Development of a conventional model to predict the electrical conductivity of polymer/carbon nanotubes nanocomposites by interphase, waviness and contact effects

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A model for electrical resistivity of composites based on contacts between fibers is developed in this paper for electrical conductivity of polymer/CNT nanocomposites. The developed model considers the influences of interphase regions and CNT waviness on percolation threshold, effective CNT concentration and network dimension. Many experimental results are applied to assess the developed model. Also, the developed model is used to study the influences of all parameters on the conductivity of nanocomposite. It is shown that thin interphase and small diameter of contact area result in poor conductivity. In addition, a desirable conductivity is obtained by the high fraction of percolated CNT in the conductive network as well as the large number of contacts between nanotubes in the nanocomposite.

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1. Introduction

Polymer nanocomposites containing carbon nanotubes (PCNT) are appealing much attention, due to their extraordinary mechanical, thermal and electrical properties which develop potential applications in electronics, shielding, conductive products, etc. [1–6]. The high aspect ratio of CNT causes the enhancement of properties by a little amount of CNT compared to conventional particles such as carbon black and clays. There are various nanotube types including single wall nanotubes (SWCNT), double wall nanotubes (DWCNT) and multi wall nanotubes (MWCNT) which can produce a high level of specific surface area (area per weight). Also, their surfaces can be functionalized with some functional groups to improve their interaction with polymer matrices [7,8].

The electrical conductivity of PCNT is based on the percolated paths of conductive CNT. A three-dimensional (3D) network of CNT usually forms in polymer matrix above a determinate concentration as percolation threshold which strongly depends on the aspect ratio of CNT and their dispersal level in polymer matrix [9–11]. Interestingly, the large aspect ratio of CNT in a range of 500–1000 can create a conductive network by very low loading of CNT. The percolation level can be experimentally assessed by measurement of electrical conductivity. However, the main mechanism for conductivity of PCNT is electron tunneling, where electrons are transferred between nanotubes by hopping [12]. In this method, the nanotubes are not bodily coupled and neighboring CNT transfers the charges by electron jumping.

The interphase regions are commonly formed in polymer nanocomposites, due to the outstanding surface area of nanoparticles and strong interfacial adhesion between matrix and filler phases [13–16]. The mechanical properties of polymer nanocomposites such as tensile modulus and strength effectively depend on the interphase properties [17–19]. So, the interphase regions play a remarkable reinforcing effect in nanocomposites. Besides, the positive role of interphase regions in the percolating structure of nanoparticles was reported, because the interphase regions can create a continuous network before the physical connection of nanoparticles was reported, because the interphase regions can create a continuous network before the physical connection of nanoparticles [20,21]. Therefore, a lower percolation threshold is observed in nanocomposites containing interphase which displays the significance of interphase in the conductivity. However, the contributions of interphase regions to the electrical conductivity of nanocomposites have not been considered.

The electrical conductivity of polymer nanocomposites particularly PCNT has been analyzed by several models. The widely used methodology is a conventional power-law model based on percolation theory of composites which expresses the conductivity by
filler concentration, percolation threshold and an exponent [22]. This model demonstrates a good fitting with the electrical conductivity of PCNT [23–25]; however, it cannot reflect the excellent physical aspects of CNT such as nano-size and surface area. Some researchers also developed the micromechanics models for conductivity of PCNT assuming different parameters such as arrangement and waviness of CNT, interphase and tunneling distance [26–28], but they generally expressed some intricate equations which are not appropriate in practice. Feng and Jiang [27] assumed the electron tunneling in PCNT by an interphase layer around CNT. Their results suggested that both electron tunneling and conductive networks contribute to the conductivity of nanocomposites, but the conductive networks are dominant at high CNT fractions. It was also indicated that the size of CNT have significant effect on the conductivity of nanocomposites. Moreover, Takeda et al. [28] considered the tunneling distance in PCNT by extending the CNT and suggested a model for conductivity of nanocomposites. Also, they formulated the tunneling distance as a function of filler volume fraction. However, some complex and unclear equations in these studies limit their application for PCNT. In fact, there is not a simple and accurate model for electrical conductivity of PCNT.

