Computational Materials Science 89 (2014) 80–88

Contents lists available at ScienceDirect

Computational Materials Science

journal homepage: www.elsevier.com/locate/commatsci

Estimating electrical conductivity of multi-scale composites with conductive nanoparticles using bidirectional time marching percolation network mapping

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ARTICLE INFO

Article history:
Received 27 October 2013
Received in revised form 24 February 2014
Accepted 18 March 2014
Available online 13 April 2014

Keywords:
Carbon nanotubes
Multi-scale composite
Electrical conductivity

ABSTRACT

A computationally efficient approach is developed to determine electrical conductivity of large specimens of multi-scale composites composed of electrically conductive nanoparticles and nonconductive matrix and micro-scale reinforcements. As a test case, the paper studies nanocomposites and multi-scale fiber/polymer composites of carbon nanotubes. Fraction of percolating carbon nanotubes is defined as a metric for estimating electrical conductivity of composites with intermediate volume fraction of carbon nanotubes. The results indicate that for multi-scale composites with high fiber/reinforcement volume fraction, the simulation size needs to be in millimeters whereas for multi-scale composites with low fiber/reinforcement volume fraction, smaller simulation sizes (in 100s of microns) are sufficient. The present research is a first step towards efficient design of fiber composites for electromagnetic applications such as lightening protection and electromagnetic shielding.

1. Introduction

Multi-scale fiber composites are defined as composite materials having reinforcements of several length scales. Multi-scale polymer composites of carbon nanotubes are emerging as a very popular choice for aerospace, marine and wind energy industries due to their enhanced electrical, thermal and self-sensing properties [1–4]. Estimation of electrical properties of these materials is an important design problem that both the industry and the academic researchers are facing in order to design composite materials for lightening strike protection, electromagnetic shielding and electromagnetic interference protection. Apart from a very large number of experimental studies on electrical and thermal characterization of CNT-based nanocomposites [2], there have been numerous studies on computational modeling of electrical transport properties of nanocomposites [5–12]. Most of these computational studies involve replacing the network of nanoparticles with a resistor network, applying Kirchoff's first law or the finite element method to solve a system of simultaneous equations [5,8,11]. However, solving a system of simultaneous equations requires considerable computing resources for realistic simulation sizes.

In this paper, we are exploring computationally low cost approaches without the need for solving simultaneous system of linear equations so that electrical conductivity of large specimen sizes of multi-scale composite materials can be simulated. Union/find [13] algorithms are a class of highly efficient algorithms used in computer science to solve the percolation problem relevant to computer networks. Union/find algorithm has been applied to nanocomposites research to find percolation threshold of nanocomposites [14]. However the disadvantage of solely relying on union/find algorithms, either the quick union/find or weighted union/find is that the original connectivity information of nanoparticles is lost in both of these algorithms. Hence while there are similarities between a network of nanoparticles and a computer network, there are major dissimilarities which cannot be neglected. These dissimilarities are illustrated in Fig. 1 where two networks are shown. From a computer networking point of view, both the networks are identical, as far as flow of information is concerned. However from the point of view of electrical/thermal transport properties, both the networks are not identical. The other dissimilarity is that when solving nanocomposite problems, the number of nodes can be much higher than computer networks. Hence recursive functions, which can be used for solving computer networking problems, can no longer be used for solving nanocomposite conductivity problems because of stack overflow errors [13,15,16]. Hence dedicated new algorithms for efficiently modeling large

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http://dx.doi.org/10.1016/j.commatsci.2014.03.032
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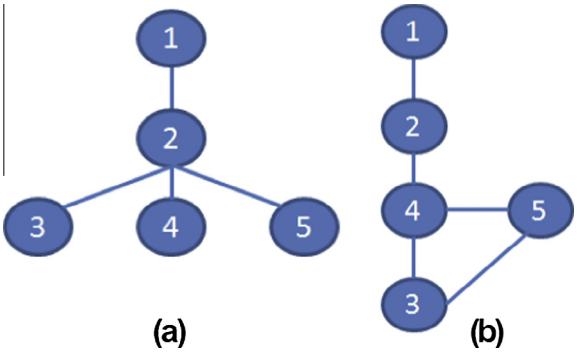

Fig. 1. Computationally equivalent but physically non-equivalent network topologies.

networks of nanoparticles are needed. We have developed a simple time-marching percolation detection algorithm which preserves the original tree network structure of the nanocomposite. We also take advantage of the spatial location of the nanoparticles to minimize our search space and to accelerate the algorithm – this is typically not done for computer networking problem as the physical location of nodes is irrelevant.

