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Thermal conductivity enhancement of carbon fiber composites

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ABSTRACT

The effective thermal conductivity enhancement of carbon fiber composites is investigated in this contribution using a three-dimensional numerical method. First a more realistic three-dimensional distribution of fibers dispersed in a matrix phase is reproduced by a developed random generation-growth method to eliminate the overrated inter-fiber contacts by the two-dimensional simulations. The energy transport governing equations are then solved through the three-dimensional structures using a high-efficiency lattice Boltzmann scheme. The resultant predictions agree well with the available experimental data. Compared with the existing theoretical models, the present method does not depend upon empirical parameters which have to be determined case by case, so that it is useful for design and optimization for new materials, beyond prediction and analysis just for existing composites.

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1. Introduction

Enormous amount of research interests have been sparked in their potential applications since the carbon fiber materials were discovered. One promising usage of carbon fibers is to act as fillers in composite materials to improve the thermal, electrical and mechanical performances [1–5]. Recently it has been reported that the thermal conductivities of carbon fiber-in-oil suspensions and carbon fiber-in-polymer composites were found to be significantly enhanced [2,3] by up to, for example, 150% with just 1% volume fraction loading of fibers [2]. However few analytical models are available for accurate predictions of the thermal property of such carbon fiber composites because of their unique structural morphology. The models treating the carbon fibers as spherical inclusions heavily underestimated the thermal conductivity enhancement [2], while those regarding the carbon fibers as infinitely long fibers grossly overestimated the enhancement effect [6] when compared with the experimental data. Several new models have thus been proposed to model and explain such anomalous thermal conductivity enhancement in carbon fiber composites in the last five years. Chen et al. proposed some simple models to include the tube-end transport effect on the thermal conductivity [7]. Very recently, Deng et al. presented analytical models on the thermal conductivity enhancements in carbon fiber composites as functions of the volume fraction, conductivity anisotropy, aspect ratio, non-straightness and interfacial thermal resistance of the carbon fibers, and the predictions agreed well with the experimen-

tal data [8]. Despite such achievements, the prediction of carbon fiber composite properties is still far from a full solution in theory. Most theoretical models suffer from their inherent nonphysical assumptions and/or the empirical parameters included.

Owing to the rapid development of computer and computational techniques, numerical modeling methods have provided an alternative in dealing with the properties of fibrous materials with complex geometries. To achieve a reliable prediction, one needs to work on two aspects: a good description of the structural details of fibrous materials, and an efficient numerical method for the solution of energy equations through the fibrous structures.

It is extremely difficult to completely describe the internal structure details of a fibrous medium due to its complex and stochastic nature. One often can only acquire the statistic-based average information such as the mean porosity, the mean fiber contacting length and the orientation range. Nonetheless there have been some attempts to reproduce the statistic-stochastic characteristics of fibrous composite materials through several different methods [9–14]. For instance, recently the tessellation-based Voronoi cells were extracted for generating the fiber distribution [9,10]. Eichhorn and Sampson [11] and Scharcanski et al. [12] investigated the stochastic characteristics of the internal structure of fibrous materials and suggested relationships between the statistical mean porosity and the micro-structural characteristics which could be helpful for fibrous structure regeneration. Moreover Wang et al. [14] recently proposed a random generation-growth algorithm for reproducing two-dimensional (2D) random microstructures of fibrous materials and investigated the effects of fiber aspect ratio, orientation angle and volume fraction on the effective thermal conductivity. However the inter-fiber

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contacts may be overrated for a 2D structure so that a three-dimensional (3D) structure is necessary for accurate modeling and predictions.