Kim et al. [29] also suggested an analytical homogenization approach to predict percolation threshold effect, tunneling role and effective electrical conductivity of polymer nanocomposites. They showed a good agreement between experimental results and calculations. Also, the thermal conductivity of polymer nanocomposites containing CNT, graphene or both of them (synergetic effect) were theoretically studied assuming the geometries of nanoparticles [29–33]. However, the available models cannot properly present the influences of interphase and waviness on the percolation threshold and conductivity.

Weber and Kamal [34] suggested a model for resistivity of polymer fiber composites which assumes the contacts between fibers and the dimensions of fiber and network. This study aims to develop this model for PCNT by the influences of interphase and CNT waviness on percolation threshold, effective CNT concentration and network level. Thus, this model is adjusted for polymer nanocomposites and proper equations for the mentioned terms are suggested. Actually, the developed methodology simply presents the percolation threshold, the volume fraction of networked CNT and conductivity of nanocomposites by filler size, waviness, interphase thickness, network fraction and contact number, while the previous models did not assume these terms for conductivity. The developed model is examined by experimental results. Moreover, the relations between the conductivity of PCNT and different parameters are explained to show the predictability of the developed model. We hope that the presented model can be applied in future studies on PCNT, because the available models cannot properly calculate the conductivity of PCNT.

2. Developed model

Most models in literature do not assume the particle-particle contacts in composites. In addition, the contacts between fibers are probably body-to-body rather than end-to-end and end-to-body [34]. As a result, the contact area is much lesser than that of end-to-end arrangement which affects the conductivity.

Weber and Kamal [34] suggested the longitudinal and transverse resistivity of polymer fiber composites assuming fiber-fiber contacts as:

\[ P_{\text{long}} = \frac{\pi R^2 \rho_N X}{\phi_N d_i \cos^2 \theta} \]  \hspace{1cm} (1)

where “R” is fiber radius, “\( \rho_N \)” is resistivity of fiber, “\( \phi_N \)” is the volume fraction of networked fibers, “d,” is diameter of contact circle, “l” is fiber length and “\( \theta \)” is angle of fiber orientation. “X” is also related to the average number of contacts between CNT in the nanocomposite (m) as:

\[ X = \frac{1}{0.59 + 0.15m} \]  \hspace{1cm} (2)

where the maximum “m” was indicated as 15.

This model can be developed for PCNT containing random distribution of CNT assuming the percolation threshold, interphase and waviness of CNT. In the case of random distribution of CNT in the nanocomposite, it can be approximated that \( \cos(\theta) = 1/3 \) [35]. Also, the resistance of CNT and nanocomposites are inversely related to their conductivities restructuring the latter equation to:

\[ \sigma = \frac{\phi_N d_i \sigma_N}{3 \pi R^2 X} \]  \hspace{1cm} (3)

where “\( \sigma_N \)” is conductivity of CNT. The formation of interphase and CNT waviness commonly affects the general properties of PCNT. The influences of these terms in percolation threshold, effective fraction of CNT and the percentages of networked CNT are given in the following.

The percolation threshold in PCNT containing random orientation of nanoparticles can be proposed [36] as:

\[ \phi_p = \frac{V}{V_{\text{ex}}} \]  \hspace{1cm} (4)

where “V” and “\( V_{\text{ex}} \)” are the volume and excluded volume of CNT, respectively. The excluded volume comprises the capacity neighboring a nanotube into which the center of a similar particle cannot enter.

“V” and “\( V_{\text{ex}} \)” for soft-core rigid spheroid-cylinders randomly oriented in three dimensions were suggested [36] as:

\[ V = \pi R^3 l + \frac{4}{3} \pi R^3 \]  \hspace{1cm} (5)

\[ V_{\text{ex}} = \frac{32}{3} \pi R^3 \left[ 1 + \frac{3}{4} \left( \frac{l}{R} \right) + \frac{3}{32} \left( \frac{l}{R} \right)^2 \right] \]  \hspace{1cm} (6)

The interphase regions around CNT frequently shift the development of a conductive network to lower filler fractions which should be considered in percolation effect. The interphase layer decreases the excluded volume [37] as:

\[ V_{\text{ex}} = \frac{32}{3} \pi (R + t)^3 \left[ 1 + \frac{3}{4} \left( \frac{l}{R + t} \right) + \frac{3}{32} \left( \frac{l}{R + t} \right)^2 \right] \]  \hspace{1cm} (7)

where “t” is interphase thickness. The interphase forms around the nanoparticles in polymer nanocomposite. The interphase thickness is assumed as the thickness of interphase regions surrounding CNT from the CNT surface to polymer matrix. So, the interphase is an intermediate phase between CNT and polymer matrix, which shows different properties than matrix and nanoparticles [38].

