Monte Carlo simulations of effective electrical conductivity in short-fiber composites

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The transport properties of conductive fiber composites are strongly dependent on the interactions between the conductive contents and their overall distribution, which is associated with the percolation and conduction of the relevant fibrous network. In this study, the fibers are modeled as randomly distributed three-dimensional cylinders with each cylinder consisting of a nonconductive core covered by a permeable conductive layer. By discretizing the interconnected surfaces of individual fibers, a finite element method is applied to evaluate the equivalent electrical conductivity of the entire system. Monte Carlo simulations are performed to quantify the relationships between the conductivity and the following factors: (1) the volume fraction, (2) the solidity of fibers, (3) the thickness of the coating layer, (4) the fiber aspect ratio, and (5) the distribution of the fiber orientation angles. In comparison with the model consisting of solid fibers, it has been shown that the coated structure can attain much higher conductivity. The associated percolation properties are also estimated from the computed conductivity, and the results show good agreement with those reported in the literature. These findings can be used as guidance in designing the next generation of multiscale conductive composites.
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I. INTRODUCTION

Over the last decades, carbon fibers have emerged as the main reinforcement filler for high performance composite materials. They have various important properties: high strength and high modulus, fatigue resistance and vibration damping, corrosion resistance, good friction and wear qualities, low thermal expansion, as well as thermal and electrical conductivity.[1] For example, compared to the traditional kinds of composite materials containing industrial carbon (carbon black, dispersed graphite) and metal powders as fillers, composites using chopped carbon fibers as conducting filler can attain a given conductivity with several times less amount of conducting filler content.[2] Therefore carbon fibers offer versatile applications in aerospace,[3] robots,[4] sporting leisure goods,[5] solar cell,[6] semiconductors,[7] and so on. However, their manufacturing costs are generally very high.

Fiberglass, on the other hand, takes advantage of relatively low cost, while providing comparably superior performance. Therefore fiberglass is also one of the most widely used fibers as reinforcement in composites for high mechanical properties nowadays. However, its applications are often limited in the field of electrical engineering due to its electrically nonconductive property. Since carbon is a good electrical conductor and easy to form as thin films,[7] composite fibers consisting of carbon layers and fiberglass cores take advantage of the low cost of fiberglass and meanwhile have the ability to conduct electricity.

Prior researches in the field of electrically conductive fiber composite were mostly experimental.[8] The manufacturing techniques of composite samples, experimental conditions, polymer matrix, and surface treatment of carbon fibers were the main focuses.[9] While these works provided first-hand information about the various important factors, validations of theories as well as new directions for further research, they have limitations because experiments alone can hardly cover every single combination of different factors.

Computational simulations, on the other hand, can be considered as "virtual" experiments. Numerical analyses of material properties including mechanical, thermal, and electrical properties are always challenging tasks in designing new composites. Yet they have the advantages of altering design parameters with a minimal cost. The molecular dynamics simulation is one of the most extensively used numerical tools, but in many cases it is computationally prohibitive due to the small time scales required. In addition, there are a variety of studies on the prediction of physical properties by using the homogenization method for composite materials with periodic microstructure.[10–12] Semianalytical models based on the effective medium theories were also developed to investigate various effects, such as fiber orientation angles and surface contact, on the effective conductivity.[13] But it is difficult to apply these methods to a material system containing stochastically distributed three-dimensional fillers. In fact, for heterogeneous fibrous materials, the transport properties are strongly correlated to the percolation phenomenon, in which the material connectivity is determined by the onset probability of the geometric percolation. This kind of material property is unpredictable from the traditional effective medium theory, and a Monte Carlo simulation scheme must be used to determine the overall statistical behaviors.

The current research concerns the potential applications of electrically conductive fibrous network, and the goal is to

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study various factors that affect the effective electrical conductivity, via Monte Carlo simulations and finite element approaches. This research could also be used to design the next generation of conductive polymer matrix composites,[14] in which the conduction and percolation behaviors of conductive composites with short fibers embedded in a thermoplastic polymer need to be thoroughly investigated. In such composites the fibers are covered with a conductive sizing material that allows the glass fibers to be used with thermoplastic powders for electrostatic spray or electrostatic fluidized bed systems for powder coating. The conductive properties of these composites are strongly dependent on the interactions between the conductive fibers and their spatial distribution.

