Journal of Applied Polymer Science

Effects of the Dispersion State and Aspect Ratio of Carbon Nanotubes on Their Electrical Percolation Threshold in a Polymer

Ha-Da Bao, Yao Sun, Zhuo-Yue Xiong, Zhao-Xia Guo, Jian Yu

Key Laboratory of Advanced Materials (MOE), Department of Chemical Engineering, Tsinghua University, Beijing 100084, People's Republic of China

Correspondence to: Z.-X. Guo (E-mail: guozx@mail.tsinghua.edu.cn) or J. Yu (E-mail: yujian03@mail.tsinghua.edu.cn)

ABSTRACT: The electrical percolation threshold of carbon nanotubes (CNTs) is correlated with their dispersion state and aspect ratio through modeling. An analytical percolation model based on excluded volume theory and developed for systems containing two types of fillers is used. CNTs are modeled as two types of fillers: single CNT and m-CNT bundle, and a variable *P* representing the dispersion state of CNTs is introduced. An equation showing the effects of the dispersion state and aspect ratio on the electrical percolation threshold of CNTs is established and verified with some of the published experimental data. It is useful for predicting the conductive behavior of polymer/CNT composites and for the design of their processing conditions. © 2012 Wiley Periodicals, Inc. J. Appl. Polym. Sci. 000: 000–000, 2012

KEYWORDS: carbon nanotubes; polymer blends; nanocomposites; modeling

Received 27 December 2011; accepted 23 February 2012; published online

DOI: 10.1002/app.37554

INTRODUCTION

Polymer/carbon nanotube (CNT) conductive composites have been extensively investigated over the last decade.[1,2] A large amount of work has been directed to their preparation and characterization. The electrical percolation threshold of CNTs is usually much lower than that of the traditional carbon fillers (such as carbon black) no matter which type of processing method is used, because of their inherent electrical conductivity and high aspect ratio. It was found that the dispersion state and aspect ratio of CNTs have significant effects[3–6] on their electrical percolation threshold in a polymer, although other factors such as polarity and crystallization behavior of the polymer, interfacial tension between CNT and polymer, and processing viscosity may also have some influence. Therefore, correlating the electrical percolation threshold of CNTs with their dispersion state and aspect ratio through modeling is of great interest in view of predicting the conductive behavior of polymer/CNT composites and material design.

The models proposed so far for the electrical percolation of CNTs in a polymer include mainly the analytical models based on excluded volume theory,[7–16] numerical simulation,[13–22] and the analytical model based on the interparticle distance concept.[23] Li et al.[23] reported the interparticle distance approach, where CNTs were modeled as a mixture of individual CNTs and spherical agglomerates. Two dispersion parameters were introduced to reflect the dispersion state of CNTs: one (ξ) is the volume fraction of agglomerated CNTs and the other (ε) is the localized volume content of CNTs in agglomerate. The correlation of percolation threshold with the dispersion state and aspect ratio of CNTs was established. However, the dispersion parameter ε is difficult to estimate. The excluded volume approach is popular in the field of carbon-filler filled conductive polymers.[8,11,24–26] It is simple and computationally less demanding compared to numerical simulation. The basic principle is: the number of objects per unit volume at percolation is inversely proportional to the excluded volume of one of the objects. For an object having an infinite aspect ratio, the proportionality constant is 1. As originally proposed by Ounaies et al.,[16] single CNTs were often modeled as spherocylinders, and the CNT agglomerates modeled as closely packed bundles of CNT cylinders with identical length to a single CNT but with a larger diameter. Grujicic et al.[13] verified the prediction of the excluded volume approach with numerical simulation, and concluded that the analytical model based on the excluded volume theory can be used to predict the percolation threshold of

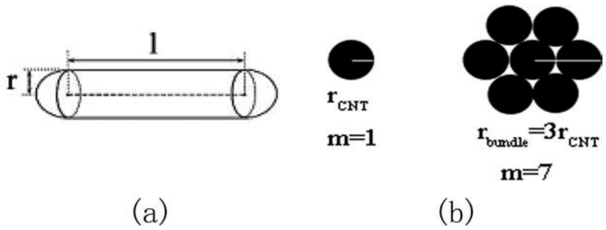

Figure 1. Illustration of (a) single CNT in length view and (b) cross-sections of single CNT and 7-CNT bundle.

