Analytical and numerical assessment of the effect of highly conductive inclusions distribution on the thermal conductivity of particulate composites

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Abstract

Highly conductive composites have found applications in thermal management, and the effective thermal conductivity (ETC) plays a vital role in understanding the thermo-mechanical behavior of advanced composites. Experimental studies show that when highly conductive inclusions embedded in a polymeric matrix the particle forms conductive chain that drastically increase the ETC of two-phase particulate composites. In this study, we introduce a random network three dimensional (3D) percolation model which closely represent the experimentally observed scenario of the formation of the conductive chain by spherical particles. The prediction of the ETC obtained from percolation models is compared with the conventional micromechanical models of particulate composites having the cubical arrangement, the hexagonal arrangement and the random distribution of the spheres. In addition to that, the capabilities of predicting the ETC of a composite by different analytical models, micromechanical models, and, numerical models are also discussed and compared with the experimental data available in the literature. The results showed that random network percolation models give reasonable estimates of the ETC of the highly conductive particulate composites only in some cases. It is found that the developed percolation models perfectly represent the case of conduction through a composite containing

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randomly suspended interacting spheres and yield ETC results close to Jeffery's model. It is concluded that a more refined random network percolation model with the directional conductive chain of spheres should be developed to predict the ETC of advanced composites containing highly conductive inclusions.

Keywords: Effective thermal conductivity, two phase-composites, particulate composites, the distribution of inclusions, highly conductive composites.

1. Introduction

Particulate composites are widely used in thermal storage management, and there is an increasing demand for efficient and highly conductive particulate composites in electronic packaging, heat sinks, and other appliances. The heat transfer capability of particulate composites can be improved by varying spatial distribution of the inclusions and thermal properties of the constituents ([1],[2]). Recently, experimental studies showed that when highly conductive inclusions are embedded in a less thermally conductive matrix, the effective thermal conductivity (ETC) of the composite increases significantly with the increase of volume fraction (V_f) of the inclusions [3]–[7]. A precise model capable of predicting the ETC of the highly conductive particulate composites plays a significant role in evaluating the thermal and stress analysis of advanced composites.

The ETC of two-phase isotropic composites depends on characteristics of the constituents such as shape, geometry, spatial distribution, thermal conductivities, V_f and matrix-inclusion interfacial effects [8], [9]. Various bounds of the ETC of composite material have been proposed, such as, based on the rule of mixture, [10], variational principle [11] and statistical correlation functions [12]. These bounds are used to define the thermodynamic limits of the ETC of two-phase composites [2].

Several analytical models have been proposed to predict the ETC of a composite material based on simplified microstructures of the composites. Maxwell [13] was one of the first persons to investigate the conduction of two-phase composites analytically by considering the dilute suspension of non-interacting spherical particles. An exact expression for the ETC of composites was obtained by solving the Laplace equation. Maxwell's approach has been extended by many researchers, such as Fricke [14], Nielsen [15], Bruggeman [16], Hamilton and Crosser [17], Benveniste [18] and Hasselman and Johnson [19]. All these modifications extended the applicability of Maxwell's approach for a variety of conditions. However, these models are not applicable to predict the ETC of composites having highly conductive inclusions [3].

Many micromechanical models have been proposed to predict the ETC of composites. Benveniste [20] formulated the ETC for multiphase systems by determining the average flux in each constituent. The homogenization was performed using the Mori–Tanaka [21] and a generalized self-consistent model [2]. Verma et al. [22] derived the ETC of two-phase composites containing spherical particles arranged in a three-dimensional cubic geometry. The arrangement was divided into multiple unit cells, and resistor model was used to determine the ETC of the unit cells. All the available micromechanical models in literature offered good predictions of the overall ETC when the V_f was relatively small or when the conductivity of the particle (Kp) was comparable to the conductivity of the matrix (Km).

Various experimental, computational and theoretical studies showed that the ETC of a composite can be significantly altered by the particle shape, size, interfacial thermal resistance aside from volume fraction and Kp/Km ratio ([2],[23]). Nan et al. [24] developed a generalized effective medium approach (EMA) formulation to compute the ETC of arbitrary ellipsoidal particulate composites with interfacial thermal resistance. The formulation accounted for the effect

of particle shape, size, orientation distribution, volume fraction and interfacial thermal resistance. With and without interfacial thermal resistance at a large Kp/Km ratio, the ETC significantly increases with the anisotropy of the inclusion shape. It was suggested that with large Kp/Km ratio, the ETC of a composite could be enhanced considerably by reinforcing the matrix with prolate inclusion (e.g., whiskers); while for small Kp/Km, the spherical particle is suitable to improve ETC.