The methodology discussed in the current research can also be used to predict transport properties of other fibrous materials including carbon nanotubes. Traditionally, the fibrous networks formed by nanotubes were mathematically simplified as two-dimensional line segments[15] or three-dimensional solid cylinders.[16] These models were more or less successful in the evaluation of the equivalent bulk properties. However, the assumption of solid geometry is in fact questionable and could be an intrinsic flaw in these models, because a nanotube by definition consists of a wall of carbon atoms rather than a solid structure. While these two morphological assumptions should not make an appreciable difference for individual fibers or bundles, the effects may be nonnegligible for bonded structures. In fact, it was found that the predicted properties for nanotube sheets assuming solid fibers could be an order of magnitude different from the results actually measured.[16] Although the explanations on this discrepancy vary, it is strongly believed by the authors of the current work that the effect of the tubal geometry must be taken into consideration in all the relevant computations.

II. METHODS

In the simulation models of the current study, each fiber was modeled as a solid cylinder covered by a thin layer of coating material. Since the conductive property of fibers was the only concern here, the nonconductive solid core was not modeled. No flaws were assumed in the material and therefore both the length and thickness maintain the same for all the fibers. The fibers were randomly distributed according to a prescribed probability density function, which was set to a constant unless otherwise indicated. The randomness in the fiber distribution was achieved by a few random parameters that controlled the center locations and the orientation angles of each fiber. The geometries were assumed fully overlapping (meaning that they can penetrate into each other) and therefore the surfaces merged together to form a new, interconnected surface when the overlap occurred, as shown in Fig. 1. This has been believed as a reasonable assumption in the case when the coating layer is sufficiently thin and penetrable.

The generated cylindrical surface of each fiber was then discretized into a finite element mesh using the commercial software MATLAB®[17] and COMSOL®,[18] and the generated nodes were subsequently used for a three-dimensional Delaunay tessellation—a method that generates indices of the points making up tetrahedrons in the tessellation of the nodes. However, in the Delaunay tessellation, the finite element mesh was generated inside a convex hull that was encompassed by all the points. To remove the redundant elements located exterior to the cylindrical surfaces, a homemade program written in C language was implemented. In the program, a numerical iteration was employed to remove the exterior elements, by checking the relative locations of the geometrical center of each element with respect to the cylindrical surfaces. The resulting elements therefore contained the interior spaces of the fibers. The outer faces of the enclosures were then sought for constructing a connected surface mesh that represents the network of the merged conductive layers, as shown in Fig. 2. This step was followed by the generation of an ABAQUS®[19] input file for the subsequent conduction analysis. This input file contained the properties of the conductive material, the mesh information, as well as

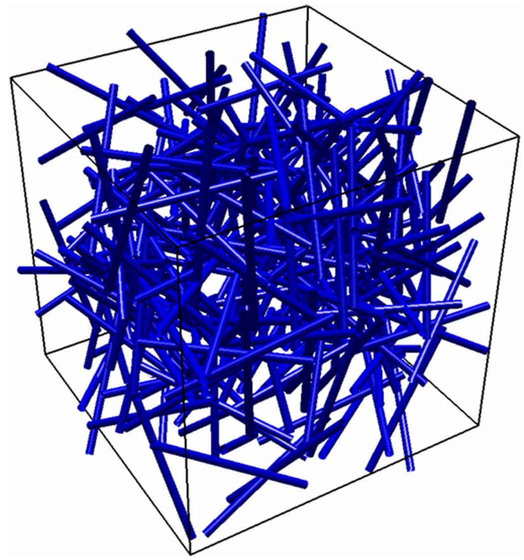

FIG. 1. (Color online) A schematic showing the random fibers in the three-dimensional space. 100 fibers of aspect ratio 20 are generated.