cylinder-like fillers with an aspect ratio as low as 350. Berhan and Sastry[14] investigated the relationship between percolation threshold and excluded volume using Monte Carlo simulation, and found a correlation between the proportionality constant and the aspect ratio of CNTs, making the excluded volume approach suitable to aspect ratio down to 15. However, in all the above-mentioned work,[13,14,16] CNTs were modeled as a constant shape, either single or closely packed bundle of CNT cylinders, due to the limit of the excluded volume theory which was originally proposed for systems containing only one type of object. In fact, both single and agglomerated CNTs can be found in a real polymer/CNT composite.[3,5] The dispersion state of CNTs should be considered for the model to approach the real situation.

In our previous work, we extended the excluded volume theory to systems containing two types of fillers.[27] In this work, the electrical percolation of CNTs is modeled by considering CNTs as two types of fillers; single CNT and *m*-CNT bundle, so that a variable *P* representing the dispersion state of CNTs can be introduced. An equation showing the effects of dispersion state and aspect ratio on the percolation threshold of CNTs is developed and analyzed with the published experimental data.

DESCRIPTION OF THE PERCOLATION MODEL

The excluded volume theory[8,24] can be expressed as follows:[27]

$$N_c = k\frac{V_{unit}}{V_{\langle ex\rangle}} \tag{1}$$

where V_{unit} is the unit volume, $V_{\langle ex\rangle}$ is the excluded volume of the object, N_c is the number of objects in the V_{unit} at percolation, and k is the proportionality constant.

If there is only one type of conductive filler, then we can divide V_{unit} into N_c equivalent small volumes, and each small volume is $V_{\langle ex\rangle}/k$. When all of the small volumes are filled with the filler, percolation occurs, and we have $N_c \cdot V_{\langle ex\rangle}/k = V_{unit}$, which is equivalent to eq. (1). An analytical model for electrical percolation of mixed carbon fillers in a polymer was proposed by dividing V_{unit} into two types of small volumes.[27]

Now, we consider CNTs as two types of fillers: single CNT as capped cylinder; and CNT agglomerate as *m* closely packed CNT bundles.[16] As shown in Figure 1, the two types of fillers have the same length *l* but different radius; *r* for a single CNT and *nr* for a CNT bundle, for example, if $m = 7$, then $n = 3$.

Here, we define $V_{\langle CNT\rangle}$ and $V_{\langle bundle\rangle}$ as the excluded volumes of a single CNT and a CNT bundle, respectively, and $k_{\langle CNT\rangle}$ and $k_{\langle bundle\rangle}$ as the respective proportionality constants of a single CNT and a CNT bundle. According to the model of mixed fillers,[27] V_{unit} is divided into two types of small volumes, $V_{\langle CNT\rangle}/k_{CNT}$ and $V_{\langle bundle\rangle}/k_{bundle}$, as shown in Figure 2.

When all of the small volumes are filled, percolation occurs. Therefore, we have the following equation:

$$V_{unit} = N'_{single} \cdot \frac{V_{\langle CNT\rangle}}{k_{CNT}} + N'_{single} \cdot \frac{V_{\langle bundle\rangle}}{k_{bundle}} \tag{2}$$

where N'_{single} and N'_{bundle} are the numbers of single CNTs and CNT bundles, respectively.

RESULTS AND DISCUSSION

$k_{\langle CNT\rangle}$ and $k_{\langle bundle\rangle}$ Expressions

It is known that the proportionality constant $k = 1$, when the aspect ratio λ (i.e., $L/2r$) is infinite. For the case where λ is not infinite, Néda et al.[12] introduced a variable S to k, that is,

$$k = 1 + S \tag{3}$$

Berhan and Sastry[14] found that

$$S = C_1\left(\frac{r}{L}\right)^{C_2} \tag{4}$$

where $C_1 = 5.231$, and $C_2 = 0.569$.

