. Conversely, the small difference of conductivity at dissimilar ranges of "σ_f" and "ρ" parameters signifies that these factors negligibly handle the conductivity.

The dissimilar levels of filler conductivity cannot operate the nanocomposite's conductivity, because it cannot modify the total conductivity of CNT and tunneling region. The excellent conductivity of CNT mainly lessens their resistance in nanocomposites. Actually, the extremely insignificant resistance of CNT cannot govern the total resistance of components and thus the CNT conductivity cannot control the whole conductivity. However, the great resistance of tunneling region governs the electron transferring in nanocomposites and very poor CNT resistance is unproductive on the conductivity. Thus, the advanced model appropriately predicts the ineffective character of CNT conductivity in the conductivity outputs. Moreover, high polymer tunneling resistivity restricts the electron moving through tunneling region. Instead, deprived tunneling resistivity encourages the charge transferring within tunneling space. Accordingly, the tunneling resistivity directly handles the tunneling resistance

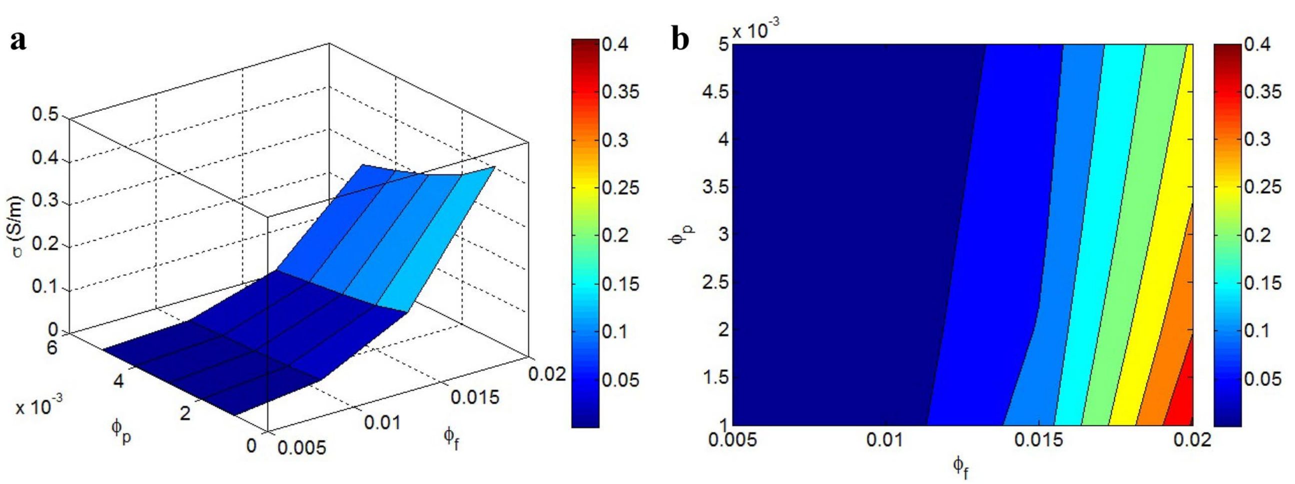

Fig. 3 Dependencies of conductivity on "ϕ_f" and "ϕ_p" by the present model: **a** 3D and **b** contour plots