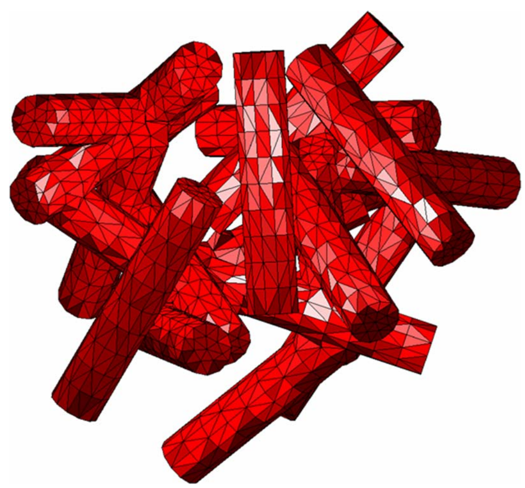

FIG. 2. (Color online) A schematic showing the surface finite element mesh generated on the interconnected fibers. 20 fibers of aspect ratio 5 are modeled.

TABLE I. Frequently used modeling parameters.

Fiber number	Coating material thickness (m)	Radius of fiber (m)	Length of fiber (m)	Unit cell length (m)
200	3×10^{-6}	3×10^{-5}	3×10^{-4}	1×10^{-3}

appropriate boundary conditions. A shell element type was used in the analysis because only the coating material contributed to the conductivity. The solid, nonconductive cores did not contribute to the overall electrical conduction and therefore were not included in the model. A steady state heat transfer analysis was performed to determine the electrical conductivity, in view of the similarity in the electrical conduction and thermal conduction (both processes are governed by Laplace's equation). In fact the computed electrical conductivity and thermal conductivity should be identical when the results are normalized. The boundary condition was specified in such a way that there was a unit voltage difference between the two opposite sides of the unit cell. It can be verified that the obtained reactive flux was equivalent to the bulk conductivity of the system.

Several parameters were used to control the geometrical morphology of the fibers such as the coating layer thickness as well as the fiber diameter and length. The frequently used modeling parameters are listed in Table I, although some of the simulations were performed using different sets of parameters. The ratio of the fiber length to the simulation cell size was maintained to be less than 1/3 to minimize the scaling effects and statistical variations in the results.

Parameters were varied for investigating their effects on the effective conductivity of the network. These parameters include the aspect ratio of fibers, the fiber number, the thickness of the coating layer, the orientation angles of fibers, and the solidity of fibers. The term "solidity" here is used to distinguish between a solid structure and a coated one.

III. RESULT

A. Convergence studies

The finite element number affects both numerical accuracy and efficiency: to accurately model the fiber interfaces, a minimum number of elements must be maintained; however, the computational time increases exponentially with the element number. Therefore, convergence tests were run to make an investigation on the sensitivity of the solution to the meshing parameters, in hopes of achieving a balance between the numerical accuracy and efficiency.

Two major parameters, the element numbers along the length and circumference of a single fiber, have been investigated. Figure 3 shows the conductivity as a function of the element number in the length direction of a fiber. Clearly, the conductivity decreases quickly at the beginning but does not show much difference when the element number exceeds 12. In fact, the conductivity only differs by 5% using 12 and 20 elements. Figure 4 shows the conductivity as a function of the meshing parameter *hcurve* that controls the cutoff curvature of the meshed geometry in COMSOL. This parameter is

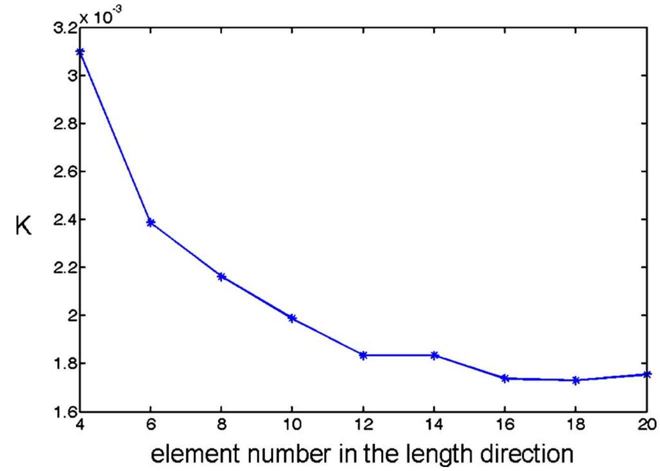

FIG. 3. (Color online) Conductivity K as a function of the element number along the length.