Therefore,

$$k_{CNT} = 1 + S_1 \tag{5}$$

$$k_{bundle} = 1 + S_2 \tag{6}$$

where $S_1 = 5.231 \cdot (2\lambda)^{-0.569}$ and $S_2 = 5.231 \cdot n^{0.569} \cdot (2\lambda)^{-0.569}$

Correlations of the Percolation Threshold with the Dispersion State and Aspect Ratio of CNTs

Entering eqs. (5) and (6) into eq. (2):

$$V_{unit} = N'_{single} \cdot \frac{V_{\langle CNT\rangle}}{1 + S_1} + N'_{bundle} \cdot \frac{V_{\langle bundle\rangle}}{1 + S_2} \tag{7}$$

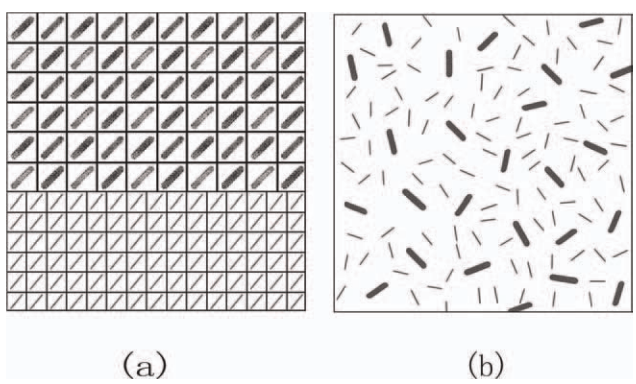

Figure 2. Schematic illustrations of the excluded volume for two-filler (single CNT and CNT bundle) systems in (a) an extreme state and (b) the real state.

We define P as the number fraction of single CNTs to total CNTs to represent the dispersion state of CNTs. Thus:

$$P = \frac{N'_{\text{single}}}{N'_{\text{single}} + mN'_{\text{bundle}}} \tag{8}$$

$0 \le P \le 1$. When $P = 0$, all of the CNTs are in an agglomerated state. The dispersion of CNTs is better when the P value increases. When $P = 1$, all of the CNTs are in a monodisperse state.

Entering eqs. (7) and (8), we get:

$$N'_{\text{single}} = \frac{m \cdot P \cdot (1+S_1) \cdot (1+S_2) \cdot V_{\text{unit}}}{m \cdot P \cdot (1+S_2) \cdot V_{\langle \text{CNT} \rangle} + (1-P) \cdot (1+S_1) V_{\langle \text{bundle} \rangle}} \tag{9}$$

$$N'_{\text{bundle}} = \frac{(1-P) \cdot (1+S_1) \cdot (1+S_2) \cdot V_{\text{unit}}}{m \cdot P \cdot (1+S_2) \cdot V_{\langle \text{CNT} \rangle} + (1-P) \cdot (1+S_1) V_{\langle \text{bundle} \rangle}} \tag{10}$$

Thus, the percolation threshold φ_c can be expressed as follows:

$$\begin{aligned}\varphi_c = \frac{V_{\text{tot}}}{V_{\text{unit}}} &= \frac{N'_{\text{single}} V_{\text{CNT}} + N'_{\text{bundle}} \cdot V_{\text{bundle}}}{V_{\text{unit}}} \\ &= \frac{m \cdot P \cdot (1+S_1) \cdot (1+S_2) \cdot V_{\text{CNT}} + (1-P) \cdot (1+S_1) \cdot (1+S_2) \cdot V_{\text{bundle}}}{m \cdot P \cdot (1+S_2) \cdot V_{\langle \text{CNT} \rangle} + (1-P) \cdot (1+S_1) \cdot V_{\langle \text{bundle} \rangle}}\end{aligned} \tag{11}$$