In nano-enhanced composites, the nanoparticles tend to agglomerate during the solid-liquid phase transition. Some studies showed enhancement in ETC with the formation of percolation networks ([25]); while others showed a reduction in ETC ([26]). The percolation threshold depends on the size and the shape of the nanoparticles ([27], [28]). Nan et al. ([24], Gao et al. [29] and Xie et al. [30]) studied the influence of the inclusions on ETC below the percolation threshold while recently Wemhoff [31] developed a percolation threshold model for composite material containing uniformly distributed and oriented cylindrical or prolate inclusion. Along the same line, Wemhoff and Webb [32] studied the influence of both spherical clustering and linear percolation network formation on the ETC of a composite. The EMA and percolation theories were employed for percolated and unpercolated areas. It was shown that both spherical clustering and linear agglomeration tend to reduce the ETC. However, the sensitivity analysis of the model suggested that linear agglomeration can increase bulk thermal conductivity. It was found that when the ratio of inclusion–matrix to inclusion-inclusion Kapitza resistance increases then the relative thermal resistance reduces through the percolation network compared to the unpercolated regions of the domain, which in turn leads to an increase in the ETC of a composite. Recently, Chatterjee et al. [33] proposed a computational heat conduction model based on percolation theory for thermal

conductivity of composites with a high volume fraction of filler in the matrix. They considered cubical particle percolation effect to compute the ETC of the composite.

Several empirical models have been proposed to predict the ETC of highly thermally conductive composites. Agari and Uno ([3], [4]) and recently by Zhang et al. [34] have shown that at higher V_f and high ratios of particle to matrix thermal conductivity, i.e., $V_f > 15\%$ and $K_p/K_m > 100$, a particle interaction in the form of a conductive chain mechanism is exist. This mechanism accelerated the heat conduction process, which was observed due to an increase in the overall ETC of the composite. It was concluded that the V_f and the geometry of the particle were responsible for forming the conductive chain mechanism. Zhou et al. [35] showed that at higher particle concentrations, some particles flocculated to form conductive chains. They introduced the heat transfer passage which took the effect of local concentration fluctuation into account to evaluate the ETC of the composites. Although, Agari and Uno [3] and Zhou et al. [35] models are applicable to predict the ETC of highly conductive composites but the parameters involved in their models need to be determined from the experiments; therefore, these models are rarely used.

The percolation threshold is usually defined in terms of the volume fraction (V_f) and highly dependent on the inclusion geometry. According to the percolation theory [36], the value of the threshold for the cubic particle, V_{fc}, is 0.3117–0.3333. When $V_f < V_{fc}$, the conductive particles are mainly dispersed, and the effect of the particles' interaction on ETC is small. When V_f goes beyond V_{fc}, the connections of the particles increase and the formations of the conductive chains dominate the rise of ETC. Yin et al. [37] mentioned that the threshold limit of percolation could go as high as 0.78. More recently, Liang et al. [28], Gao and Li [38], Wemhoff [31], Wang et al. [27] also observed the dependence of particle size and shape on the percolation threshold.

The focus of this study is to present three-dimensional FE percolation models with an attention to find the effect of high conductivity percolation path on the overall ETC of a composite. In this study, FE percolation models were developed purely on thermal conduction physics and analyzed the influence of increasing inclusions' volume fraction on the ETC of the particulate composites numerically. Moreover, for comparison, we have also investigated the 3D micromechanical models based on the conventional spatial distribution of the inclusions, namely, cubical arrangement, hexagonal arrangement, and random distribution. It is emphasized here that there is no study available that explicitly analyze how these conventional 3D micromechanical models will behave at higher volume fractions and the higher mismatch between particle and matrix thermal conductivity. Previously developed micromechanical model and analytical model by the authors for predicting the ETC of two-phase composite were also considered. The comparisons of the proposed models with the experimental data were performed and analyzed.

2. Details of the proposed percolation model

Agari and Uno [3] demonstrated that with the increase of volume fraction of particles, the chain of the highly conductive particles, i.e., high conductivity percolation paths are formed that drastically increase the ETC of the composite. Percolation is a phenomenon in which the highly conductive particles distributed randomly in the matrix form conductive chains. The inspiration for the geometry of the proposed models comes from the experimental work of Agari and Uno [3] and the percolation theory that is widely used in electrical engineering. We believe that it is worth investigating the role of high conductivity percolation path on the ETC of the particulate composite. The information on the three-dimensional spatial distributions of inclusion are usually not known for the experimental data considered in this study, and generally, the actual microstructure of composite is usually not available in the literature. Therefore, we created

percolation models to study the conductive chain mechanism in the particulate composites. The 2D Illustration of the typical percolation model considered in this study is shown in Figure 1. The figure clearly shows that the particles (in red) touching each other participate in percolation and accelerate the heat conduction while particles (in blue) which do not form a conductive chain do not participate in percolation.