inversely proportional to the circumferential element number. For the two limiting cases with hcurve=0.4 and 1.6, the total number of elements in the cross sectional plane of a fiber is 68 and 4, respectively. It can be seen that the maximum change in the conductivity is approximately 30% regardless of the substantial change in the curvature cutoff parameter. Therefore our conclusion is that the computed conductivity is not very sensitive to the circumferential element number. In practice, 16 elements were used in the cross sectional plane of the model to ensure sufficient accuracy.

B. Parametric studies

The relationship between the conductivity and the coating thickness is presented in Fig. 5. Apparently, the relationship is strictly linear and the curve has a zero intersection with the horizontal axis. This is because the volume of the conductive material varies linearly with the coating thickness.

By adding more fibers into the system, the total volume of the coating material will increase accordingly. Because of the presumed overlapping condition at the joints, the in-

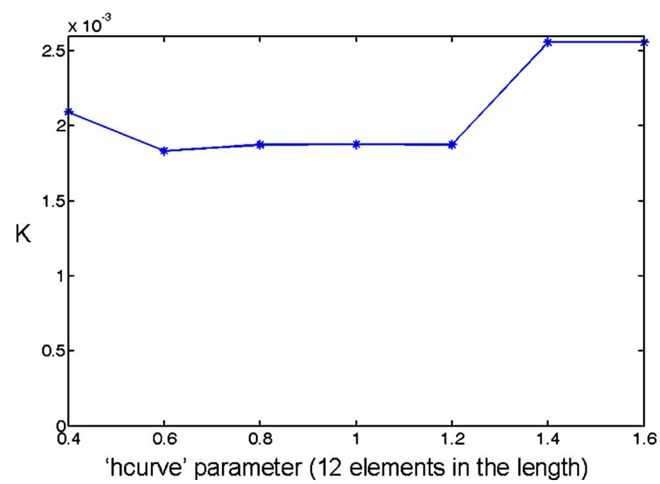

FIG. 4. (Color online) Conductivity K as a function of the element number on the cross sectional plane of a fiber. "hcurve" is a curvature cutoff parameter for a meshed geometry.

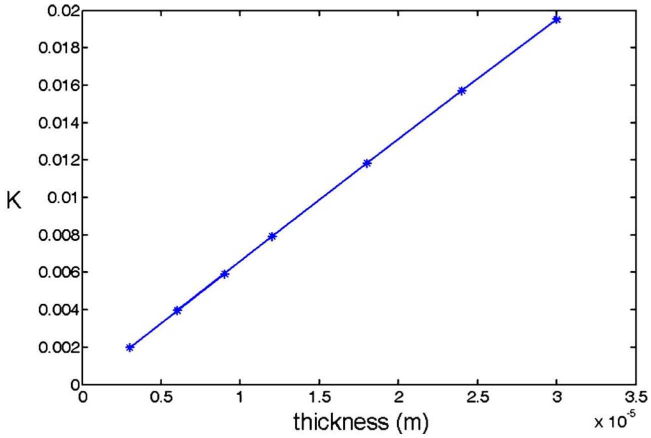

FIG. 5. (Color online) Conductivity K as a function of the coating layer thickness.

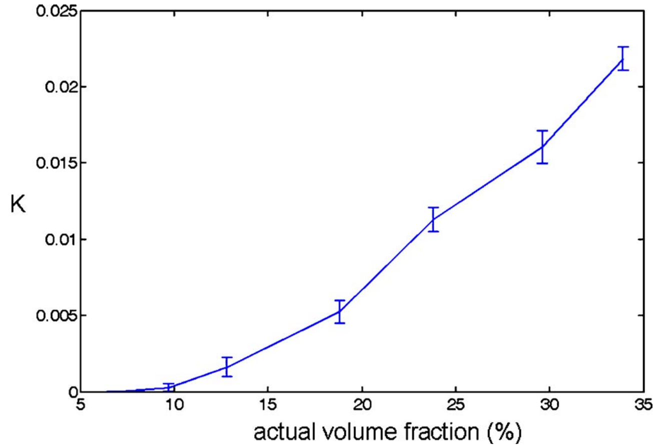

FIG. 7. (Color online) Conductivity K as a function of the fiber volume fraction.