$V_{\langle \text{CNT} \rangle}$, $V_{\langle \text{bundle} \rangle}$, V_{CNT}, and V_{bundle} are known as follows:

$$V_{\langle \text{CNT} \rangle} = \frac{32}{3}\pi r_{\text{CNT}}^3 + 8\pi l_{\text{CNT}} r_{\text{CNT}}^2 + 4 l_{\text{CNT}}^2 r_{\text{CNT}} \cdot \langle \sin(\gamma) \rangle \tag{12}$$

$$V_{\langle \text{bundle} \rangle} = \frac{32}{3}\pi r_{\text{bundle}}^3 + 8\pi l_{\text{CNT}} r_{\text{bundle}}^2 + 4 l_{\text{CNT}}^2 r_{\text{bundle}} \cdot \langle \sin(\gamma) \rangle \tag{13}$$

$$V_{\text{CNT}} = \frac{4}{3}\pi r_{\text{CNT}}^3 + \pi l_{\text{CNT}} r_{\text{CNT}}^2 \tag{14}$$

$$V_{\text{bundle}} = \frac{4}{3}\pi r_{\text{bundle}}^3 + \pi l_{\text{CNT}} r_{\text{bundle}}^2 \tag{15}$$

where $\langle \sin(\gamma) \rangle$ is the average value of $\sin(\gamma)$ for two sticks and γ is the angle between them.

Entering eqs. (12)–(15) to eq. (11):

$$\varphi_C = \frac{m \cdot P \cdot (1+S_1) \cdot (1+S_2) \cdot \left(\frac{4}{3}\pi r_{\text{CNT}}^3 + \pi l_{\text{CNT}} r_{\text{CNT}}^2\right) + (1-P) \cdot (1+S_1) \cdot (1+S_2) \cdot \left(\frac{4}{3}\pi r_{\text{bundle}}^3 + \pi l_{\text{CNT}} r_{\text{bundle}}^2\right)}{m \cdot P \cdot (1+S_2) \cdot \left(\frac{32}{3}\pi r_{\text{CNT}}^3 + 8\pi l_{\text{CNT}} r_{\text{CNT}}^2 + 4 l_{\text{CNT}}^2 r_{\text{CNT}} \cdot \langle \sin(\gamma) \rangle\right) + (1-P) \cdot (1+S_1) \cdot \left(\frac{32}{3}\pi r_{\text{bundle}}^3 + 8\pi l_{\text{CNT}} r_{\text{bundle}}^2 + 4 l_{\text{CNT}}^2 r_{\text{bundle}} \cdot \langle \sin(\gamma) \rangle\right)} \tag{16}$$

$r_{\text{bundle}} = n \cdot r_{\text{CNT}}$, and for a random distribution $\langle \sin(\gamma) \rangle = \pi/4$.[14] Thus:

$$\varphi_c(P, \lambda) = \frac{m \cdot P \cdot (1+S_1) \cdot (1+S_2) \cdot (3\lambda + 2) + (1-P) \cdot (1+S_1) \cdot (1+S_2) \cdot (3n^2\lambda + 2n^3)}{2m \cdot P \cdot (1+S_2) \cdot (3\lambda^2 + 12\lambda + 8) + 2(1-P) \cdot (1+S_1) \cdot (3n\lambda^2 + 12n^2\lambda + 8n^3)} \tag{17}$$

We assume that a CNT bundle is seven closely packed CNTs as shown in Figure 1, that is, $m = 7$, $n = 3$, $S_1 = 5.231 \cdot (2\lambda)^{-0.569}$, $S_2 = 5.231 \cdot 3^{0.569} \cdot (2\lambda)^{-0.569} = 9.77 \cdot (2\lambda)^{-0.569}$.

Based on eq. (17), Figure 3 can be drawn, showing the effects of the dispersion state P and aspect ratio λ of CNTs on the percolation threshold, where P and λ can be varied simultaneously.

As shown in Figure 3, for a fixed value of P the percolation threshold changes about three orders of magnitude when λ changes from 10^1 to 10^4. For example, if $P = 1$, the percolation threshold decreases from 7.29 to 0.005 vol %; if $P = 0$, the percolation threshold decreases from 20.5 to 0.016 vol %. In addition, for a fixed value of λ, the percolation threshold may change significantly when P changes from 0 to 1. For example, if $\lambda = 10^2$, the percolation threshold decreases from 2.02 to 0.61 vol %. This finding indicates that both P and λ are important factors affecting the percolation threshold of CNTs and one need to make CNTs retain both a high degree of dispersion and a high aspect ratio if low percolation threshold is intended to be obtained. This is in agreement with the finding of Li et al. derived from the interparticle distance model.[23]

