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Analysis of electrical conductivity of carbon nanotube-reinforced two-phase composites

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ABSTRACT

This short paper presents a simple approach for calculating the effective electrical conductivity of carbon nanotubes-reinforced two-phase composite materials. The approach is developed by modifying the traditional effective medium approximation to include the effect of percolation threshold. The predicted electrical conductivity from the present approach is validated by using published experimental results. It is shown that the effective electrical conductivity of two-phase composites can be calculated by using the lower bound of the effective medium approximation if the inclusion volume fraction is below its percolation threshold, and the projection of the upper bound of the effective medium approximation if the inclusion volume fraction exceeds its percolation threshold.

1. Introduction

Carbon nanotubes (CNTs) have excellent thermal and electrical conductivities. Because of their nanoscale cross-section, electrons propagate only along the longitudinal axis of the CNTs. As a result of this, CNTs are generally referred to as one-dimensional conductors. Owing to the large aspect ratio of CNTs, the incorporation of small amount of CNTs in polymers can remarkably improve the thermal and electrical conductivities of the CNTs-filled polymer composites [1–4]. In recent years, great efforts have been made in developing smart sensors by using CNTs-reinforced composites for various industrial applications [5–8].

To understand how the CNTs influence the electrical conductivity of CNTs-reinforced composites, experimental investigations have been carried out on the electrical conductivity mechanism of CNTs-reinforced composites. For example, Kim et al. [9] investigated the electrical conductivity of chemically modified multi-walled CNTs reinforced epoxy composites. Taherian [10] presented an experimental study on the electrical conductivity of polymer-based nanocomposites. Jang et al. [11] reported the electrical conductivity of CNTs-reinforced cement--based composites with and without moisture. Jang and Li [12] examined the influence of thermal and mechanical loadings on the electrical conductivity of CNTs-reinforced composites. Apart from the experimental work, numerical simulations have been also carried out on the electrical conductivity of CNTs-reinforced multi-phase composites by using statistical continuum theory [13–16], micromechanics approaches [17–21], and multiscale modelling techniques [22–24]. The numerical simulations are able to handle complicated situations and take account into various individual influences such as the aspect ratio, orientation, shape and distribution of CNTs dispersed in the matrix on the local and global conductivity of the composites. Moreover, the simulation can also be used to predict the effective electrical conductivity of the composite.

An important issue for promoting the use of CNTs-reinforced composites in engineering fields is the development of theoretical models able to provide a quantitative prediction of their overall electrical conductivity. Recently, Zare and Rhee [25] presented a simple model for the prediction of the electrical conductivity of CNTs-polymer nanocomposites by assuming the filler properties, interphase dimension, network level, interfacial tension, and tunnelling distance. Kim et al. [26] proposed a prediction model for the electrical conductivity and percolation threshold of nanocomposites containing spherical particles by using three-dimensional random representative volume elements. Mazaheri et al. [27] proposed a theoretical model for describing the effective electrical conductivity and percolation behaviour of polymer-graphene nanocomposites with various exfoliated filleted nanoplatelets. Fang et al. [28] developed a theoretical calculation model for the effective electrical conductivity of CNTs-reinforced two-phase composites by using a penalty function. The model considers the aspect

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ratio effect of CNTs on the effective electrical conductivity of the composite materials. A review paper was provided by Radzuan et al. [29] on the electrical conductivity models for conductive polymer composites.

The literature survey on existing analytical models shows that, although there are many works existed in literature, many of them are too comprehensive and/or too complicated to be applied. Some of them were developed empirically based on some special conditions and thus are difficult to be applied widely. Note that CNTs are the tube-like fibres with very large aspect ratios. This makes the CNTs-reinforced composite have very low percolation threshold [30,31]. Therefore, it is vital in the prediction models to include the effect of the percolation threshold on the electrical conductivity of the CNTs-reinforced composites. In this paper a simple analytical model is presented to calculate the effective electrical conductivity of CNTs-reinforced two-phase composite materials. The model is developed by modifying the upper and lower bounds of the effective medium approximation by including the effect of percolation threshold. The model is validated by using the experimental results published in literature.

2. Effective electrical conductivity of two-phase composite materials

Consider a two-phase composite material, one of which is the continuous polymer matrix and the other of which is the randomly dispersed small inclusions (CNTs) distributed in the matrix. The properties of the composite material are a function of the properties of the two constituent materials, their volume fractions, and the geometry of the dispersed phase. The latter includes the shape and size of the inclusions, as well as their distribution and orientation in the matrix.