Next to predict the effective thermal conductivity of a fibrous medium with a given structure, the energy equations must be solved through the multiphase structure. The structural complexities bring two challenges when the governing equations are to be solved by conventional numerical tools such as the finite difference [15] and finite element methods [10]. The first is the constraint of conjugate heat transfer at interfaces: for steady pure heat conduction through multiple phases, temperature and heat flux continuities have to be ensured at the interfaces when the contact thermal resistance is negligible [16,17], thus demanding extremely high computational resources for a fibrous medium with numerous such interfaces. The second is the requirement of grid refinement for complex structures: the accuracy of a conventional numerical method is strongly dependent on the grid size so that an extra fine grid is needed for the complex geometry. Consequently when dealing with such multiphase conjugate heat transfer problems in fibrous materials, these requirements confine the computational domain into a very limited area. To overcome these two difficulties, we [18] developed a high-efficiency lattice Boltzmann algorithm to solve the energy equations in multiphase materials where the continuity constraints are satisfied and much less grid number is required for the same accuracy. The predictions agree well with the existing experimental data for granular multiphase porous media.

The objective of this contribution is to investigate the effective thermal conductivity of carbon fiber composites with higher accuracy and efficiency: three-dimensional random microstructures of carbon fiber composites are reproduced by the random generation-growth (RGG) method and then the energy transport equations are solved by the developed D3Q15 lattice Boltzmann method. The predicted results will be validated with the available experimental data.

2. Numerical methods

2.1. Random generation-growth algorithm for 3D microstructures

Consider a three-dimensional two-phase fibrous composite, assuming each fiber of equal length represented by a straight line. For simplification, we generate the structure in a 3D cubic system with fibers uniformly distributed in the system and each fiber has a unit square cross section. Different from the 2D cases [14], a 3D case an angle pair (φ, θ) to define a fiber's orientation as shown in Fig. 1. The structure generation process can be described as follows:

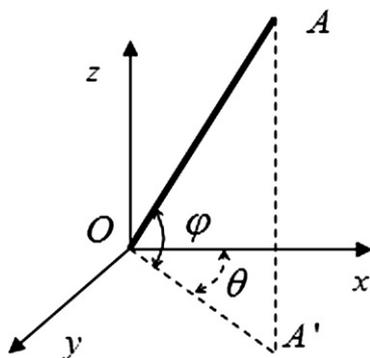


Fig. 1. A fiber oriented in a three-dimensional space. The fiber is represented by a straight line OA. A' is the projection of A on x-y plane. (θ, φ) is the orientation angle pair of the fiber OA.

- (i) Randomly locate the fiber cores based on a core distribution probability, c_d , which is defined as the probability of a given cell being a core of a fiber. Apparently the value of c_d is determined by the fiber number density.
- (ii) Assign a random numbers pair to the orientation angles (φ, θ) for each fiber core to define the fiber orientation. For isotropic materials, the orientation angles can be any values within $0 \sim 2\pi$.
- (iii) Grow each fiber from the core along (φ, θ) direction until the fiber length l reach the specified value or the volume fraction of fibers attains the given level.

The flowchart of the generation process and more of the parameters discussion can refer to our previous work in [14].

2.2. Lattice Boltzmann algorithm

To calculate the effective thermal conductivity of fibrous materials, we have to solve the energy transport equations for the temperature and heat flux fields. For a steady pure thermal conduction with no phase change, no convection and no contact thermal resistance, the equations to be solved are a series of Poisson equations subject to temperature and heat flux continuity constraints at the phase interfaces

$$(\rho c_p)_f \left(\frac{\partial T}{\partial t} \right) = \nabla \cdot (k_f \nabla T) \quad (1)$$

$$(\rho c_p)_s \left(\frac{\partial T}{\partial t} \right) = \nabla \cdot (k_s \nabla T) \quad (2)$$

where subscript f represents the fluid, and s the solid; T is the temperature, ρ the density, k the thermal conductivity, and c_p the specific heat capacity. At the interfaces between the two phases once in equilibrium, there are

$$T_{f,int} = T_{s,int}, \quad (3)$$

$$k_f \frac{\partial T}{\partial \hat{n}} \Big|_{f,int} = k_s \frac{\partial T}{\partial \hat{n}} \Big|_{s,int}, \quad (4)$$