Furthermore, the high aspect ratio of CNT (length to diameter) causes waviness which declines the effectiveness of nanotubes in nanocomposites. An equivalent nanotube with effective length of \( l_{eq} \) can be assumed for a curved nanotube according to Fig. 1a which defines a waviness parameter as:

\[ u = \frac{l}{l_{eq}} \]  \hspace{1cm} (8)

where “u = 1” shows an straight nanotube (no waviness), while the higher levels of “u” exhibit more waviness and less effective length.
The interphase waviness and waviness do not change the volume of CNT, but “\(V_{ex}\)” assuming interphase and CNT waviness can be presented supposing \(l_{eq} = l/u\) as:

\[
V_{ex} = \frac{32}{3} \pi (R + t)^3 \left[ 1 + \frac{3}{4} \left( \frac{l}{u} \right) + \frac{3}{2} \left( \frac{l}{R + t} \right)^3 \right]
\]

(9)

Now, the percolation threshold by the influences of interphase and waviness can be expressed as:

\[
\phi_p = \frac{\pi R^3 l + (4/3) \pi R^3}{2} \left[ 1 + \frac{2}{3} \left( \frac{l}{u} \right) + \frac{2}{3} \left( \frac{l}{R + t} \right)^3 \right]
\]

(10)

It is shown in the next section that this equation can present useful calculations for percolation threshold in PCNT.

Both CNT and interphase layer can be included in effective CNT (Fig. 1b), because they simultaneously affect the overall properties of PCNT. The volume fraction of effective CNT assuming interphase and waviness [27] can be stated as:

\[
\phi_{eff} = \frac{(R + t)^3 (l/u + 2t)}{R^3 l/u} \phi_f
\]

(11)

where “\(\phi_f\)” is CNT volume fraction in PCNT.

Also, a number of CNT participates in the conductive network after percolation threshold. “\(t\)” parameter as the percentages of percolated CNT [27] is considered by:

\[
f = \frac{\phi_{eff}^{1/3} - \phi_f^{1/3}}{1 - \phi_f^{1/3}}
\]

(12)

Therefore, the volume fraction of network nanoparticles in PCNT can be expressed as:

\[
\phi_N = f \phi_{eff}
\]

(13)

The waviness or waviness also worsens the nature conductivity of CNT [26]. The role of waviness in “\(\sigma_N\)” can be stated as:

\[
\sigma_{Na} = \frac{\sigma_{Na}}{u}
\]

(14)

By substituting of Eqs. (13) and (14) into (3), a developed model is derived which can predict the electrical conductivity of PCNT by the influences of interphase, waviness and contact properties as:

\[
\sigma = f \phi_{eff} d \sigma_N \frac{d \sigma_{Na}}{3 \pi R^2 Xu}
\]

(15)

This simple model can be easily used to estimate the electrical conductivity of PCNT. Also, this model shows the dependence of PCNT conductivity to different parameters.

3. Results and discussion

3.1. Evaluation of developed model by experimental results

The developed model is applied to predict the conductivity in some samples.