In literature five simple approaches have been widely used to estimate the effective electrical conductivity of the two-phase composite material [28,32,33], namely, parallel, series, spherical (upper and lower), and cubic (upper and lower) approximations (see Fig. 1). The corresponding mathematical formulations used for the calculation of the effective electrical conductivity of the two-phase composite material in these models can be expressed as follows,

$$\sigma_e = \sigma_i V_i + \sigma_m V_m \tag{1}$$

$$\sigma_e = \frac{(\sigma_m + 2\sigma_i) + 2V_m(\sigma_m - \sigma_i)}{(\sigma_m + 2\sigma_i) - V_m(\sigma_m - \sigma_i)}\sigma_i \tag{2}$$

$$\sigma_e = \frac{(\sigma_i + 2\sigma_m) + 2V_i(\sigma_i - \sigma_m)}{(\sigma_i + 2\sigma_m) - V_i(\sigma_i - \sigma_m)}\sigma_m \tag{3}$$

$$\sigma_e = \sigma_i\left(1 - V_m^{2/3}\right) + \frac{V_m^{4/3}}{\frac{V_m}{\sigma_m} + \frac{V_m^{2/3} - V_m}{\sigma_i}} \tag{4}$$

$$\sigma_e = \sigma_m\left(1 - V_i^{2/3}\right) + \frac{V_i^{4/3}}{\frac{V_i}{\sigma_i} + \frac{V_i^{2/3} - V_i}{\sigma_m}} \tag{5}$$

$$\sigma_e = \frac{1}{\frac{V_i}{\sigma_i} + \frac{V_m}{\sigma_m}} \tag{6}$$

where σ_e is the effective electrical conductivity of the composite material, V_i and V_m are the volume fractions of the inclusions and matrix ($V_i + V_m \equiv 1$), σ_i and σ_m are the electrical conductivities of the inclusions and matrix, respectively. Eqs. (1)–(6) represent the parallel, spherical (upper and lower bounds), cubic (upper and lower bounds), and series approximations, respectively. Eqs. (2) and (4) are for the case where the matrix is perfectly enclosed by the inclusions (Fig. 1b and c) and thus they provide the upper bound of the prediction; whereas Eqs. (3) and (5) are for the case where the inclusions are perfectly enclosed by the matrix (Fig. 1e and f) and thus they provide the lower bound of the prediction. Mathematically, Eqs. (3) and (5) can be obtained directly from Eqs. (2) and (4) by swapping σ_i and V_i with σ_m and V_m, respectively.

The difference of the predicted electrical conductivities between the above six approximations is graphically shown in Fig. 2, where the conductivity of inclusions is assumed to be 50 times that of the matrix. It can be seen from the figure that the parallel and series models provide the highest and lowest values, whereas the spherical and cubic models provide similar predictions for the upper or lower bounds. The difference between the upper bound curves (parallel, spherical upper bound and cubic upper bound) or between the lower bound curves (series, spherical lower bound and cubic lower bound) reflects the influence of the configuration or pattern of the inclusions distributed in the matrix on the effective electrical conductivity of the composite material.