where the subscript "int" corresponds to the interfaces and \hat{n} represents the unit normal vector to the interfaces. As stated above, the geometry complexity and the huge numbers of such interfaces demand huge computational costs when using conventional partial differential equation solvers. Recently, the lattice Boltzmann method (LBM) has been developed to solve effectively the fluid–solid conjugate heat transfer [18,19]. Compared with the conventional PDE solvers, the most important advantages of LBM are the easy implementations of multiple inter-particle interactions and complex geometry boundary conditions [20–22], and the conservation laws can hold automatically without additional computational efforts [23]. LBM has gained several successes in predicting the effective thermal conductivities of conventional granular porous media verified with available experimental data [18,19]. Here we present a three-dimensional scheme of LBM and then use it to predict the effective thermal conductivity of the carbon fiber composites.

For solving the energy transport governing equations in a three-dimensional multiphase system, the energy evolution equation can be generally given as [18]

$$g_z(\mathbf{r} + \mathbf{e}_z \delta_t, t + \delta_t) - g_z(\mathbf{r}, t) = -\frac{1}{\tau_n} [g_z(\mathbf{r}, t) - g_z^{eq}(\mathbf{r}, t)], \quad (5)$$

where \mathbf{r} is the location vector, t the real time, δ_t the time step, \mathbf{e}_z the discrete lattice velocity, g^{eq} the equilibrium distribution of the evolution variable g_z , and τ the dimensionless relaxation time with the subscript n representing the n th phase. For a three-dimensional fifteen-speed (D3Q15) lattice Boltzmann model there are

$$g_x^{eq} = \begin{cases} 0 & \alpha = 0, \\ T/9 & \alpha = 1-6, \\ T/24 & \alpha = 7-14 \end{cases} \quad (6)$$

$$\mathbf{e}_x = \begin{cases} (0, 0, 0) & \alpha = 0, \\ (\pm 1, 0, 0)c, (0, \pm 1, 0)c, (0, 0, \pm 1)c & \alpha = 1-6, \\ (\pm 1, \pm 1, \pm 1)c & \alpha = 7-14 \end{cases} \quad (7)$$

and

$$\tau_n = \frac{9}{5} \frac{k_n}{(\rho C_p)_n c^2 \delta_t} + 0.5, \quad (8)$$

where k is the thermal conductivity of each phase, (ρc_p) the volume thermal capacities which has to be set identical for different phases based on the conjugate heat transfer effect [24] and the c a pseudo sound speed whose value can take any positive value theoretically only to ensure τ_n value within (0.5,2) [25].

For the isothermal boundary treatment, we follow the bounce-back rule of the non-equilibrium distribution proposed by Zou and He [26]. For the insulated boundary, a specular reflection treatment is implemented to avoid energy leak along the surfaces [18].

The temperature and the heat flux can then be calculated according to [18,27]

$$T = \sum_{\alpha} g_x, \quad (9)$$

$$q = \left(\sum_{\alpha} \mathbf{e}_x g_x \right) \frac{\tau_n - 0.5}{\tau_n}. \quad (10)$$

After the temperature field is solved, the effective thermal conductivity, k_{eff} , can be determined

$$k_{eff} = \frac{L \cdot \int q \cdot dA}{\Delta T \int dA}, \quad (11)$$

where q is the steady heat flux through the system cross section area dA between the temperature difference ΔT with a distance L .