Several samples including ultrahigh molecular weight polyethylene/MWCNT (\(R = 8\) nm, \(l = 8\) \(\mu\)m and \(u = 1.2\)) from [39], polycarbonate/acylonitrile butadiene styrene/MWCNT (\(R = 5\) nm, \(l = 1.5\) \(\mu\)m and \(u = 1.2\)) from [22], poly (lactic acid)/MWCNT (\(R = 15\) nm, \(l = 5\) \(\mu\)m and \(u = 1.5\)) from [40] and ultrahigh molecular weight polyethylene/MWCNT (\(R = 25\) nm, \(l = 100\) \(\mu\)m and \(u = 1.15\)) from [41] were chosen for analysis. The interphase thickness (\(t\)) can be determined by comparing the experimental levels of “\(\phi_p\)” through measurement of electrical conductivity with the calculations of Eq. (10). The experimental results of “\(\phi_p\)” were reported as 0.0007, 0.002, 0.0048 and 0.0002 for ultrahigh molecular weight polyethylene/MWCNT, polycarbonate/acylonitrile butadiene styrene/MWCNT, poly (lactic acid)/MWCNT and ultrahigh molecular weight polyethylene/MWCNT samples, respectively. When these results are applied to Eq. (10), “\(t\)” values of 7, 5, 5 and 15 nm are calculated for the reported samples, respectively. These ranges are correct, because the interphase thickness cannot exceed the gyration radius of polymer chains [42]. So, Eq. (10) can be used to estimate the interphase thickness in PCNT by the experimental levels of percolation threshold. These results show that the interphase forms in these samples and the percolation threshold cannot be calculated without the assumption of interphase regions. The conductivity of MWCNT is assumed as \(10^{50}\) S/m. Moreover, “\(d_c\)” as the diameter of contact circle was suggested as \(4 \times 10^{-5}\) times of fiber diameter in the conventional model [34]. Since “\(d_c\)” between CNT is much less than that of fibers, \(d_c = 10^{-6}\) R is considered in the developed model.

Using the dimensions of CNT and interphase, the electrical conductivity can be calculated for the reported samples. Fig. 2 illustrates the acceptable arrangement between the predictions and the experimental results at all CNT fractions. The maximum difference between experimental and theoretical values was considered as 10%, which is logical. It can be suggested that the developed model accounting the influences of CNT waviness and interphase can suitably predict the electrical conductivity of PCNT. In addition, the values of “\(m\)” as number of contacts between CNT can be estimated by the developed model. “\(m\)” parameter is calculated as 60, 46, 30,000 and 400 for ultrahigh molecular weight polyethylene/ MWCNT, polycarbonate/acylonitrile butadiene styrene/MWCNT, poly (lactic acid)/MWCNT and ultrahigh molecular weight polyethylene/MWCNT samples, respectively. Weber and Kamal [34] suggested the maximum level of “\(m\)” for conventional fiber com-
posites as 15. However, the number of contacts increases in PCNT, due to the larger aspect ratio of CNT in comparison to micro-fibers. Conclusively, the developed equations can demonstrate proper calculations for electrical conductivity, interphase thickness and contact number.

### 3.2. Roles of parameters in the conductivity

The roles of different parameters in the electrical conductivity of PCNT are investigated by the developed model. It should be noted that the influences of various parameters on the conductivity of nanocomposites have not been presented in the earlier works. The parametric study can guide the researchers to optimize the main parameters in the conductivity of nanocomposites. The constant levels of $\phi_f$, $R$, $l$, $u$, $t$, $m$, $r$ and $d_c$ are considered as 0.02, 10 nm, 10 µm, 1.4, 5 nm, 50, 105 S/m and 10^{-5} nm for all calculations.

Fig. 3 depicts the influences of $R$ and $u$ parameters on the electrical conductivity based on the developed model at average levels of other factors by 3D and contour plots. A very poor conductivity near to insulating is observed at $R > 30$ nm and $u > 1.5$, which demonstrates the negative effects of high levels of $R$ and $u$ on conductivity. However, the best conductivity as 1.1 S/m is observed at $R = 10$ nm and $u = 1$. As a result, thin and straight CNT cause positive impacts on conductivity of PCNT, while the waviness of thick nanotubes cannot improve the conductivity of polymer matrices.

The thin CNT commonly produce a high level of surface area in PCNT, which increases the interphase fraction and also, affects the characteristics of polymer chains. So, thin CNT can decrease the percolation level by introducing a large surface area per volume and promoting the interphase regions. On the other hand, these advantages of thin nanotubes cause a high effective volume fraction of nanoparticles and more percentages of percolated filler. In addition, the nano-size strongly affects the interfacial interactions between polymer matrices and nanoparticles as well as the nature conductivity of nanotubes [42–44]. Accordingly, it is reasonable to obtain a high electrical conductivity by thin CNT. The same role of CNT size in the conductivity of PCNT was also explained in previous studies [45]. Moreover, the waviness decreases the effective length of nanotubes in PCNT. Since the reduction of effective nanotubes length weakens the percolation level and nature conduction of CNT in addition to the fraction of networked CNT, the waviness drops the potential advantages of CNT in PCNT which depends to their excellent length. A high percolation threshold, poor conductivity and low networkability of waved CNT finally result in a weak conductivity in PCNT, as suggested by the developed model. Therefore, the present model properly predicts the effects of $R$ and $u$ parameters on the conductivity of PCNT.