creased volume may not be linearly proportional to the number of the added fibers. In addition, the number of the intersections of fibers (and hence the number of conducting paths) will increase with the fiber number. As a consequence, the effective conductivity increases monotonically with the fiber number as well as the volume fraction. This is consistent with the experimental observation reported in the literature related to nickel-coated carbon fiber composites, in which the electrical resistivity showed filler content dependence.[20] Below a certain threshold of fiber number, however, zero conductivity has been obtained because there are no percolation paths existing in the system. Figures 6 and 7 show the effects of the fiber number and the volume fraction, respectively, on the electrical conductivity. The estimated value of the percolation threshold from Fig. 7 is around 10% in terms of volume fraction, which is consistent with the lower bound value obtained from the percolation analysis for particle aspect ratio equal to 5 in the literature.[21]

The fiber aspect ratio, defined as the ratio of the fiber length to the fiber diameter, is an important parameter that often appears in the literature related to micro- and nanoscale materials research. Figures 8 and 9 show the effective conductivity as a function of fiber number and volume fraction for various fiber aspect ratios. In Fig. 8, the fiber number changes from 200 to 800 with an increment of 100 each time. The change in aspect ratio was achieved by varying the fiber diameter while maintaining the same fiber length and coating thickness. Three distinct aspect ratios were investigated: 7, 10, and 15. Since the volume of the coating material is proportional to the fiber diameter, the effective conductivity decreases with fiber aspect ratio assuming the same fiber number used. However, by assuming the same volume fraction of fibers, the effective conductivity increases with the aspect ratio as shown in Fig. 9, due to the reduced percolation threshold for high aspect ratio fibers. In fact the percolation threshold for each aspect ratio can be estimated by linearly extrapolating the curve in Fig. 9 and finding the intersection with the horizontal axis. The approximated threshold in terms of volume fraction is 7%, 5%, and 3% for fiber aspect ratio 7, 10, and 15, respectively. This explains why a high aspect ratio has been seen as one of the greatest advantages for carbon fibers, especially for carbon nanotubes. The above results are consistent with the former theoretical research on the relationship between the percolation threshold and the particle aspect ratio, regardless of the fibrous or ellipsoidal geometries assumed.[21] The results are also consistent with the experimental results reported by previous researchers. For example, Carmona *et al.*[22] reported a percolation volume fraction of 4.5% for carbon fiber com-

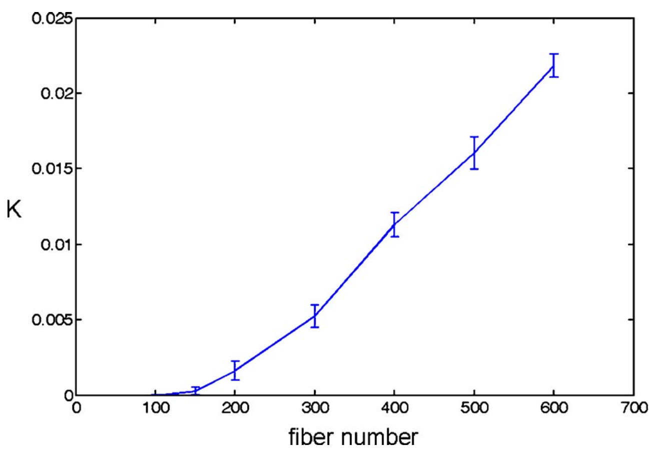

FIG. 6. (Color online) Conductivity K as a function of the fiber number.