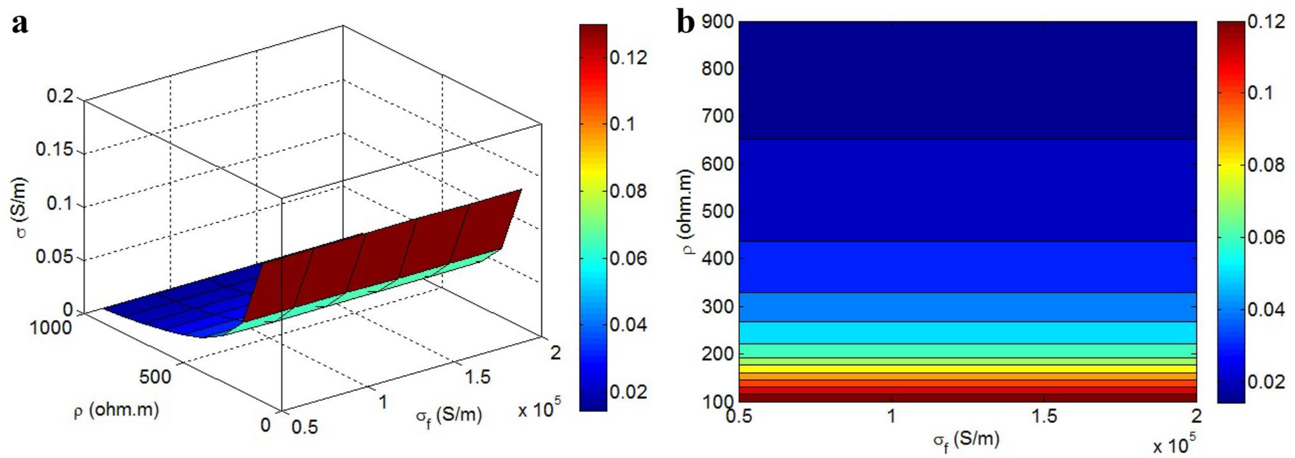

Fig. 4 Calculations of conductivity by various "σ_f" and "ρ" ranges according to the developed model: **a** 3D and **b** contour plots

determining the conductivity of PCNT. Generally, the tunneling resistivity adversely manages the nanocomposite's conductivity, because it's high level limits the electron transportation via tunneling region. These signs accept the forecasts of advanced model for conductivity at dissimilar extents of tunneling resistivity.

Figure 5 displays the conductivity variation at the changed intensities of "d" and "λ" by the advanced model. Large "d" and short "λ" improve the conductivity, but short "d" introduces an insulated nanocomposite. Actually, wide and short tunnels positively affect the conductivity, while only short tunneling diameter causes insulating. So, the dimensions of tunneling region change the conductivity of PCNT. These outputs imply that very short tunneling diameter between adjacent CNT in nanocomposites cannot produce a desirable conductivity, but both long tunneling diameter and small tunneling distance maximize the conductivity.

Wide tunnels enlarge the contact area between CNT, which strengthens the charge conveyance through tunnels. Indeed, wide tunnels enhance the efficiency of CNT covering the tunneling region for transportation of electrons. Nevertheless, small tunneling width producing small interaction zone poorly transfers the electrons through tunneling region weakening the conductivity. It can be said that the tunneling diameter manipulating the contact area between adjacent CNT dominantly controls the extent of electron transferring via tunneling zone. Hence, the tunneling diameter directly governs the nanocomposite's conductivity confirming the advanced model.

The tunneling space between CNT commonly contains the insulated polymer film. A bulky tunnel reveals a dense insulated polymer layer amid CNT, which obviously worsens the tunneling efficacy. Therefore, lengthy tunnels directly increase the conflicts limiting the charge shifting. Conversely,

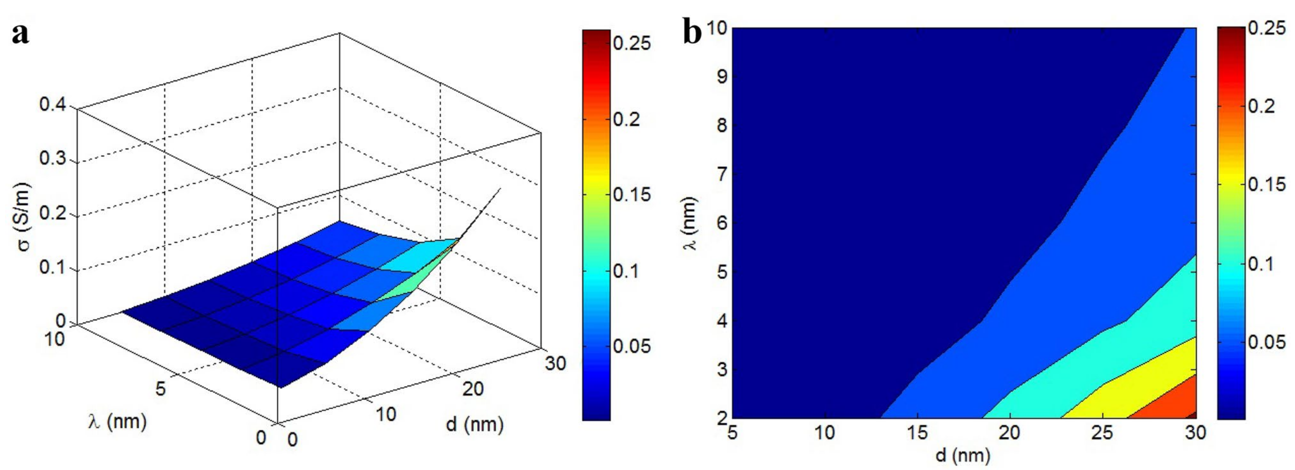

Fig. 5 Association of nanocomposite's conductivity to "d" and "λ" factors by the present model: **a** 3D and **b** contour plots