Fig. 4 also exhibits the effects of $\phi_f$ and $\phi_p$ parameters on the conductivity of PCNT according to the developed model. $\phi_f = 0.025$ and $\phi_p = 0.001$ cause the highest conductivity as 0.7 S/m indicating that the best conductivity is achieved by the highest $\phi_f$ and the least $\phi_p$. However, an insulating is observed by slight $\phi_f$ and high $\phi_p$ which generally show the positive and negative effects of $\phi_f$ and $\phi_p$ parameters on the conductivity of PCNT, respectively.

These evidences establish that a large number of CNT and small percolation threshold produce a high conductivity in nanocomposites. The role of filler volume fraction is justified based on the exceptional conductivity of CNT which causes an insulating polymer matrix to be conductive. The CNT nanoparticles produce a conductive network in polymer matrix improving the electrical conductivity of resulted nanocomposite. However, the conductivity occurs above a special filler concentration which is named percolation threshold. In fact, the nanotubes before percolation cause an
insignificant conductivity in PCNT, but at the percolation threshold, the conductivity of PCNT rapidly raises and a conductive nanocomposite is made. Therefore, a little fraction of CNT as well as a high percolation threshold cannot supply suitable conditions for remarkable conductivity of PCNT, whereas a high level of filler concentration above a low percolation threshold easily produces a conductive network, which promotes the conductivity of PCNT. Accordingly, it is logical to say that a high filler fraction and low percolation threshold provide a high conductivity in PCNT. The similar results were also reported in other papers which confirm the current observations [46,47].

The conductivity of PCNT at dissimilar intensities of ‘‘l’’ and ‘‘rN’’ parameters as the length and conductivity of CNT is also shown in Fig. 5. The poor values of these parameters approximately introduce non-conductivity, but an extraordinary conductivity is obtained at the high levels of ‘‘l’’ and ‘‘rN’’. l < 10 μm and σN < 10^5 S/m result in a weak conductivity near to 0, but a great conductivity of 5 S/m is observed at l = 25 μm and σN = 3.7 * 10^7 - S/m. Conclusively, the length and conductivity of CNT directly affect the conductivity of PCNT demonstrating that long and super-conductive CNT are necessary to obtain high-conductive PCNT.

Clearly, long CNT are well percolated at a low volume fraction in PCNT. Also, they produce a big network which can effectively transfer more charges. Additionally, they provide a big interfacial area with polymer chains which results in strong interfacial interaction between polymer chains and conductive nanotubes. As a result, long CNT confidently grow the dimensions of filler network and characteristics of polymer chains which improve the conductivity of PCNT. Moreover, the high conductivity of CNT raises the conductivity of polymer chains and promotes the conductivity of produced nanocomposite. In other words, the conductivity of PCNT mainly depends on the conductivity of nanoparticles, because the polymers commonly are insulator. So, a higher conductivity of nanoparticles more motivates the conduction of polymer chains in PCNT. However, it was mentioned that the conductivity of CNT is weakened by defects, waviness and thick nanoparticles which should be considered when using CNT [26]. Convincingly, it can be confirmed that the developed model correctly shows the dependence of PCNT conductivity to CNT length and conduction.

Fig. 6 depicts the variation of conductivity at different ranges of ‘‘t’’ and ‘‘d_c’’ parameters as interphase thickness and diameter of contact area. These parameters show direct influences on the conductivity, where the low ranges of ‘‘t’’ and ‘‘d_c’’ cause poor conductivity, but the great levels of these parameters produce a considerable conductivity. As observed, t < 3 nm and d_c < 1.4 - * 10^-5 nm result in σ = 0.3 S/m, but t = 11 nm and d_c = 3 * 10^-5 nm produce the best conductivity as 4 S/m. Therefore, the interphase thickness and diameter of contact circle directly affect the conductivity of PCNT. In other words, thick interphase and large contact circle can increase the conductivity, while thin interphase and small contact area reduce it.
The positive effect of interphase thickness on the conductivity is clearly correlated to its roles in the percolation threshold, effective nanoparticles and volume fraction of networked CNT. A thick interphase around nanoparticles undoubtedly decreases the distance between nanotubes which facilitates the formation of filler network at low filler concentration. Also, a thicker interphase produces a better effective nanotube which increases the effectiveness of nanoparticles in conductivity of PCNT (Fig. 1b). Besides, a thick interphase positively contributes to the fraction of networked nanotubes, which strongly manages the conductivity. Generally, the interphase regions surrounding the nanotubes provide a potential condition for improvement of electrical conductivity in PCNT beside CNT. It means that the interphase grows the conducting efficiency of nanoparticles in PCNT.