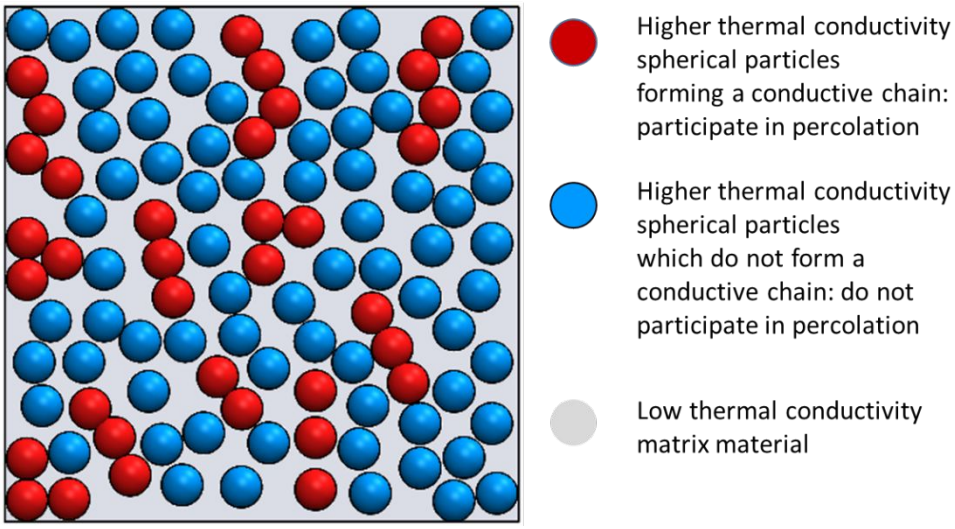


Figure 1: Percolation model showing particles forming a conductive chain and participating in percolation paths, and randomly distributed particles in low thermal conductivity matrix.

3. Modeling Approach

Three different modeling approaches are considered, namely, finite element homogenization, micromechanical homogenization, and analytical modeling.

3.1. Finite element homogenization

3.1.1. Detailed Micromechanical models

The thermal conductivity of the particulate composite depends on the microstructural feature of the composite such as the shape, size distribution, spatial distribution, and orientation distribution of the reinforcing inclusions in the matrix [10]. Mostly, real composites possess inclusions with random distributions. The periodic arrangement of the inclusions is usually considered to simplify

the problem and acquire insight into the effect of microstructure on the effective properties [39]. For example, a particulate composite with a 3D periodic array of particles, an RVE (unit cell) shown in Figure 2(a) and (b) is sufficient to conclude the whole composite [40]. These RVEs are used to study the ETC of particulate composites representing the dilute effects of distribution. On the other hand, RVE with a random distribution of particles is also studied by several researchers [29],[41]. However, the recent experimental studies show that effective thermal properties of the particulate composites increase drastically with highly conductive inclusions in a matrix due to the formation of the conductive chain of particles. The effect of highly conductive inclusion has not been investigated in the literature. In this study, four types of the microstructural arrangements, namely, 3D cubical arrangement, hexagonal arrangement, random distribution, and random network percolation models are considered to investigate their ETC when the ratio of thermal conductivity of particle to the matrix is very high. Figure 2 shows the RVE of these different microstructural arrangements.