a short tunneling distance displays a thin polymer film between adjacent CNT improving the tunneling effect. These observations validate the opposite correlation between conductivity and tunneling distance. Many models in literature have focused on the tunneling length/distance to investigate the tunneling efficacy in nanocomposites [3, 53, 54]. In fact, many studies have disregarded the roles of polymer tunneling resistivity and tunneling diameter in the tunneling conductivity. So, the tunneling distance is a main parameter governing the electron tunneling in nanocomposites.

Figure 6 reveals the "f" and "R_{tun}" impacts on the conductivity complying with the advanced model. $f=0.5$ and $R_{tun}=2$ Ω produce the conductivity of 0.7 S/m, but an insulated sample is detected at $f<0.24$ or $R_{tun}>15$ Ω. These results demonstrate that the big nets and poor tunneling resistance positively handle the conductivity, while the low part of linked CNT or high tunneling resistance causes insulating. In fact, the formation of large conductive networks and providing poor tunneling resistance are necessary to improve the conductivity. However, small networks or high tunneling resistance cannot grow the charge transferring in polymer nanocomposites.

The ratio of linked CNT and the extent of nets straightly handle the conductivity. The conductivity of nanocomposites mainly associates to the net magnitude [55, 56] and thus "f" is an important parameter managing the conductivity. The high number of CNT in the nets facilitates the charge transferring in nanocomposites, but low "f" produces small networks limiting the transferring of electrons. Moreover, very small percentage of networked CNT cannot effectively transport the electrons producing an insulating. Besides, the tunneling conflict significantly governs the electron transporting within the samples.

A poor resistance raises the charge transferring thorough tunneling region, while a high tunneling resistance mainly limits the tunneling effect. As mentioned, the total conflict of components (filler and tunnels) only depends on the tunneling resistance, because the extraordinary CNT conductivity introduces too weak conflict. Accordingly, the tunneling resistance negatively affects the conductivity. More, very high tunneling resistance does not allow the electron tunneling, which induces an insulating. These explanations show the correct associations of conductivity to "f" and "R_{tun}" terms approving the advanced model.

Comparisons between calculations and experimental data

The experimental data of conductivity are linked to the model's calculations. Poly (vinyl chloride) (PVC)/multi walled CNT (MWCNT) ($l=16$ µm, $R=8$ nm, $u=1.2$ and $\phi_p=0.0005$) [57], epoxy (DGEBA-based epoxy resin cured with an amine hardener)/single walled CNT (SWCNT) ($l=2$ µm, $R=1$ nm, $u=1.6$ and $\phi_p=0.0003$) [58], epoxy (bisphenol A type epoxy resin cured with an amine type hardener)/MWCNT ($l=30$ µm, $R=8$ nm, $u=1.2$ and $\phi_p=0.0002$) [59] and poly (ethylene terephthalate) (PET)/MWCNT ($l=1$ µm, $R=5$ nm, $u=1.2$ and $\phi_p=0.001$) [60] samples were chosen from valid studies. More details about the materials and characterizations were provided in the original references. Equation 14 can calculate the interphase thickness for samples by percolation threshold and CNT dimensions. The ranks of "t" are predicted as 3.5, 3.2, 7 and 22 nm for PVC/MWCNT, epoxy/SWCNT, epoxy/MWCNT and PET/MWCNT samples, in that order. These grades display the development of dissimilar interphase zone in these samples. PET/MWCNT and epoxy/SWCNT nanocomposites present the thickest and the thinnest interphase, respectively. These outcomes indicate that the interphase region differently add to the nets and conductivity of samples.