3. Results and discussion

Since the present lattice Boltzmann solver has been validated for predictions of effective thermal conductivities of granular porous media by comparing with the available theoretical solutions and experimental data in our previous work [18,19], we employ it here directly for three-dimensional carbon fiber composites and compare the numerical results with the existing experimental data. In our simulations, random microstructures of carbon fiber composites are generated on a 3D grid system, and then the energy transport governing equations solved through the structures by the high-efficiency lattice Boltzmann method. Similar to other types of porous media [18], the porous structures thus generated exhibit realistic stochastic features, thus leading to fluctuations around an averaged result of each trail with given parameters. We have studied such numerical uncertainty and found that the fluctuation is strongly dependent on the grid number and slightly affected by the fiber volume fraction. A larger grid number and a higher fiber volume fraction will lead to smaller fluctuations.

Consider a carbon fiber-in-oil suspension with low fiber loadings. Choi et al. have reported anomalous thermal conductivity enhancement in such mixtures [2], which could not be accurately predicted by the established models for the conventional granular porous media [6,7]. Very recently, Deng et al. have proposed new analytical models and resulted in good agreements between the predictions and the experimental data [8]; however a few free parameters in their models still have to be determined empirically and case by case. While in our numerical method, no empirical parameters exist. We generate the fibers in a $L_c \times L_c \times L_c$ grid sys-

tem. Since we know from the experiment that the average aspect ratio of the fibers is very high but still a finite value, we generate the fibers with length $L = 2L_c$ by performing a periodic treatment when the growth meet the boundaries. Such a treatment can ensure the high-aspect-ratio effect without losing the fiber-end transport effect when the aspect ratio of fibers is very high, such as larger than 1000. Our numerical tests have proved that the predicted effective thermal properties seldom vary with the grid number (L_c) if the other parameters are given when L_c is no smaller than 50. For saving computational efforts, we use $L_c = 60$ for our simulations in this contribution. The fiber volume fraction is controlled by varying values of the fiber core distribution probability, c_d .

Fig. 2 compares the predicted effective thermal conductivities of carbon fiber-in-oil suspensions using the present method with the measured data and predictions by other theoretical models. The properties used in simulations include thermal conductivity of carbon fibers $k_f = 2000$ W/m K and thermal conductivity of the oil mass $k_m = 0.1448$ W/m K. Because the fiber loadings are very low, the fluctuations in our numerical predictions are quite remarkable (nearly 10%) but the relative errors might still be comparable with those of experimental data. The present predictions appear a little lower than the experimental data, which may lie in the convective heat transfer in the fiber-oil suspension is not counted in our model. Compared with the results from other theoretical models, the carbon fibers enhancement result is actually in between that predicted by the spherical particle model, also known as the Hashin and Shtrikman (HS) lower bound, and the HS higher bound [28]. The interaction direct derivative (IDD) analytical model can also provide good results with well-estimated empirical parameters, $p = 2000$ and $\eta = 0.155$ and $q \rightarrow 1$ corresponding to random fiber distribution [8].

Frusteri et al. [29] have recently measured the thermal conductivity of a phase change material containing carbon fibers. The fiber loading is much higher (up to 10 wt.%). We compare their experimental data with our numerical predictions and also with those from Deng et al.'s theoretical models. The thermal properties used in our simulations are $k_f = 180$ W/m K and $k_m = 0.47$ W/m K [30]. Fig. 3 shows good agreements between our predictions and the measured data when the volume fraction is lower than 6%. When the fiber fraction is high, the inter-fiber contacts will affect the effective thermal property. Because no thermal contact resistance [7] is considered in our model, the predicted thermal