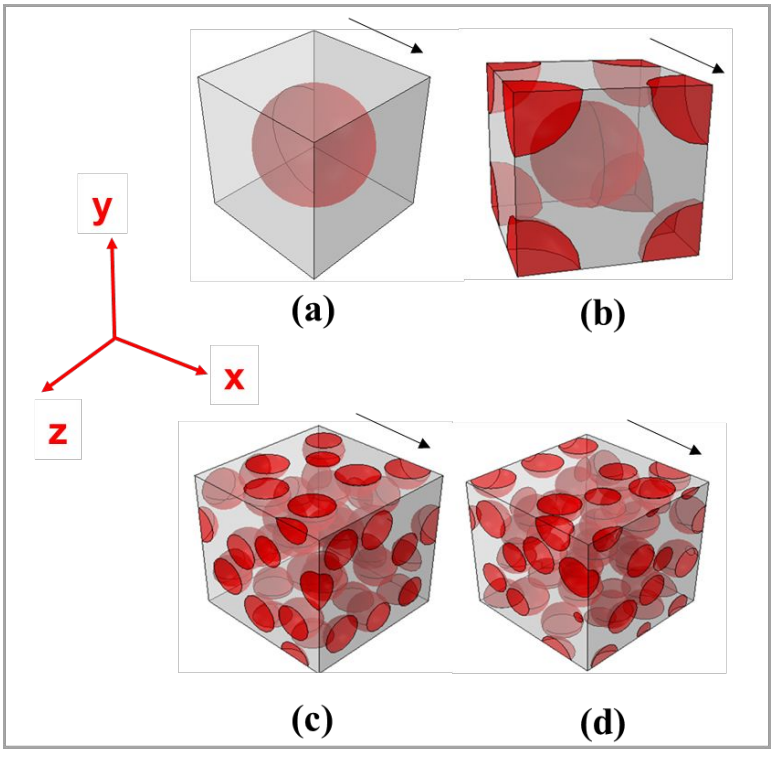


Figure 2 Examples of particulate composite RVE considering detailed microstructural arrangements of particles at volume fraction 30%, (a) Cubical arrangement. (b) Hexagonal arrangement, (c) Random distribution, (d) Random network percolation model. The arrow represents the heat transfer direction.

3.1.2. Governing Equation and Constitutive relations

For a continuum body with volume, and surface, , the thermal equilibrium governing equation for the temperature field can be defined as [42]:

$$q_{i,i} = 0 \tag{1}$$

with temperature boundary conditions $\theta = \overline{\theta}$ on Γ_θ and/or heat flux $h_i = q_i n_i = \overline{h}_i$ on Γ_h. Where $\theta = T - T_0$ is temperature change, T is the absolute current temperature, T_0 is an absolute reference temperature, h is the normal heat flux, is the outward vector which is normal to the boundary $\Gamma = \Gamma_\theta \cup \Gamma_h$; $\overline{h}$ and $\overline{\theta}$ are macroscopic heat flux and temperature on the related boundaries.

Microscopic Constitutive Model

The matrix and particle are assumed to be locally isotropic and homogeneous. The thermal constitutive law that governs each material or phase in a RVE is given by the Fourier's law of heat conduction

$$q_i = -K_{ij}\varphi_j \text{ , where } \varphi_j = \frac{\partial \theta}{\partial x_j} \tag{2}$$

Where, q_i, φ_j and K_{ij} are the components of the heat flux vector, temperature gradient vectors and the consistent tangent thermal conductivity tensor.

Macroscopic Constitutive Model

The macroscopic constitutive relation is obtained by solving the heat conduction problem on heterogeneous RVE with specified boundary conditions. Based on the imposed boundary conditions, either the macroscopic heat flux or the macroscopic temperature gradient field is calculated by averaging or homogenizing the microscopic counterparts. The effective thermal conductivity is calculated by using the macroscopic constitutive relationships relating the average heat flux ($\overline{q_i}$), and average temperature gradient ($\overline{\varphi_j}$), and is expressed by the Fourier law of heat conduction as:

$$\overline{q_i} = -\overline{K_{ij}}\,\overline{\varphi_j} \text{ , where } \overline{\varphi_j} = \frac{\partial \overline{\theta}}{\partial x_j} \tag{3}$$

Where $\overline{K_{ij}}$ are the components of the effective thermal conductivity tensor.

3.1.3. Finite Element Models

Finite element models for four types of micromechanical arrangements as shown in Figure 2 were generated with a different volume fraction of the particles ranging from 0-50%. The commercial finite element analysis software ABAQUS was used to carry out the heat transfer analysis. RVE's were considered to have highly conductive particles embedded in a low thermal conductivity matrix. The thermal conductivity of the matrix and particles used in this study are given in Table 1. Figure 3 (a) shows the example of the random composite having the macroscopic scale (L) and microscopic scale (l). An example of RVE having a random distribution of particles at 30% volume fractions is shown in Figure 3(b). Meshed RVE with 10-noded quadratic heat transfer elements (DC3D10) showing 6 boundary faces with respect to the axes directions is also shown in Figure 3(c). Each node in the FE model has one degree of freedom of temperature (T).

The following assumptions were made in creating the finite element models [43]:

(1) The characteristic size of the heterogeneities is assumed to be much smaller than the dimension of an RVE, which in turn is supposed to be small compared to the characteristic length of the macroscopic structure.

(2) Both the matrix and the particle phases are homogeneous and isotropic.

(3) Perfect bonding between the particle and matrix with negligible thermal contact resistance.