Figure 7 illustrates the trial data as well as the conductivity estimations for the reported samples. The predictions of

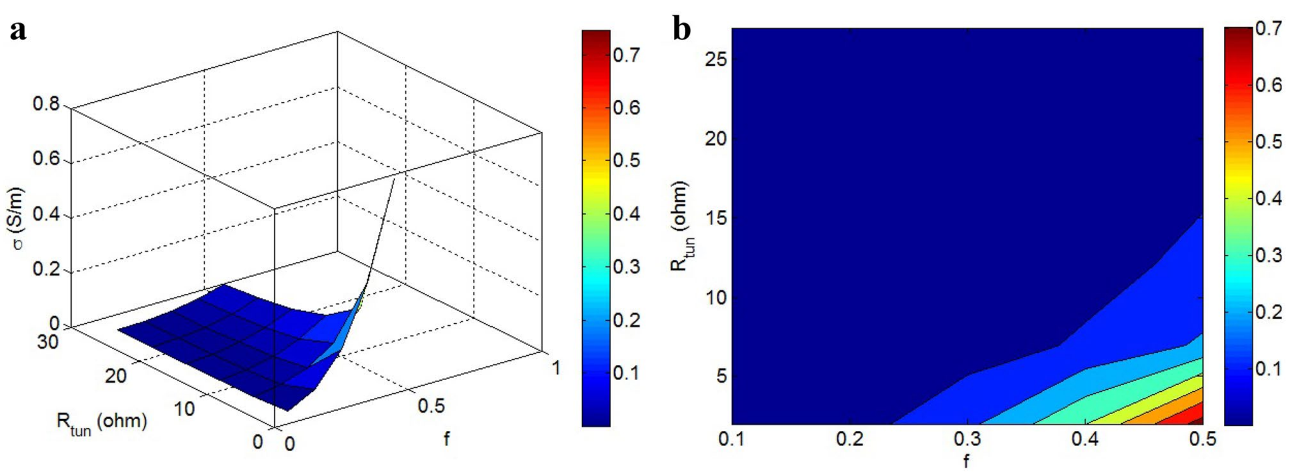

Fig. 6 Conductivity of PCNT supposing "f" and "R_{tun}" variables by the established model: **a** 3D and **b** contour plots