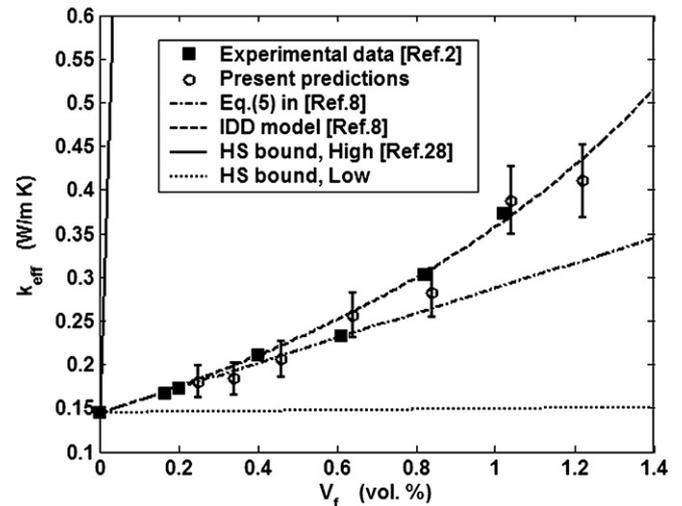


Fig. 2. Measured and predicted effective thermal conductivities of fiber-in-oil suspensions. The parameters are $k_f = 2000$ W/m K, $k_m = 0.1448$ W/m K; for straight cable model, $p = 2000$ and $\eta = 0.155$; for IDD model, $p = 2000$ and $\eta = 0.155$ and $q \rightarrow 1$.

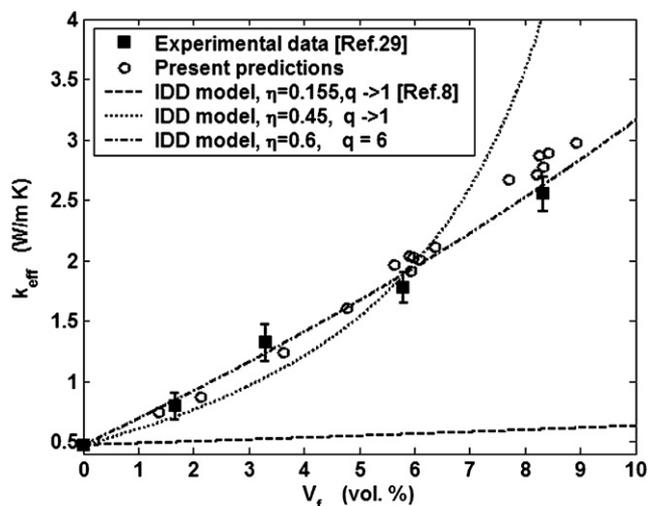


Fig. 3. Measured and predicted effective thermal conductivity of phase change materials containing carbon fibers. The properties used are $k_f = 180 \text{ W/m K}$, $k_m = 0.47 \text{ W/m K}$.

conductivities are a little higher than but still comparable with the experimental data when the fiber fraction is high. The numerical relative errors due mainly to the fibers distribution randomness are at the same level as those of experimental measurements. With respect to the experimental data, the accuracy of the IDD model [8] strongly depends on the estimations of the empirical parameters. The IDD predictions are much lower than the measurements when we use the same set of parameters as used for the above fiber-in-oil suspension case where $p = 2000$ and $\eta = 0.155$ and $q \rightarrow 1$; the predictions will improve if we use $\eta = 0.45$ and $q \rightarrow 1$ corresponding to the random isotropic distribution of fiber. However when the parameters are set as $\eta = 0.6$ and $q = 6$, then the predictions of IDD model agree well with both the experimental data and our numerical results. Whereas the present numerical prediction method does not need any empirical parameters and it is thus very suitable for design and optimization of new materials, instead of just for prediction and analysis of existing composites.

4. Conclusions

A three-dimensional numerical method has been presented for predictions of the effective thermal conductivity of carbon fiber composites. The random generation-growth method is developed for generating three-dimensional fibers distribution in a mass phase, so as to avoid the overrated inter-fiber contacts in two-dimensional cases. The energy transport governing equations for the three-dimensional structures are solved through using a high-efficiency lattice Boltzmann scheme. The predicted thermal conductivity enhancements using the present method agree with the available experimental data for both a low-loading fiber-in-oil suspension and a high-fiber-loading phase change materials. Compared with the existing theoretical models, the present method does not require any empirical parameters so that it is highly suitable for design and optimization of new materials, in addition to prediction and analysis for existing composites.

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