(4) No voids in the matrix and particle.

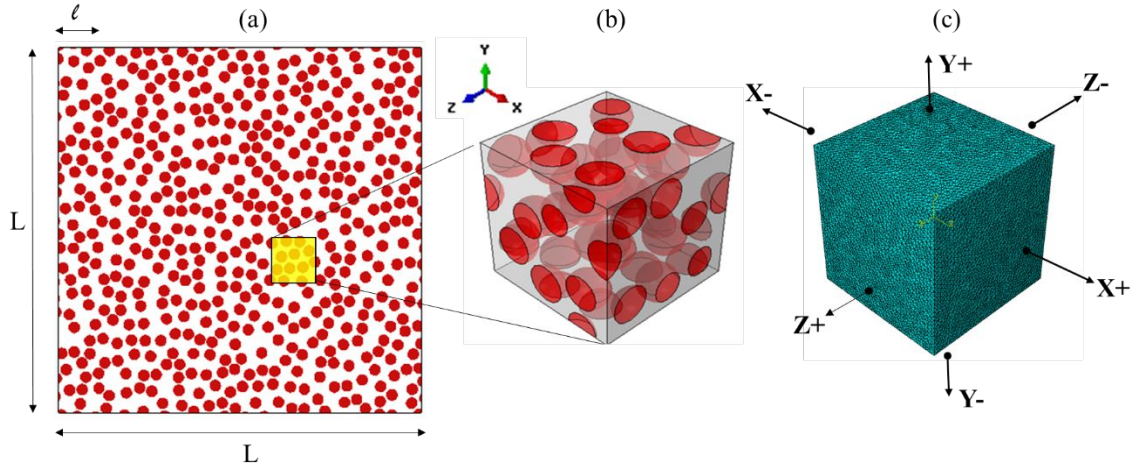


Figure 3 (a) Random distribution of particle (b) RVE (c) Meshed RVE with 10-noded quadratic heat transfer tetrahedron elements (DC3D10) showing 6 boundary faces with respect to the axes directions.

Table 1. Thermal conductivity of materials ([3], [4], [7], [5], [44], [6]).

Material	Thermal Conductivity (Wm^{-1} K^{-1})	Material	Thermal Conductivity (Wm^{-1} K^{-1})
Polystyrene	0.1549	Silica	1.5
Epoxy	0.195	Alumina	36
High density polyethylene (HDPE)	0.543	Aluminum	204
Polyvinyl chloride	0.1687	Graphite	209.3
CaO (calcium oxide)	15.07	SCAN	220
MgO (magnesium oxide)	54.85		

3.1.4. Boundary conditions

The effective thermal properties of the heterogeneous materials can be determined by applying four types of boundary conditions [45],[46], to the RVE. The four boundary conditions are listed here as follows:

1. Natural Boundary Condition (NBC)

$$h = \overline{q}_i n_i, \qquad \langle q_i \rangle = \overline{q}_i, \qquad \forall \mathrm{x}_i \in \partial B, \tag{4}$$

2. Essential Boundary Condition (EBC)

$$\theta = \overline{\varphi}_i x_i, \qquad \langle \varphi_i \rangle = \overline{\varphi}_i, \qquad \forall \mathrm{x}_i \in \partial B, \tag{5}$$

3. Periodic Boundary Conditions (PBC)

$$\begin{aligned} &\theta(\mathrm{x}_i + L_i) = \theta(\mathrm{x}_i) + \overline{\varphi}_i (x_i^+ - x_i^-), \\ &h_i(\mathrm{x}_i + L_i) = -h_i(\mathrm{x}_i) \qquad \forall \mathrm{x}_i \in \partial B, \end{aligned} \tag{6}$$

So that $\langle q_i \rangle = \overline{q}_i$, where $\overline{q}_i$ and $\overline{\varphi}_i$ are constant vectors.

4. Mixed Boundary Condition (MBC)

$$(\theta - \overline{\varphi}_i x_i)(h - \overline{q}_i n_i) = 0, \qquad \forall \mathrm{x}_i \in \partial B, \tag{7}$$

Where, ∂B is the surface boundary of the RVE, n_i is the outer unit normal vector to ∂B, and L is the length of the periodicity. Generally, for the case of PBC and EBC, $\overline{\varphi}_i$ is applied; while $\overline{q}_i$ is applied for NBC. In the case of MBC, EBC is applied on one pair of parallel faces, and NBC is applied on the other pairs. Previous investigations have found that the MBC and PBC are much more accurate in the micromechanical analysis of composite materials for both periodic materials and random materials ([47]). Moreover, MBC and PBC yield RVE size independent results while the results obtained from EBC and NBC converge to those of MBC and PBC as l increases as shown by [48]. Since cubical and hexagonal arrangements are periodic so PBC can be applied;

while MBC should be used for random and percolation models. This study is also utilized MBC to evaluate effective thermal conductivity due to scale-independence.