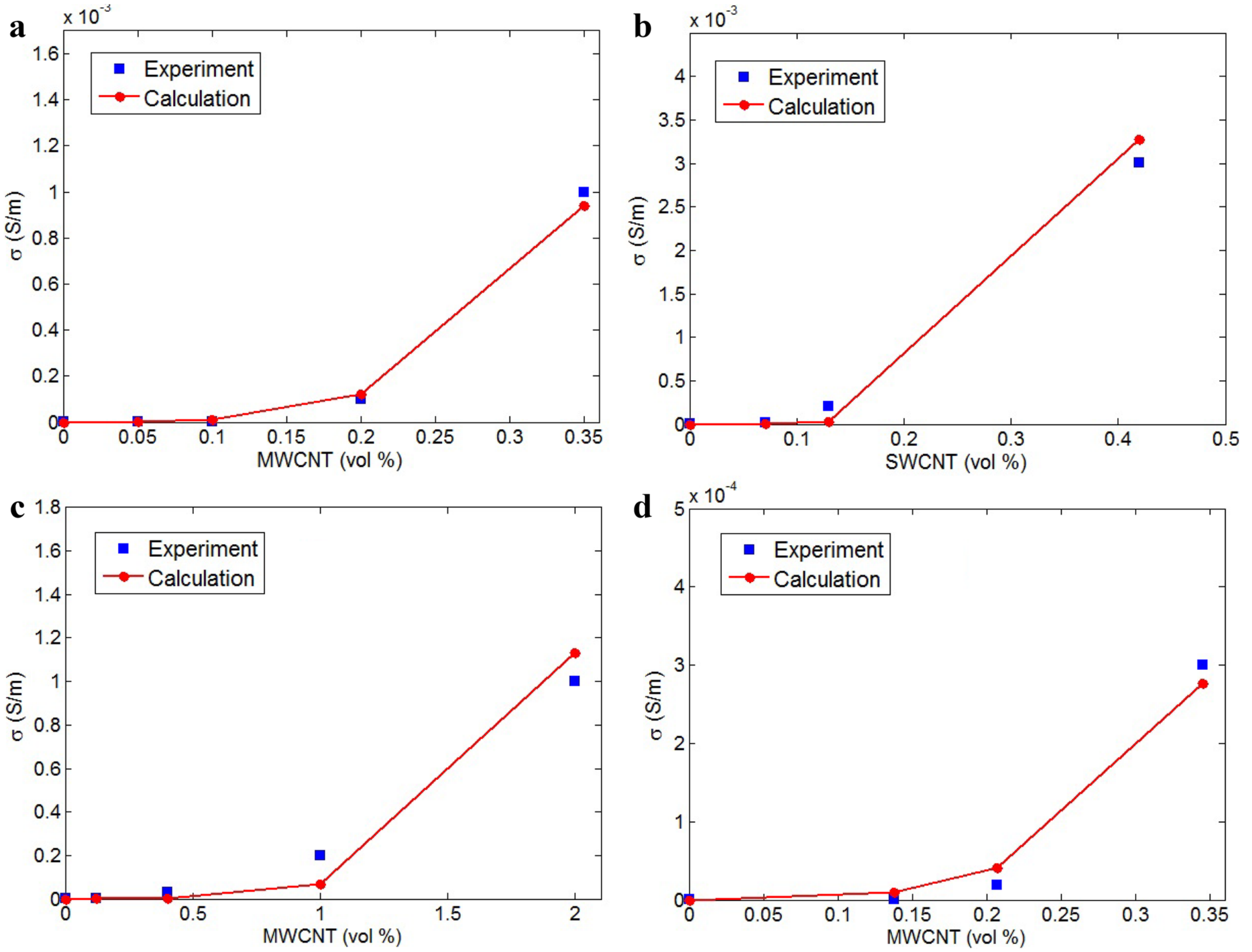

Fig. 7 Comparison between experimental data and calculations of conductivity by the established model for **a** PVC/MWCNT [57], **b** epoxy/SWCNT [58], **c** epoxy/MWCNT [59] and **d** PET/MWCNT [60] samples

developed model precisely comply with the experimental grades at whole samples. The proper agreements between empirical and theoretical data approve the developed model. Therefore, the advanced model correctly suggests the conductivity considering the conductive nets, interphase and tunneling region. This model is attractive, because the existed models cannot adequately consider the roles of effective terms in the conductivity. The ranks of (ρ, λ, d) are calculated as (150, 3.5, 12), (1750, 8, 0.6), (2500, 7, 10) and (3000, 7.5, 5) (ohm.m, nm, nm) for PVC/MWCNT, epoxy/SWCNT, epoxy/MWCNT and PET/MWCNT samples, correspondingly. The different tunneling properties show the dissimilar levels of tunneling resistance controlling the conductivity in the samples. PVC/MWCNT sample exhibits poor tunneling resistivity, short and wide tunnels declining the conflict and cultivating the conductivity. However, epoxy/SWCNT displays the highest tunneling resistance, which reveals the smallest level of charge moving among the specimens. As mentioned, the tunneling resistance dominantly handles the total resistance of components and the conductivity of nanocomposites.

Conclusions

Hui-Shia model was simplified and advanced for PCNT conductivity assuming the network fraction, interphase district and CNT tunnels. The rational influences of all factors on the conductivity and the acceptable agreements between empirical data and predictions support the developed model. Reedy CNT and poor waviness develop the conductivity, although only dense CNT produce an insulated sample. The calculations indicate that these factors expressively handle the conductivity. Thick interphase and large CNT grow the conductivity, but thin interphase or short CNT cannot modify it. Also, both high CNT loading and low percolation onset cultivate the conductivity, whereas a low filler concentration causes an insulated sample. Additionally, the polymer tunneling resistivity negatively manages the conductivity, while CNT conduction is a useless factor. Generally, polymer tunneling resistivity and CNT conductivity negligibly handle the conductivity. Moreover, wide and short tunnels positively handle the conductivity, whereas only very short tunneling diameter causes the insulating. The high ratio of linked CNT and poor tunnel conflicts positively govern the conductivity, while small nets or high conflicts in tunnels cause the insulated specimen.