Examination of Eq. (17) with Published Experimental Data

Ounaies et al.[16] used CNTs having an aspect ratio (λ) of 2143 in the preparation of polyimide/CNT composites by solution compounding, and found that the electrical percolation threshold was 0.05 vol %. The prediction from their analytical model based on the excluded volume theory is 0.024 vol % if all of the CNTs are in the monodisperse state, and 0.076 vol % if all of the CNTs are in the agglomerated state as 7-CNT bundles. None of the predicted values are in agreement with the experiment result. According to the model presented in this work, $P = 0.29$; that is, 29% of the total number of CNTs are in the

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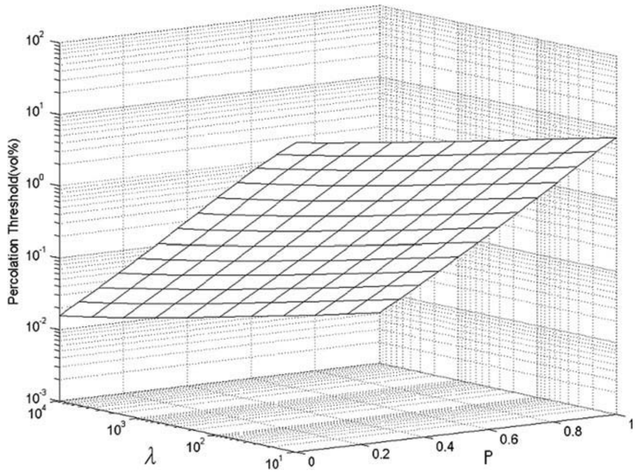

Figure 3. Effects of the degree of dispersion *P* and aspect ratio *λ* on the percolation threshold of CNTs in a polymer.

monodisperse state, and the rest of the CNTs are in the agglomerated state.

Our research group[3] prepared polyoxymethylene (POM)/CNT, polyamide 6 (PA6)/CNT, and polypropylene (PP)/CNT composites by melt mixing, and found that the electrical percolation thresholds were 0.70, 1.09, and 0.90 vol %, respectively. Since it is known that CNTs can be shortened during melt compounding,[28] CNTs were isolated from the above-mentioned three composites by dissolving the polymers in hexafluoroisopropanol, formic acid, and refluxing xylene, respectively, and observed under TEM (Figure 4). The lengths and the diameters of CNTs are measured using AutoCAD analyzing software. The average aspect ratios of CNTs after compounding were 106, 95, and 91. Taking the aspect ratio (λ) as 10^2 for calculation using eq. (17), the values of *P* representing the dispersion state of CNTs are 0.85, 0.43, and 0.60 for the cases of POM/CNT, PA6/CNT, and PP/CNT composites, respectively, suggesting that 85, 43, and 60% of total numbers of CNTs were in the monodisperse state. In this way, the experimental results are reasonably explained. It should be noted that Figure 4 shows the CNTs isolated from the polymer/CNT composites, therefore can only be used to measure the aspect ratios of CNTs, but not for estimation of the dispersion states of CNTs in the composites.

The *P* values calculated from eq. (17) for the above-mentioned examples along with three other examples are illustrated in Figure 5. It is clear that the experimental results can be reasonably explained by the variable *P* across a wide range of CNT aspect ratios. The references, matrix types and processing conditions for all the samples shown in Figure 5 are listed in Table I