3.1.5. Homogenization Method

From the above homogenization procedures, we can see that the flux and temperature on the boundary of the RVE are sufficient to calculate the effective thermal conductivity of the composites. The macroscopic heat flux and the macroscopic temperature gradient fields were computed as the volume averages of the microscopic counterparts, and they were related to each other by the macroscopic constitutive formulations ([49]).

$$\overline{q_i} = \langle q_i \rangle = \frac{1}{V}\int_V q_i dV = \frac{1}{V}\int_\Gamma h x_i d\Gamma \tag{8}$$

$$\overline{\varphi_i} = \langle \varphi \rangle = \frac{1}{V}\int_V \nabla\varphi dV = \frac{1}{V}\int_\Gamma \theta n_i d\Gamma \tag{9}$$

Where V is the volume of the RVE. It can be seen from Eq. 8 and 9 that the volume average heat flux and temperature gradient are related to the flux on the boundary of the RVE.

For isotropic case, the ETC tensor can be expressed as $\overline{K_{ij}} = \overline{k}\delta_{ij}$. Thus according to macroscopic Fourier's law, the ETC can be calculated as $\overline{k} = \|\overline{q_i}\| / \|\overline{\varphi_i}\|$. To eliminate the rate effects and external influence, all RVE heat transfer simulation should be carried out under steady-state heat conduction.

It is worth mentioning that for highly conductive inclusions, increasing the volume fraction of the inclusion would cause acceleration in the diffusion process, which cannot be captured by a Fourier's law of heat conduction presented in Eq. (1) [50]. However, if one neglects size dependent conduction and assumes steady state conditions, then different diffusion equations will yield the same effective thermal conductivity for the composite [47].

3.2. Micro-thermal homogenization

Khan and Muliana (2010) developed a micromechanical model for the effective thermal properties of a particle reinforced composite. In this study, we employed the same model for computing the effective thermal conductivity of the composite containing highly conductive inclusion in a less thermally conductive matrix and analyze its suitability. For brevity, we have reviewed the basic equations describing the ETC formulation. Figure 4 illustrates the simplified micromechanical model (RVE) for the particulate composite. In the model, a microstructure with the cubical arrangement of cubic particles in a homogeneous matrix was assumed. A representative volume element (RVE) with a cubic particle embedded in the center of the matrix with cubic domain. A one-eight unit-cell consisting of four sub-cells was modeled due to symmetry. The first sub-cell was a particle constituent, while subcells 2, 3, and 4 were representing the matrix constituents. The micromechanical relations gave equivalent homogeneous thermal responses from the heterogeneous microstructures and simultaneously recognize thermal constitutive behaviors of the individual constituents. The micromechanical formulations were designed to be compatible with commercial finite element analyses software, i.e., ABAQUS [1]. In ABAQUS, the effective responses from the micromechanical relations were implemented at each material point (Gaussian integration point) within the finite elements as shown in Figure 4.

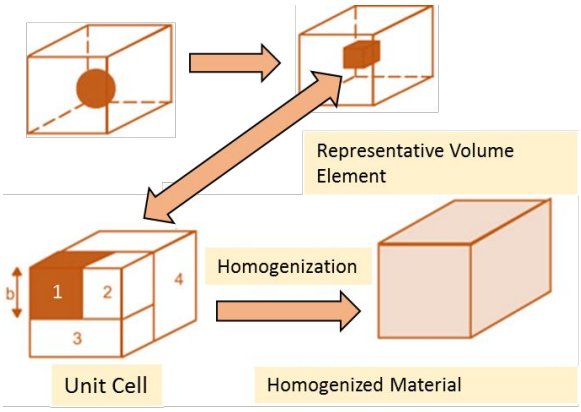


Figure 4 Representative unit-cell model for the particulate composite with cubic particle embedded in a matrix.