References

1. Su F, Miao M (2014) Effect of MWCNT dimension on the electrical percolation and mechanical properties of poly (vinylidenefluoride-hexafluoropropylene) based nanocomposites. Synth Met 191:99–103
2. He L, Tjong SC (2013) Low percolation threshold of graphene/polymer composites prepared by solvothermal reduction of graphene oxide in the polymer solution. Nanoscale Res Lett 8(1):132
3. Zare Y, Rhee KY (2019) The effective conductivity of polymer carbon nanotubes (CNT) nanocomposites. J Phys Chem Solids 131:15–21
4. Zhang YF, Du FP, Chen L, Yeung KW, Dong Y, Law WC et al (2020) Supramolecular ionic polymer/carbon nanotube composite hydrogels with enhanced electromechanical performance. Nanotechnol Rev 9(1):478–488
5. Liu Y, Jiang X, Shi J, Luo Y, Tang Y, Wu Q et al (2020) Research on the interface properties and strengthening–toughening mechanism of nanocarbon-toughened ceramic matrix composites. Nanotechnol Rev 9(1):190–208
6. Jiang Q, Tallury SS, Qiu Y, Pasquinelli MA (2020) Interfacial characteristics of a carbon nanotube-polyimide nanocomposite by molecular dynamics simulation. Nanotechnol Rev 9(1):136–145
7. Qian J, Pu J-H, Zha X-J, Bao R-Y, Liu Z-Y, Yang M-B et al (2019) Effect of aspect ratio of multi-wall carbon nanotubes on the dispersion in ethylene-α-octene block copolymer and the properties of the Nanocomposites. J Polym Res 26(12):275
8. Tondnevis F, Keshvari H, Mohandesi JA (2019) Physico-mechanical and in vitro characterization of electrically conductive electrospun nanofibers of poly urethane/single walled carbon nano tube by great endothelial cells adhesion for vascular tissue engineering. J Polym Res 26(11):256
9. Jebril S, Doudou BB, Zghal S, Dridi C (2019) Non-isothermal crystallization kinetics of hybrid carbon nanotube-silica/polyvinyl alcohol Nanocomposites. J Polym Res 26(11):254
10. Zidan HM, Abdelrazek EM, Abdelghany AM, Tarabiah AE (2019) Characterization and some physical studies of PVA/PVP filled with MWCNTs. J Mater Res Technol 8(1):904–913
11. Liu S, Wu G, Chen X, Zhang X, Yu J, Liu M et al (2019) Degradation Behavior In Vitro of Carbon Nanotubes (CNTs)/Poly (lactic acid) (PLA) Composite Suture. Polymers 11(6):1015
12. Irzhak VI, Dzhardimalieva GI, Uflyand IE (2019) Structure and properties of epoxy polymer nanocomposites reinforced with carbon nanotubes. J Polym Res 26(9):220
13. Arumugam S, Ramamoorthy P, Chakkarapani LD (2019) Biodegradable dendrimer functionalized carbon nanotube-hybrids for biomedical applications. J Polym Res 26(8):182
14. Poothanari MA, Xavier P, Bose S, Kalarikkal N, Komalan C, Thomas S (2019) Compatibilising action of multiwalled carbon nanotubes in polycarbonate/polypropylene (PC/PP) blends: phase morphology, viscoelastic phase separation, rheology and percolation. J Polym Res 26(8):178
15. Handique J, Dolui SK (2019) A thermally remendable multiwalled carbon nanotube/epoxy composites via Diels-Alder bonding. J Polym Res 26(7):163
16. Behdinan K, Moradi-Dastjerdi R, Safaei B, Qin Z, Chu F, Hui D (2020) Graphene and CNT impact on heat transfer response of nanocomposite cylinders. Nanotechnol Rev 9(1):41–52