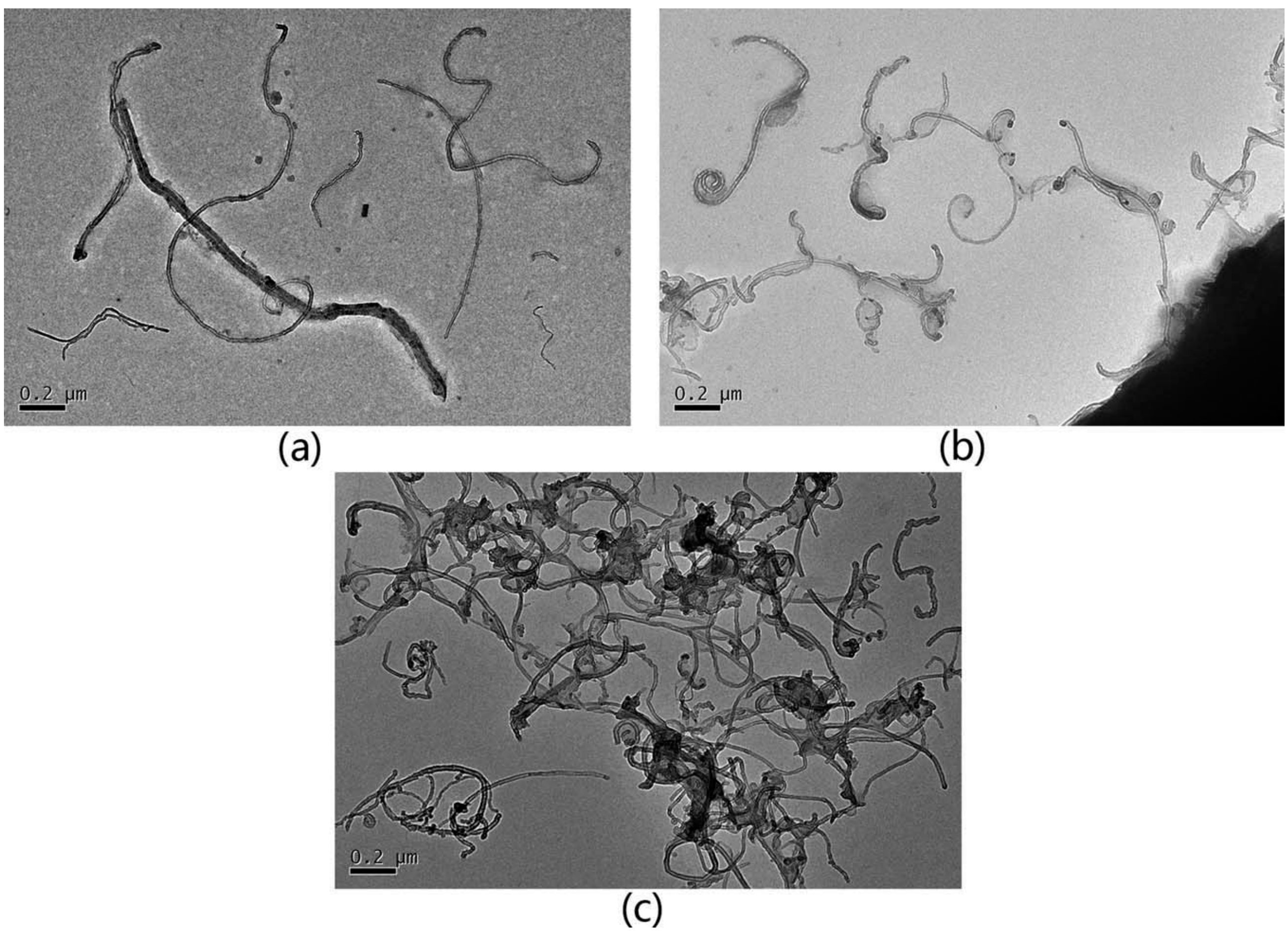

Figure 4. TEM photographs of the CNTs isolated from (a) POM/CNT, (b) PA6/CNT, and (c) PP/CNT composites.

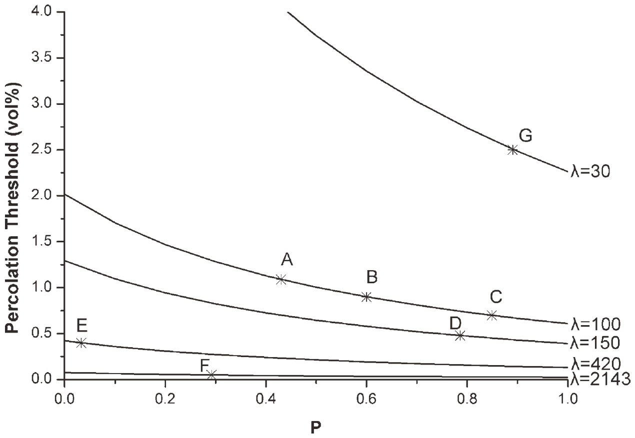

Figure 5. Effect of the degree of dispersion (*P*) on the percolation threshold of CNTs at different aspect ratios (λ). A, B, C, D, E, F, and G show the calculated *P* values for the data published in Refs. 3(A–C), 4(E), 5(D), 16(F), and 29(G).