The micromechanical model was formulated by considering an RVE with a single inclusion embedded in a cubic matrix. Periodic boundary conditions were imposed on the selected RVE model. A volume averaging method based on a spatial variation of the temperature gradient in each subcell was adopted to determine the effective thermal conductivity of the particle reinforced composites. The average heat flux and temperature gradient are shown here:

$$\bar{q}_i = \frac{1}{V}\sum_{m=1}^{N} \int_{V^{(m)}} q_i^{(m)}(x_k^{(m)})dV^{(m)} \approx \frac{1}{V}\sum_{m=1}^{N} V^{(m)} q_i^{(m)} \tag{10}$$

$$\bar{\varphi}_i = \frac{1}{V}\sum_{m=1}^{N} \int_{V^{(m)}} \varphi_i^{(m)}(x_k^{(m)})dV^{(m)} \approx \frac{1}{V}\sum_{m=1}^{N} V^{(m)} \varphi_i^{(m)} \tag{11}$$

Where N is the total number of sub-cells. The average heat flux within an FE scheme was solved numerically, and an incremental approach was used to obtain the ETC expression. The incremental average heat flux can be expressed as:

$$d\bar{q}_i = -\bar{K}_{ij} d\bar{\varphi}_j \tag{12}$$

The homogenized temperature gradient and heat flux relations are summarized as follows:

$$d\bar{\varphi}_i = \frac{1}{V^{(A)}}\left[V^{(1)} d\varphi_i^{(1)} + V^{(2)} d\varphi_i^{(2)}\right] = d\varphi_i^{(3)} = d\varphi_i^{(4)} \tag{13}$$

$$d\bar{q}_i = \frac{1}{V}\left[V^{(A)} dq_i^{(A)} + V^{(3)} dq_i^{(3)} + V^{(4)} dq_i^{(4)}\right] \tag{14}$$

$$dq_i^{(A)} = dq_i^{(1)} = dq_i^{(2)} \tag{15}$$

Where, the total volume of the subcells 1 and 2 in Eqs. (13) and (14) is $V^{(A)} = V^{(1)} + V^{(2)}$.

Let $\boldsymbol{M}^{(m)}$ be the concentration tensor that relates the average temperature gradient of each subcell with the overall temperature gradient across the unit cell. The temperature gradient in each subcell is given by:

$$d\varphi_i^{(m)} = M_{ij}^{(m)} d\bar{\varphi}_j \tag{16}$$

and the incremental form of the heat flux in each subcell is expressed as:

$$dq_i^{(m)} = -K_{ij}^{(m)} M_{jk}^{(m)} d\bar{\varphi}_k \tag{17}$$

Using Eq. (17), the average incremental heat flux in the unit-cell model is approximated as:

$$d\bar{q}_i = -\frac{1}{V}\sum_{m=1}^{4} V^{(m)} K_{ij}^{(m)} M_{jk}^{(m)} d\bar{\varphi}_k \tag{18}$$

Comparing the above equation with Eq. (12) gives the tangent effective thermal conductivity matrix of the composite, which is:

$$\overline{K}_{ik} = -\frac{1}{V}\sum_{m=1}^{4} V^{(m)} K_{ij}^{(m)} M_{jk}^{(m)} \tag{19}$$

Detail expression of the $\boldsymbol{M}^{(m)}$ matrix can be found elsewhere [1].

3.3. Analytical Model

Khan et al. [51] proposed an analytical model for ETC of an isotropic two-phase composite material consists of a highly conductive particle as inclusions embedded in a low thermally conductive polymeric matrix. An ideal contact between the inclusions and the matrix was assumed with no porosity in the composite. The ETC expression was derived assuming a unidirectional heat flow, neglecting thermal convection, radiation, and the contact resistance between the matrix and the inclusions. The underlying assumption and procedure to obtain the ETC expression is summarized below.

A unit cube (1x1x1) of a two-phase composite material was assumed to have inclusions dispersed in a matrix with some statistical spatial distribution as shown in Figure 5 (a). Some form of continuous distribution of inclusions was usually assumed to determine the analytical expression for the ETC of a composite [52]. The outer surfaces of a cube parallel to xy and xz planes were perfectly insulated, i.e., the direction of heat transfer is along the x-axis. The two-

phase composite was hypothetically sliced into numerous thin layers parallel to the yz plane as shown by the vertical lines in Figure 5 (b). It was assumed that the driving potential (temperature gradient) for heat conduction in x-direction was uniform through each layer. Each composite layer had the inclusions and the matrix fractions as shown in Figure 5 (c). Without changing the ETC of each layer, both inclusions and matrix can equivalently be represented by histograms of matrix and inclusions to compute the effective resistance of each slice which in turn gives the effective resistance (ETC) of a unit cell. Alternatively, the sequence of the layers can be re-arranged into a continuous distribution function of the inclusions to compute the effective resistance of the unit cell Figure 5 (d). The model obtained was geometrically invariant along the z-axis.