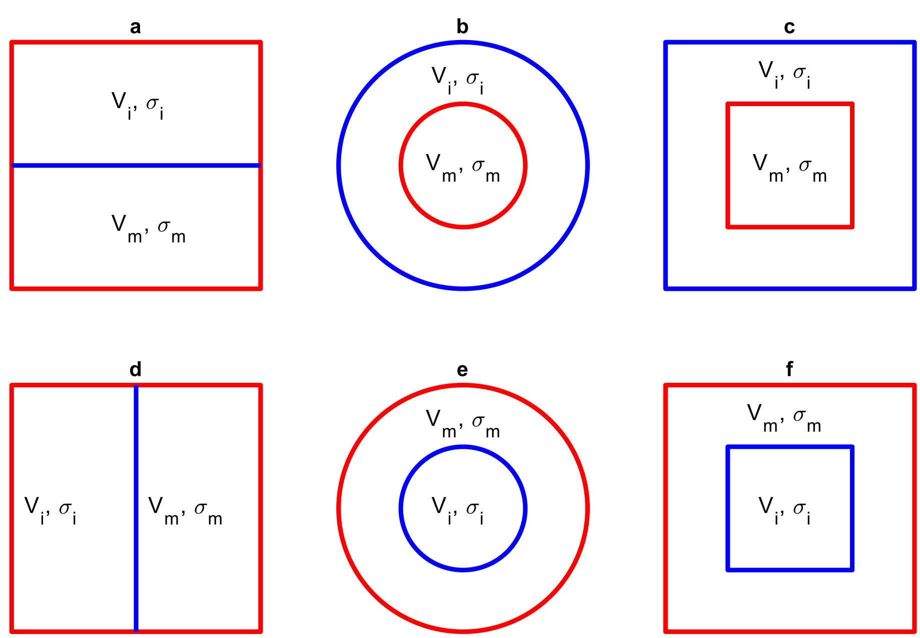

Fig. 1. (a) Parallel, (b) spherical (upper bound), (c) cubic (upper bound), (d) series, (e) spherical (lower bound), and (f) cubic (lower bound) models for estimating effective properties of two-phase composite materials.

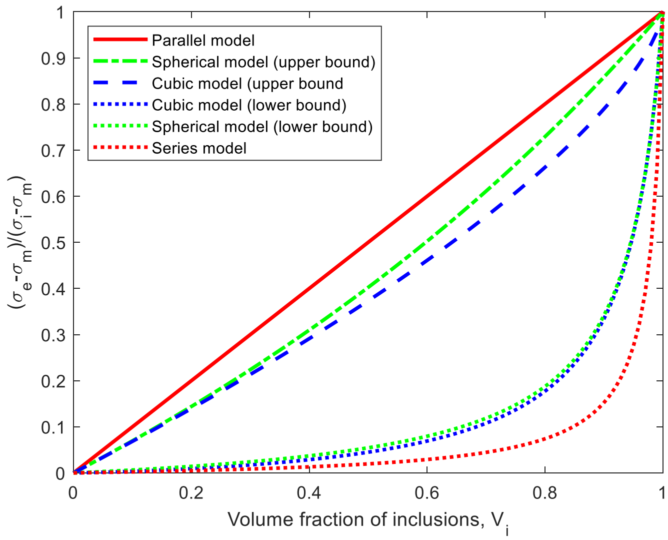

Fig. 2. Variation of effective electrical conductivity of two-phase composite material with volume fraction of inclusions (σ_i =50σ_m).

Physically, all the three upper bound curves (parallel, spherical and cubic upper bound models) represent the composite where the inclusions are perfectly percolated throughout the matrix; whereas all the three lower bound curves (series, spherical and cubic lower bound models) represent the composite where the inclusions are perfectly isolated within the matrix. Since the series and parallel models represent the two extreme cases in the upper and lower bound predictions, to use the spherical or cubic model would be more appropriate, particularly for the matrix composites [28,32,33]. Also, it is anticipated that, for a matrix composite with a small volume fraction of inclusions, individual inclusions would be more likely to be isolated and thus its effective electrical conductivity would be close to its lower bound curve. In opposite, for a matrix composite with a large volume fraction of inclusions, individual inclusions would be more likely to be connected and thus its effective electrical conductivity would be close to its upper bound curve. This implies that, for a real matrix composite its effective electrical conductivity will increase with increased volume fraction of inclusions, initially from the lower bound curve, then to the upper bound curve. The departure point from the lower bound curve to the upper bound curve is known as the percolation threshold of the composite. However, it should be mentioned here that, after the percolation threshold, part of the inclusions become connected, but some of the inclusions would remain isolated. Thus, the effective electrical conductivity of the composite material would be still below its upper bound value although it is much higher than its lower bound value. The exact value of the effective electrical conductivity at a given volume fraction of inclusions is dependent on the proportion of the volume fractions between the connected and isolated inclusions [30].

The percolation threshold of a two-phase composite material is dependent on the aspect ratio of inclusions and their distribution and dispersion in the matrix. According to the statistical continuum theory, for a composite with randomly distributed inclusions the percolation threshold of the composite can be determined based on the aspect ratio of the inclusions [27,30,31]. The higher the aspect ratio of the inclusions, the lower the percolation threshold of the composite. Similarly, for a given volume fraction of inclusions, the proportion between the connected and isolated inclusions would also be dependent on the aspect ratio of the inclusions, which can be effectively expressed in terms of the percolation threshold.