2. Details of the computational model and algorithm: Nanocomposites and micro-scale composites

In our model, we have assumed that the contact resistance between nanoparticles is much larger than the resistance of the nanoparticles themselves. We have also assumed that the polymer matrix is perfectly non-conducting. These are standard assumptions used in almost all prior studies on electrical percolation of nanocomposites. We have also neglected the tunneling resistance since tunneling distances are usually of the order of angstroms. The present algorithms for electrical conductivity modeling of nanocomposites has four main steps: (1) creating random nanoparticle network (2) creating nodal tree network of physically (electrically) connected nanoparticles (3) removing dangling bonds (4) removing nanoparticle nodes not connected to 'electrodes' (5) application of Kirchoff's first law. These steps are shown in Fig. 2. The fifth step is optional, as will be discussed later in the paper. In the first step, random nanoparticle networks are generated, depending on nanoparticle volume fraction and geometry. The network tree of the nanoparticles is determined using the procedure shown in Fig. 3. We are assuming that the nanoparticles are very long aspect ratio (~1000) rods, corresponding to carbon nanotubes (CNTs), the most widely used nanoparticles for polymer nanocomposites. Only nanoparticles in neighborhood of each nanoparticle are tested for intersection criteria. For those nanoparticles which are close (directly connected) to the electrodes, connecting nodes are defined as 0 (left electrode) and $N+1$ (right electrode). Where N is the number of nanoparticles in the simulation model.

Once the nodal network tree of each nanoparticle is determined, next, the 'dangling bonds' are removed. Dangling bonds are defined as those nodes which are connected to only one other node. This process is followed iteratively till there are no more dangling bonds. Once dangling bonds are removed, those nodes which are not connected to either the left or right electrode are removed. This is a computationally intensive task and it can be done either by using a recursive function (for example: weighted union/find algorithm) or by using a time marching scheme illustrated in Fig. 4. In the time marching scheme, the nodes connected to left and right electrodes are determine step by step till convergence is reached. In the first step of the algorithm, nodes connected to left electrode are marked as connected (red[1] CNTs in Fig. 4). In the second step, all nodes connected to those nodes are marked as connected. This process is continued till there are no more new nodes to be added. This process is repeated for right electrode as well. Whereas the search direction for the left electrode is from left to right, the search direction for right electrode is from right to left. It has been found that using opposite search direction for the two electrodes drastically reduces the algorithm execution time. For a random nanoparticle of 2400 nanoparticles, the entire tree network for the nanoparticles was determined in just four iterations (time steps), as shown in Fig. 4. Finally, the intersection of two results (nodes connected to left electrode and nodes connected to right electrode) is taken. The intersection of the elements connected to left and right electrodes is the percolation pathway of the nanoparticle nodal network (see Fig. 4).

Time marching algorithm is used here just because recursion depth becomes too high due to large number of nanoparticles [15] for the recursive function implementation which is required for algorithms such as the union/find algorithm. The time marching algorithm does not require large memory or exceptionally faster CPU clock speed for its successful execution. However the time marching algorithm may be significantly slower than the recursive algorithm (such as weighted union/find algorithm). The algorithm continues till the number of elements connected to the electrodes becomes constant.

Only those nanoparticles which are not dangling bonds and are connected to both the left and right electrodes contribute towards electrical transport in the nanocomposite. In addition, dangling bonds can lead to singular matrix in case a system of linear equations (using Kirchoff's law) needs to be solved. Once the network of nanoparticles which are connected to the electrical network and contribute to its electrical resistance has been determined, Kirchoff's law can be applied at each node as shown in Fig. 5. The conduction matrix is determined, as given by Eq. (1) and the matrix is solved for current in each node. Once the current in each node is determined, the overall incoming current can be determined by summing up the currents in the left-most nodes. Similarly, the outgoing current is determined by summing the currents in the right most nodes. Once the incoming and outgoing currents are known, the total resistance of the network can be calculated. Fig. 6 illustrates the formulation of the system of simultaneous equations in the form of matrices. As it can be seen, the conduction matrix is a sparse matrix and there are fast algorithms developed for solving matrices such as these. Parallel processing is also possible using these faster algorithms which will be discussed in our future research papers.

$$\sum_j \frac{V_i - V_j}{R_{ij}} = 0 \tag{1}$$

The modeling approach developed in this section for nanocomposites can be used for multi-scale (micro) composites as well. The only difference is the geometrical domain over which the model is applied. Fig. 7 illustrates the application of algorithm to a multi-scale hybrid composite having CNT volume fraction of 0.01 vol%. The CNTs are 15 μm long with an aspect ratio of 15,000 and the microspheres are 40 μm in diameter. As it can be seen from Fig. 7, the microspheres disrupt the continuous electrical conduction pathways. Red color of the CNTs indicates that they are part of the network of CNTs extending from left to the right electrode (spanning cluster). Blue color indicates that the CNTs are not a part of the spanning cluster formed between the electrodes. As the algorithm developed for nanocomposites is computationally very

[1] For interpretation of color in Figs. 4, 7 and 10, the reader is referred to the web version of this article.