It is also observed that “$d_c$” parameter directly controls the conductivity of PCNT, i.e. a better conductivity is obtained by a greater diameter of contact ring, which shows the positive effect of contact area on the conductivity of PCNT. The significance of contact area may be correlated to the tunneling effect between nanotubes as the main mechanism of conductivity in PCNT [27,48,49]. It was reported that the resistance of tunneling distance is inversely depended to contact areas [27]. In fact, a high level of “$d_c$” increases the contact area which declines the resistance of tunneling distance and improves the conductivity of PCNT. As a result, the developed model correctly shows the conductivity of PCNT as a function of interphase thickness and contact area.

The dependence of PCNT conductivity on “$f$” and “$m$” parameters as fraction of percolated CNT and number of contacts is exhibited in Fig. 7. The conductivity gets higher levels by more values of these parameters, while their small ranges meaningfully decrease the conductivity. The best conductivity as 1.6 S/m is observed at $f = 0.5$ and $m = 90$, while the low ranges of $f < 0.17$ and $m < 30$ cause poor conductivity. So, the fraction of percolated CNT and number of contacts directly change the conductivity of nanocomposites, as expected.

The level of “$f$” explicitly shows the size and density of filler network in PCNT, because “$f$” parameter shows the percentages of nanotubes included in the conductive network after percolation threshold. A high level of “$f$” indicates that a large number of nanoparticles are involved in networked regions which obviously demonstrate a large and dense network. However, a lesser level of “$f$” depicts more dispersed nanotubes in PCNT which produce a small network. Since a bigger network is able to transfer more charges, the electrical conductivity of PCNT shows a direct dependence on network dimension and “$f$” parameter. The literature studies also demonstrated the positive role of network size and density in the conductivity of nanocomposites [50]. Additionally, the high number of contacts between CNT increases the possibility of networking or tunneling effect, whereas the nanotubes with high separation distance cannot promote the conductivity even by electron hopping [27]. In fact, an upper value of “$m$” more progresses the efficiencies of CNT for production of a conductive
nanocomposite by a low loading of nanofiller. So, the developed model accurately justifies the roles of \( f \) and \( m \) parameters in the conductivity of PCNT.

4. Conclusions

The conventional model for electrical resistivity of fiber composites assuming the contacts between fibers was developed for PCNT taking into account the influences of interphase and CNT waviness on percolation threshold, effective concentration of CNT and network dimensions. An acceptable arrangement between the predictions and the experimental data is shown which confirms the predictability of the developed model. Also, the developed model can estimate the interphase thickness and number of contacts as calculated for the reported samples.

An insulating is observed at \( R > 30 \) nm and \( \eta > 1.5 \) which demonstrates the negative effects of high \( R \) and \( \eta \) parameters on conductivity of PCNT. Also, \( \phi_{p} \) and \( \phi_{f} \) parameters show positive and undesirable effects on conductivity of nanocomposites. Similarly, \( l < 10 \) \( \mu \)m and \( \sigma_{c} < 10^{5} \) S/m result in a weak conductivity signifying the direct influences of both \( \sigma_{c} \) and \( \phi_{f} \) parameters on PCNT conductivity. Additionally, \( f < 4 \) nm and \( d_{c} < 1.5 \times 10^{-3} \) nm result in poor \( \sigma = 0.3 \) S/m which show the positive impacts of these parameters on conductivity of nanocomposites. Moreover, the fraction of percolated CNT and number of contacts directly control the conductivity. So, the developed model logically shows better conductivity by the higher levels of CNT concentration, length, conduction and straightness, interphase thickness, diameter of contact area, percentages of percolated CNT and number of contacts beside the smaller ranges of CNT radius and percolation threshold.

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