We herein first consider an extreme case, in which any increased volume fraction of inclusions from the percolation threshold point, V_i-V_p, is assumed to be connective, whereas the volume fraction of the inclusions V_p corresponding to the percolation threshold is assumed to remain isolated. In this case the effective electrical conductivity of the composite is given by the curve OB of the lower bound curve for $V_i < V_p$, and the curve BC for $V_i > V_p$, which is the modified upper bound curve by shifting the origin point of the upper bound curve to point B, as shown in Fig. 3, where the lower and upper bound curves are plotted based on the spherical model. It is obvious that the curve BC will be over-predicted when V_i is close to V_p and under-predicted when V_i is close to 1. The former is because not all of the increased inclusions would be connective when V_i is close to V_p; whereas the latter is because the originally isolated inclusions will turn to be connective when V_i is close to 1. In other words, the ratio between the connected and isolated inclusions should be proportional to the value of V_i-V_p. To take this into account, one way is to horizontally project the upper bound curve AD in Fig. 3 to BD. Mathematically, this can be expressed as follows.

For $V_i < V_p$:

$$\sigma_e = \frac{(\sigma_i + 2\sigma_m) + 2V_i(\sigma_i - \sigma_m)}{(\sigma_i + 2\sigma_m) - V_i(\sigma_i - \sigma_m)}\sigma_m \approx \frac{(1+2V_i)\sigma_m}{1-V_i} \tag{7}$$

For $V_i \geq V_p$:

$$\sigma_e = \frac{(\sigma_m + 2\sigma_i) + 2\left(1 - V_i^*\right)(\sigma_m - \sigma_i)}{(\sigma_m + 2\sigma_i) - \left(1 - V_i^*\right)(\sigma_m - \sigma_i)}\sigma_i \approx \frac{2V_i^*\sigma_i}{3 - V_i^*} \tag{8}$$

$$V_i^* = V_A + \left(\frac{V_i - V_p}{1 - V_p}\right)^\alpha (1 - V_A) \tag{9}$$

where V_i* is the projected volume fraction of inclusions, α is a constant to be determined (α=1 and α>1 represent the linear and nonlinear projections, respectively), and V_A is the volume fraction of inclusions at point A (see Fig. 3), which can be determined from the upper bound curve according to the condition $\sigma_e=\sigma_A=\sigma_B$ as follows,

$$V_A = 1 - \frac{\left(1 - \frac{\sigma_A}{\sigma_i}\right)(2\sigma_i + \sigma_m)}{\left(2 + \frac{\sigma_A}{\sigma_i}\right)(\sigma_i - \sigma_m)} \tag{10}$$

where σ_A and σ_B is the effective electrical conductivity at point A and B as shown in Fig. 3, which can be determined from the lower bound curve at the point $V_i=V_B=V_p$ as follows,

$$\sigma_A = \sigma_B = \frac{(\sigma_i + 2\sigma_m) + 2V_p(\sigma_i - \sigma_m)}{(\sigma_i + 2\sigma_m) - V_p(\sigma_i - \sigma_m)}\sigma_m \tag{11}$$

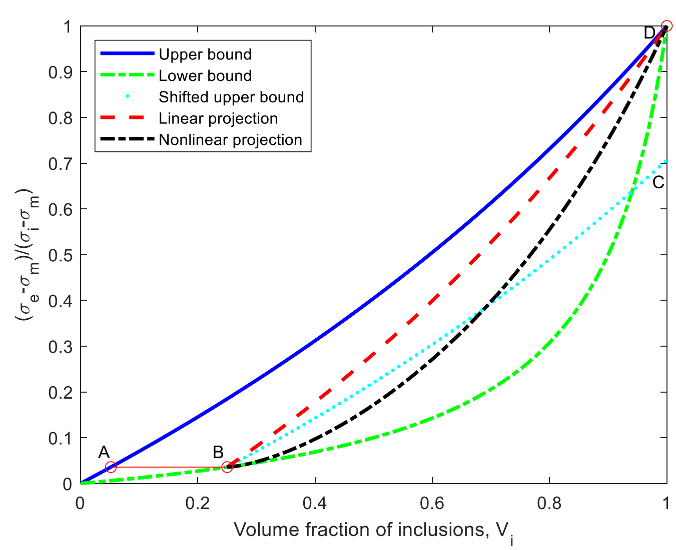

Fig. 3. Variation of effective electrical conductivity of two-phase composite material with volume fraction of inclusions after percolation threshold (σ_i =25σ_m).