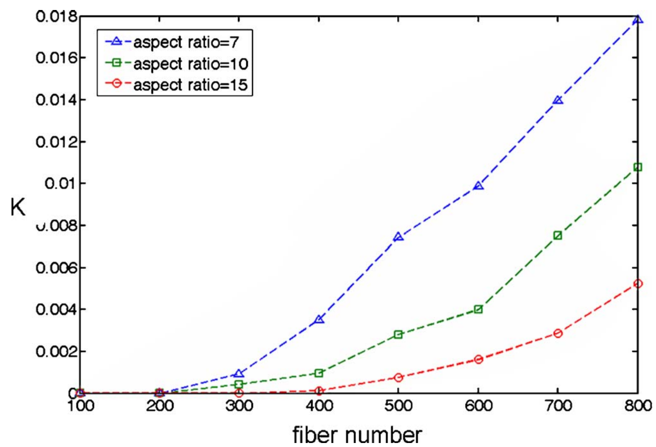

FIG. 8. (Color online) Conductivity K as a function of the fiber number for different fiber aspect ratios.

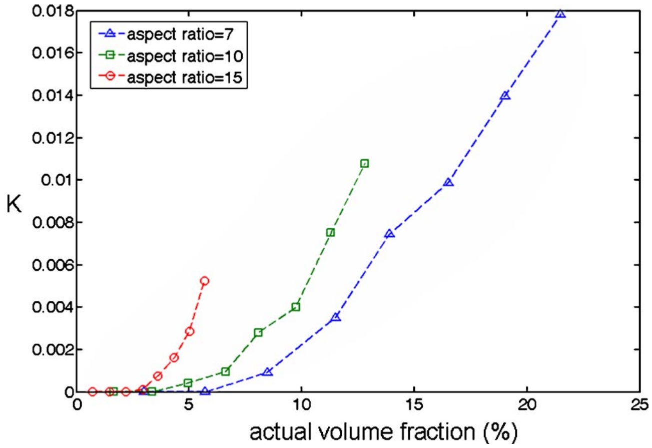

FIG. 9. (Color online) Conductivity K as a function of the fiber volume fraction for different fiber aspect ratios.

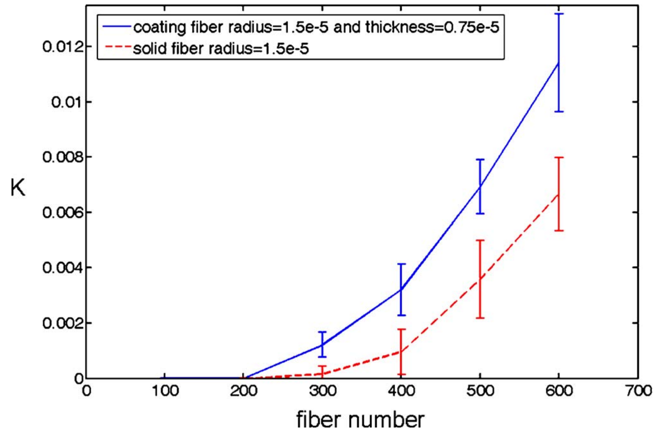

FIG. 11. (Color online) Comparison of conductivity between two types of fibers: (1) solid fiber without coating material and (2) coated fiber with a nonconductive core and a conductive coating layer.

posites with the aspect ratio of carbon fibers greater than 10. This result agrees well with the upper limit of the percolation threshold predicted by the current study.

In the applications involving electrically conductive fibers, electrical conductivity depends on the conducting "chains" of fillers. In addition to the filler content, the orientation of filler is also a very important factor affecting the bulk material properties. Realistic applications may involve controlling factors for angles, but in the current study related to the effects of fiber orientation, a uniform distribution was assumed. Figure 10 shows the effect of the fiber orientation angle, θ on the effective conductivity. θ has been defined as the angle between the fiber axis and the direction of the potential gradient. It varied from 0 to $\pi/2$ with an increment of $\pi/18$, while the angles of the projections of the fibers on the x-y plane were randomly distributed. Both the mean values and standard deviations are presented in Fig. 10. It has been noticed that the standard deviations of the results vary with the fiber diameter. When the volume fraction of the coating material is sufficiently low, the uncertainties in the results are significantly large. The fiber diameter used in Fig. 10 was set to 1×10^{-4} m. Clearly, the effective conductivity decreases with the orientation angle. When θ=90°, the fibers were directed in the x-y plane that is perpendicular to the z axis. The contact between fibers in the x-y plane does not contribute to the formation of the conducting paths along the z axis, and therefore the conductivity reaches its minimum value. When θ is zero, however, the fibers were parallel to the z axis and thus aligned with the direction of voltage gradient. Thereby the chance of forming conducting paths is the greatest, leading to the maximum effective conductivity in the computational result.