17. Lee SY, Hwang JG (2019) Finite element nonlinear transient modelling of carbon nanotubes reinforced fiber/polymer composite spherical shells with a cutout. Nanotechnol Rev 8(1):444–451
18. Zare Y, Rhee KY (2018) A power model to predict the electrical conductivity of CNT reinforced nanocomposites by considering interphase, networks and tunneling condition. Engineering, Composites Part B
19. Kim S, Zare Y, Garmabi H, Rhee KY (2018) Variations of tunneling properties in poly (lactic acid)(PLA)/poly (ethylene oxide)(PEO)/carbon nanotubes (CNT) nanocomposites during hydrolytic degradation. Sens Actuators, A 274:28–36
20. Razavi R, Zare Y, Rhee KY (2019) The roles of interphase and filler dimensions in the properties of tunneling spaces between CNT in polymer nanocomposites. Polym Compos
21. Ryvkina N, Tchmutin I, Vilčáková J, Pelíšková M, Sáha P (2005) The deformation behavior of conductivity in composites where charge carrier transport is by tunneling: theoretical modeling and experimental results. Synth Met 148(2):141–146
22. Balberg I, Azulay D, Toker D, Millo O (2004) Percolation and tunneling in composite materials. Int J Mod Phys B 18(15):2091–2121
23. Zare Y, Rhee KY, Park S-J (2019) Modeling the roles of carbon nanotubes and interphase dimensions in the conductivity of nanocomposites. Res Phys 15:102562
24. Zare Y, Rhee KY (2017) Development and modification of conventional Ouali model for tensile modulus of polymer/carbon nanotubes nanocomposites assuming the roles of dispersed and networked nanoparticles and surrounding interphases. J Colloid Interface Sci 506:283–290
25. Zare Y (2015) Assumption of interphase properties in classical Christensen–Lo model for Young's modulus of polymer nanocomposites reinforced with spherical nanoparticles. RSC Adv 5(116):95532–95538
26. Seidel G, Puydupin-Jamin AS (2011) Analysis of clustering, interphase region, and orientation effects on the electrical conductivity of carbon nanotube–polymer nanocomposites via computational micromechanics. Mech Mater 43(12):755–774
27. Hassanzadeh-Aghdam MK, Mahmoodi MJ, Ansari R (2019) Creep performance of CNT polymer nanocomposites-An emphasis on viscoelastic interphase and CNT agglomeration. Compos B Eng 168:274–281
28. Nikfar N, Zare Y, Rhee KY (2018) Dependence of mechanical performances of polymer/carbon nanotubes nanocomposites on percolation threshold. Phys B Condens Matter
29. Razavi R, Zare Y, Rhee KY (2018) A model for tensile strength of polymer/carbon nanotubes nanocomposites assuming the percolation of interphase regions. Colloids Surf, A 538:148–154
30. Zare Y, Rhee KY, Park SJ (2019) A developed equation for electrical conductivity of polymer carbon nanotubes (CNT) nanocomposites based on Halpin-Tsai model. Res Phys 102406
31. Qiao R, Brinson LC (2009) Simulation of interphase percolation and gradients in polymer nanocomposites. Compos Sci Technol 69(3):491–499
32. Baxter SC, Robinson CT (2011) Pseudo-percolation: Critical volume fractions and mechanical percolation in polymer nanocomposites. Compos Sci Technol 71(10):1273–1279
33. Maiti S, Suin S, Shrivastava NK, Khatua B (2013) Low percolation threshold in polycarbonate/multiwalled carbon nanotubes nanocomposites through melt blending with poly (butylene terephthalate). J Appl Polym Sci 130(1):543–553