Substituting Eq. (11) into (10), it yields,

$$V_A = 1 - \frac{(2\sigma_i + \sigma_m)\left[1 - \frac{(\sigma_i+2\sigma_m)+2V_p(\sigma_i-\sigma_m)}{(\sigma_i+2\sigma_m)-V_p(\sigma_i-\sigma_m)} \times \frac{\sigma_m}{\sigma_i}\right]}{(\sigma_i - \sigma_m)\left[2 + \frac{(\sigma_i+2\sigma_m)+2V_p(\sigma_i-\sigma_m)}{(\sigma_i+2\sigma_m)-V_p(\sigma_i-\sigma_m)} \times \frac{\sigma_m}{\sigma_i}\right]} \tag{12}$$

Substituting Eq. (12) into (9), it yields,

$$V_i^* = 1 + \left[\left(\frac{V_i - V_p}{1 - V_p}\right)^{\alpha} - 1\right] \frac{(2\sigma_i + \sigma_m)\left[1 - \frac{(\sigma_i+2\sigma_m)+2V_p(\sigma_i-\sigma_m)}{(\sigma_i+2\sigma_m)-V_p(\sigma_i-\sigma_m)} \times \frac{\sigma_m}{\sigma_i}\right]}{(\sigma_i - \sigma_m)\left[2 + \frac{(\sigma_i+2\sigma_m)+2V_p(\sigma_i-\sigma_m)}{(\sigma_i+2\sigma_m)-V_p(\sigma_i-\sigma_m)} \times \frac{\sigma_m}{\sigma_i}\right]} \tag{13}$$

The approximation equation of Eqs. (7) and (8) is applied for the case of $\sigma_i \gg \sigma_m$; whereas the exact equation of Eqs. (7) and (8) does not have this limitation and thus can be applied to any two-phase composites. Fig. 3 also shows the two projected curves BD (red colour and black colour) of the upper bound curve AD calculated by using Eqs. (8) and (13), in which the red curve is the linear projection with α=1 and the black curve is the nonlinear projection with α=1.5. The comparison of the two projected curves BD with the curve BC for the extreme case indicates that the use of the nonlinear projection would be more accurate and thus should be employed.

3. Model validation

To validate the present prediction model, the comparison between the calculated effective electrical conductivity using Eqs. (7), (8) and (13) and that measured in experiments is made for four different two-phase composite materials. The values used in the prediction for the percolation threshold and electrical conductivities of the matrix and inclusions are given in Table 1, which are directly taken from corresponding experiments. The measured conductivity was evaluated by means of a dielectric analyzer in the specified frequency range. During the experiments the specimens used for the conductivity measurement were silver-pasted to minimize the contact resistance between the composites and the electrodes. Fig. 4 shows the comparison between the predicted and measured effective electrical conductivities of the ethylene-octene-copolymer composites containing multi-walled CNTs and carbon fibers as the inclusions, respectively. The details of the experimental work can be found in Ref. [34]. Fig. 5 shows the comparison between the predicted and measured effective electrical conductivities of the multi-walled CNTs-reinforced epoxy composites. The experimental data were taken from Refs. [9,12]. It is evident from these comparisons that there is excellent agreement between the predicted and measured effective electrical conductivities. This demonstrates that the present analytical model is able to reflect the influence of CNTs on the electrical conductivity of the CNTs-reinforced composite materials and the predicted electrical conductivity is reliable and accurate.

4. Conclusion

This paper has presented an analytical model for predicting the effective electrical conductivity of CNTs-reinforced two-phase composite materials. The model has been validated using published experimental results. From the present study, the following conclusions can be drawn.