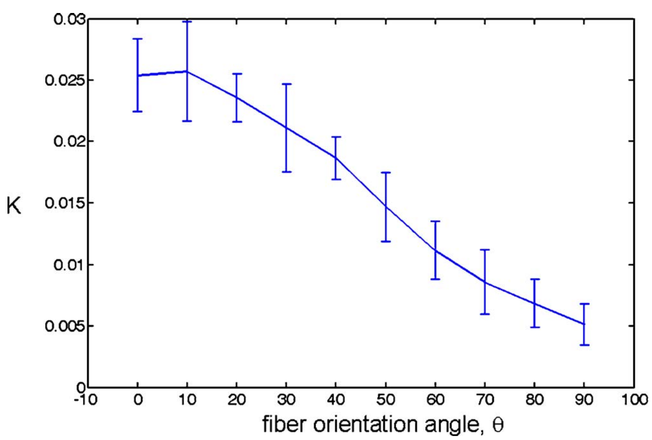

FIG. 10. (Color online) Variation of conductivity against the fiber orientation angle θ. This angle is defined as the maximum angle between the fiber axis and the potential gradient.

The effective conductivity is compared in Fig. 11 for two different types of fibers: (1) fibers with conductive coating material and nonconductive cores and (2) fibers made of conductive material only. The core aspect ratios of the two fibrous systems were the same (both of them were 10) because of the same core diameters and lengths used. In addition, it can be easily verified that the volumes of the conductive materials were also the same in the two models. Clearly, the conductivity of the coated structure is higher than that of the solid one. For example, when the fiber number is 600, the effective conductivity of the coated model is approximately 50% higher than that of the solid model. This is because in the coated structure the conductive material has a larger mean diameter and therefore a greater probability of making touch with other fibers, whereas in the solid structure, the conductive material was concentrated within a relatively smaller space. Therefore, coated fibers can attain higher conductivity than solid fibers when the same amount of conductive material is used. This agrees well with the experimental results showing that at the same fiber content, the conductivity of the composites filled with nickel-coated carbon fibers is much greater than that of ordinary carbon-fiber-filled composites.[23] This improved performance should merit the use of a coated structure for fibrous materials in engineering practice.

IV. DISCUSSIONS

A. Effects of imperfect coating layer and electrical anisotropy

In the computational model presented in this study the coating material has been modeled as a uniform layer that encompasses a solid core. This is an idealized yet useful simplification in the simulation. However in reality, during the coating process the fiberglass may not be fully covered and the coating thickness could vary. In the literature relevant to the fabrication processes of coating carbon,[24] it was found that the porous nature of the coated carbon and the low carbonization temperature led to the low electrical conductivity of coated carbon. It was further claimed that a better process would be decomposition of hydrocarbons by chemical vapor deposition at temperatures below 1000 °C, which would produce carbon films of smooth surfaces, containing randomly distributed pores of a few hundred nanometers in size. The inhomogeneous characteristics of coating materials would reduce their ability to conduct electricity. Therefore, if this factor were considered, the advantage of coated carbon fiberglass media over solid carbon fiber showed in Fig. 11 would probably be reduced. Further, the electrical anisotropy[25] of fibers in carbon fiber composites could induce another source of inhomogeneity in the material and may impact the overall effective electrical conductivity.