along with the aspect ratios (λ) and dispersion variables (*P*). Only limited examples are listed, because analyzing all of the published results is impossible due to the unclear description of CNTs and the possible rupture of CNTs during compounding. In Ref. 29, three types of CNTs were used, only the shortest (also the lowest aspect ratio) is shown in Table I. According to our model, the other two types of CNTs must be severely fractured during melt compounding, for example, the real aspect ratio could be down to 50 for CNT3 (about a quarter of the original aspect ratio i.e., 200). Besides, CNT1 is heavily agglomerated as shown by the TEM photographs,[29] a much bigger m value should be used for the model to understand the relatively high percolation threshold.

As indicated by the shape of the curves shown in Figure 5, at a low aspect ratio the dispersion state of CNTs has a significant effect on their percolation threshold. The effect decreases when the aspect ratio increases. At a very high aspect ratio, the effect is little.

Limitations

The model presented in this work is not suitable for cases where CNTs are not isotropically distributed in the polymer, because filler orientation is not considered in the model. It cannot explain the ultra low percolation threshold (<0.005 vol %) obtained with a CNT aspect ratio of $<10^4$. The possible reasons could be:

1. The model only takes two factors (P and λ) into account. In some cases, one or more other factors may not be ignored.
2. CNTs are modeled as straight cylinders and CNT agglomerates modeled as closely packed CNT bundles. In fact, CNTs are wavy and CNT agglomerates are irregularly shaped and composed of randomly arranged CNTs. In this model, CNT dispersion is only represented by a bimodal distribution of diameters for simplicity; while in reality, CNTs have a wide range of dispersion.
3. The model is based on the excluded volume theory and thus gives statistical percolation threshold. In some cases, kinetic percolation, which takes the particle movement and re-aggregation into account, should be considered.

CONCLUSIONS

An electrical percolation model of CNTs is proposed based on excluded volume theory by considering CNTs as two types of fillers: single CNT and 7-CNT bundle. An equation showing the effects of the dispersion state and aspect ratio of CNTs on their electrical percolation threshold in a polymer is derived from the model. The equation predicts that both the dispersion state and aspect ratio are important factors affecting the percolation threshold of CNTs. The percolation threshold changes about three orders of magnitude when the aspect ratio of CNTs changes from 10^1 to 10^4. At a low aspect ratio, the dispersion state of CNTs has a significant effect on their percolation threshold. The effect decreases when the aspect ratio increases. At a very high aspect ratio, the effect is little. The equation is verified with some of the published experimental data, showing the applicability of the model. Therefore, it can provide a general guidance for the design of the processing conditions of polymer/CNT composites.

ACKNOWLEDGMENTS

The authors thank Beijing Key Laboratory of Green Reaction Engineering and Technology, Department of Chemical Engineering, Tsinghua University for kindly providing MWCNTs. This work was supported by National Natural Science Foundation of China (No. 50973053) and the Specialized Research Fund for the Doctoral Program of Higher Education (No. 20090002110072).

Table I. The References, Matrix Types, and Processing Conditions for all the Samples

Matrix type	Aspect ratio	Processing method	Percolation threshold (vol %)	Dispersion state (*P*)	Reference	Symbol
Polyimide/CNT	2143[a]	Solution compounding	0.05	0.29	16	F
POM/CNT	100[b]	Melt mixing	0.70	0.85	3	C
PA6/CNT	100[b]	Melt mixing	1.09	0.43	3	A
PP/CNT	100[b]	Melt mixing	0.90	0.60	3	B
Epoxy resin/CNT	420[a]	Solution compounding	0.40	0.03	4	E
Polycarbonate/CNT	150[a]	Melt mixing	0.48	0.79	5	D
PP/CNT	30[a]	Melt mixing	2.5	0.89	29	G

[a]Original data., [b]Measured data after compounding.

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