- The effective electrical conductivity of the CNTs-reinforced two-phase composite materials can be calculated in terms of the electrical conductivities of the two constituent materials, the percolation threshold of the composite, and the volume fraction of the inclusions.
- For the volume fraction of inclusions below the percolation threshold of the composite, all the inclusions can be treated to be isolated, and thus the effective electrical conductivity of the CNTs-reinforced two-phase composite material can be calculated by using the lower bound of the effective medium approximation as demonstrated in the present examples.
- For the volume fraction of inclusions above the percolation threshold of the composite, part of the inclusions is connected and part of them remains isolated, and thus the effective electrical conductivity of the

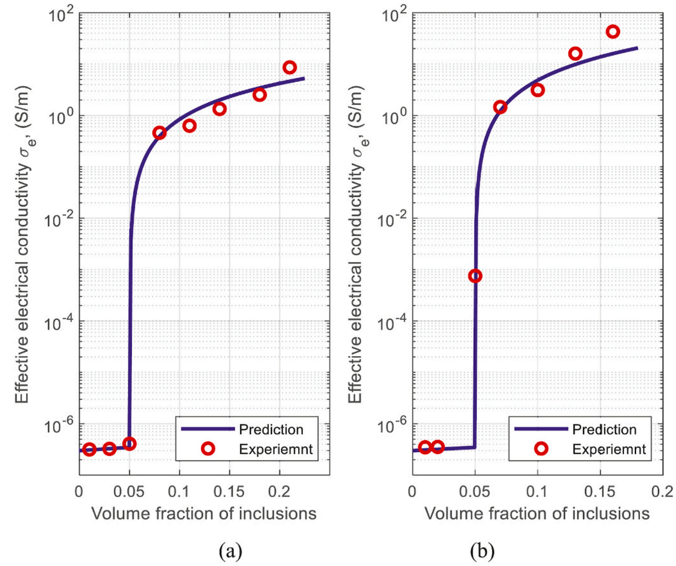

Fig. 4. Comparison of predicted and measured effective electrical conductivities of ethylene-octene-copolymer composites with (a) multi-walled CNTs and (b) carbon fibres as inclusions.

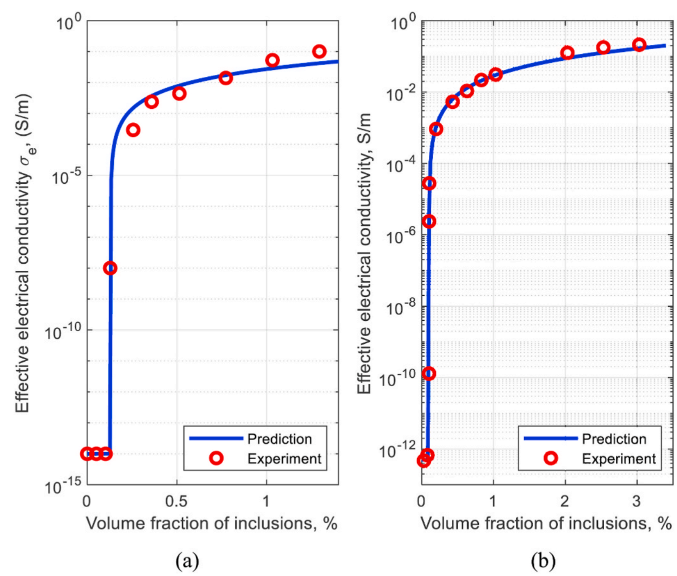

Fig. 5. Comparison of predicted and measured effective electrical conductivities of epoxy composites with (a) multi-walled CNTs and (b) A4 multi-walled CNTs as inclusions.

Table 1
Parametric values employed in calculations (α=1.5).

Case	Percolation threshold (V_p, %)	Conductivity of inclusions (σ_i, s/m)	Conductivity of matrix (σ_m, s/m)
Fig. 4(a) [34]	5.0	100	3.0 x 10^{-7}
Fig. 4(b) [34]	5.0	600	3.0 x 10^{-7}
Fig. 5(a) [12]	0.13	50	1.0 x 10^{-14}
Fig. 5(b) [9]	0.10	50	5.0 x 10^{-13}

CNTs-reinforced two-phase composite material can be calculated by using the nonlinear projection of the upper bound curve of the effective medium approximation as demonstrated in the present examples.

- The effects of aspect ratio and distribution pattern of inclusions on the effective electrical conductivity of the composite can be characterized by the projection of the effective medium approximation using the percolation threshold of the composite.

CRediT authorship contribution statement

Zaiwei Li: Conceptualization, Methodology, Investigation, Formal analysis, Writing – original draft, Validation, Writing – review & editing. **Long-yuan Li:** Conceptualization, Methodology, Investigation, Formal analysis, Writing – original draft, Validation, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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