B. Tunneling effects

In the current model, the electrical conduction occurs only when two fibers have overlapping volume. In reality charge transfer takes place along filler network with a direct electric contact between neighboring fibers. The larger the conducting chains in the network, the lower the electrical resistance of the system would be. From a set of experiments of adding fillers to film composite material,[2] it was observed that there was a sharp decrease in the electrical resistance for the content of filler in the form of polydisperse carbon fibers in comparison with equisized particulate filler. This observation supported the contact mechanism between conducting chains responsible for the conduction in composites at a relatively large size scale. However, when the size scale reaches submicron or nanometer,[26] the conductivity of a filled system is determined by the size of the space between conducting filler particles which electrons jump across according to a tunneling or emission mechanism of conductivity. This is especially important in modeling nanotube networks. Although the tunneling effects were not considered in the current work, they will likely be incorporated in our future study. A strategy for appropriately modeling the tunneling effects would be to construct a number of "gap elements" around neighboring fibers. Each gap will have a gap resistivity to approximate the tunneling effect. The gap resistivity can be determined either from experiments or from fundamental quantum theories.

C. Effect of contact resistance

In the current model, in the case when the spatial locations of some portions of two fibers coincided, they were simulated as a merged joint at their original locations. Therefore the electrical resistances between the contact surfaces were not considered. During the mixture and compression processes in manufacturing, however, mechanical deformations along the contact surfaces of fibers are inevitable. When impermeable coating layers or rough surfaces are involved, the contact resistance may be significant. Overall, since the electrical contact resistance was ignored in the current model, the computed effective conductivities could have been overestimated compared to the actual values.

D. Effect of surface treatment

The physical and chemical treatment on the surface coating is not considered in the present models. Besides the content and arrangement of carbon fibers, the physical-chemical interaction at the fiber-matrix interface plays an important role in the electrical properties of the composites. Previous researches showed that different surface treatment methods could result in significantly different electrical conductivity.[27] To incorporate this effect in future study, the material properties for single fibers should be adjusted by a factor prior to the finite element analysis. The required parameter can be determined inversely from experiments.

E. Temperature effect

The effect of temperature has not been included in the current work. In practice, the stability of the electrophysical properties could be an important performance property, primarily influenced by temperature.[28] The coefficient of thermal expansion can vary greatly between the polymer matrix and the carbon fillers, and usually the former is much higher than the latter. The study on the relationship between temperature and film composite materials with carbon fiber as the fillers[2] showed a sharp increase in the resistance of the film composite. It was then hypothesized that the thermal expansion of polymer matrix interrupted contacts between conducting particles and that the contacts between the fibers were restored when temperature decreased. A study of this effect will require a complicated analysis on the coupled thermal, mechanical and electrical processes, which will be part of our research objectives in the future.

V. CONCLUSIONS

This research studied the effective electrical conductivity of a three-dimensional network consisting of coated fibers as opposed to widely studied noncoated solid fibers. Specifically, short fibers were modeled as randomly distributed cylinders with each containing a nonconductive core and a thin conductive coating layer. A Monte Carlo simulation method in conjunction with the finite element discretization scheme was employed to investigate the conductivity of the system as functions of various geometrical and material parameters.

Several conclusions have been drawn, as follows:

(1) The effect of the coating material on the conduction and percolation of short fibers has been quantified. It has been shown that there exists a linear relationship between the volume of the coating material and the overall effective conductivity.
(2) The addition of an insignificant amount of fiber (3%–5%) can cause substantial increase in conductivity near the percolation threshold. A quantitative relationship has been obtained between the volume fraction and the conductivity for short fibers.

(3) The effect of fiber aspect ratio has been studied for several representative values of aspect ratio. Fibers with larger aspect ratio yield larger effective conductivity assuming the same volume fraction.

(4) Conductivity is strongly dependent on the fiber orientation angle. Its maximum value is obtained when the fibers are aligned with the direction of electric current or the applied potential gradient. The relationship has been quantified between the orientation angle and the effective conductivity.

(5) Comparisons between the solid and coated models revealed that the volume of the conductive material is not the only factor determining the overall effective conductivity, and that coated fibers can attain much higher conductivity than solid fibers. But this advantage could be undermined by inhomogeneous properties of conductive materials in actual applications.

These conclusions are consistent with formerly reported experimental results and theoretical predictions found in the literature. Since the contact problems were not modeled in the current research, the immediate future work will include the development of an efficient tool for modeling contact resistance as well as the interactions among the thermal-mechanical-electrical couplings.

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