34. Maiti S, Shrivastava NK, Khatua B (2013) Reduction of percolation threshold through double percolation in melt-blended polycarbonate/acrylonitrile butadiene styrene/multiwall carbon nanotubes elastomer nanocomposites. Polym Compos 34(4):570–579
35. Jang SH, Yin H (2015) Effective electrical conductivity of carbon nanotube-polymer composites: a simplified model and its validation. Mater Res Express 2(4):045602
36. Feng C, Jiang L (2013) Micromechanics modeling of the electrical conductivity of carbon nanotube (CNT)–polymer nanocomposites. Compos A Appl Sci Manuf 47:143–149
37. Seidel GD, Lagoudas DC (2009) A micromechanics model for the electrical conductivity of nanotube-polymer nanocomposites. J Compos Mater 43(9):917–941
38. Zare Y, Rhee KY (2019) Simplification and development of McLachlan model for electrical conductivity of polymer carbon nanotubes nanocomposites assuming the networking of interphase regions. Compos B Eng 156:64–71
39. Zare Y, Rhee KY (2020) Development of Expanded Takayanagi Model for Tensile Modulus of Carbon Nanotubes Reinforced Nanocomposites Assuming Interphase Regions Surrounding the Dispersed and Networked Nanoparticles. Polymers 12(1):233
40. Zare Y, Rhee KY (2020) Modeling the effect of interfacial conductivity between polymer matrix and carbon nanotubes on the electrical conductivity of nanocomposites. RSC Adv 10(1):424–433
41. Zare Y (2015) Estimation of material and interfacial/interphase properties in clay/polymer nanocomposites by yield strength data. Appl Clay Sci 115:61–66
42. Zare Y (2016) A model for tensile strength of polymer/clay nanocomposites assuming complete and incomplete interfacial adhesion between the polymer matrix and nanoparticles by the average normal stress in clay platelets. RSC Adv 6(63):57969–57976
43. Zare Y, Rhee KY, Park SJ (2018) A modeling methodology to investigate the effect of interfacial adhesion on the yield strength of MMT reinforced nanocomposites. J Ind Eng Chem
44. Shia D, Hui C, Burnside S, Giannelis E (1998) An interface model for the prediction of Young's modulus of layered silicate-elastomer nanocomposites. Polym Compos 19(5):608–617
45. Mohiuddin M, Hoa SV (2013) Estimation of contact resistance and its effect on electrical conductivity of CNT/PEEK composites. Compos Sci Technol 79:42–48
46. Al-Saleh MH (2015) Influence of conductive network structure on the EMI shielding and electrical percolation of carbon nanotube/polymer nanocomposites. Synth Met 205:78–84
47. Zare Y, Rhee KY (2017) Development of a conventional model to predict the electrical conductivity of polymer/carbon nanotubes nanocomposites by interphase, waviness and contact effects. Applied Science and Manufacturing, Composites Part A
48. Ji XL, Jiao KJ, Jiang W, Jiang BZ (2002) Tensile modulus of polymer nanocomposites. Polym Eng Sci 42(5):983
49. Berhan L, Sastry A (2007) Modeling percolation in high-aspect-ratio fiber systems. I. Soft-core versus hard-core models. Phys Rev E 75(4):041120
50. Rafiee R (2013) Influence of carbon nanotube waviness on the stiffness reduction of CNT/polymer composites. Compos Struct 97:304–309
51. Li J, Ma PC, Chow WS, To CK, Tang BZ, Kim JK (2007) Correlations between percolation threshold, dispersion state, and aspect ratio of carbon nanotubes. Adv Func Mater 17(16):3207–3215
52. Takeda T, Shindo Y, Kuronuma Y, Narita F (2011) Modeling and characterization of the electrical conductivity of carbon nanotube-based polymer composites. Polymer 52(17):3852–3856
53. Zare Y, Rhee KY (2020) Significances of interphase conductivity and tunneling resistance on the conductivity of carbon nanotubes nanocomposites. Polym Compos
54. Zare Y, Rhee KY (2017) Development of a Model for Electrical Conductivity of Polymer/Graphene Nanocomposites Assuming Interphase and Tunneling Regions in Conductive Networks. Ind Eng Chem Res 56(32):9107–9115
55. Pu JH, Zha XJ, Zhao M, Li S, Bao RY, Liu ZY et al (2018) 2D end-to-end carbon nanotube conductive networks in polymer nanocomposites: a conceptual design to dramatically enhance the sensitivities of strain sensors. Nanoscale
56. Deng H, Lin L, Ji M, Zhang S, Yang M, Fu Q (2014) Progress on the morphological control of conductive network in conductive polymer composites and the use as electroactive multifunctional materials. Prog Polym Sci 39(4):627–655
57. Mamunya Y, Boudenne A, Lebovka N, Ibos L, Candau Y, Lisunova M (2008) Electrical and thermophysical behaviour of PVC-MWCNT nanocomposites. Compos Sci Technol 68(9):1981–1988
58. Gojny FH, Wichmann MH, Fiedler B, Kinloch IA, Bauhofer W, Windle AH et al (2006) Evaluation and identification of electrical and thermal conduction mechanisms in carbon nanotube/epoxy composites. Polymer 47(6):2036–2045
59. Kim YJ, Shin TS, Do Choi H, Kwon JH, Chung YC, Yoon HG (2005) Electrical conductivity of chemically modified multiwalled carbon nanotube/epoxy composites. Carbon 43(1):23–30
60. Logakis E, Pissis P, Pospiech D, Korwitz A, Krause B, Reuter U et al (2010) Low electrical percolation threshold in poly (ethylene terephthalate)/multi-walled carbon nanotube nanocomposites. Eur Polymer J 46(5):928–936