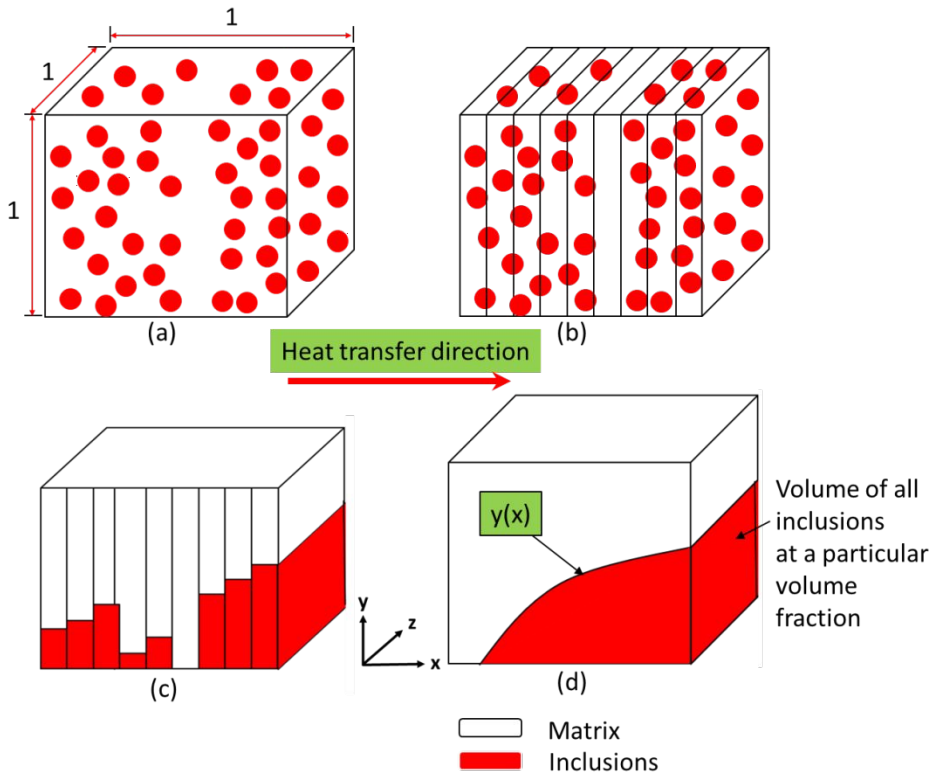


Figure 5. A cubic unit cell for the study of the ETC of a two-phase composite material (after [28]). (a) Inclusions randomly distributed in the matrix, (b) hypothetically sliced thin layers, (c) histogram of the layers and (d) corresponding equivalent continuous distribution of the histogram.

The unidirectional effective heat flux ($\overline{q}_x$) in a unit cube can be expressed as:

$$\bar{q}_x = -k_e \frac{\partial \bar{T}}{\partial x_x} \tag{20}$$

Where, is the ETC of a composite and $\partial \bar{T} / \partial x_x$ is the uniform temperature gradient through each layer. For a unit length along the x-axis, the expression for effective resistance ( ) of a unit cell is given by [52]

$$\mathrm{R_e} = \int_0^1 \frac{dx}{k_m + (k_p - k_m)\,\mathrm{y(x)}} + \frac{1-2x}{k_m}; \qquad k_e = \frac{1}{R_e} \tag{21}$$

The is a function that describes the variation in the collective volume of the inclusions (that may vary differently for different distributions) from one V_f to the other V_f of the inclusions. The volume under the surface was formed by the curve projection on xy-plane represents the volume of all inclusions at a particular volume fraction. The ETC ( ) was obtained from Eq. (21) by taking the inverse of the equivalent overall resistance of the unit cell ($\mathrm{R_e}$) obtained for different distributions. These calculations were only possible if one assumed that the driving potential (temperature gradient) for heat conduction along x-direction was uniform through each layer.

Khan et al. [51] assumed that the sequence of the layers shown in Figure 5(c) can be represented by a linear distribution function of the inclusion material and obtained the following expression for the ETC of a two-phase composite as a function of V_f and the thermal conductivity of the constituents:

$$k_e = \left[\begin{array}{l} \dfrac{1}{(k_p - k_m)} \ln\left\{ k_m + \sqrt{2V_f}\,(k_p - k_m) + \sqrt{2V_f}\,(k_p - k_m) \right\} \\ \quad - \dfrac{1}{(k_p - k_m)} \ln\left\{ k_m + \sqrt{2V_f}\,(k_p - k_m) \right\} + \dfrac{1 - \sqrt{2V_f}}{k_m} \end{array} \right]^{-1} \tag{22}$$

4.2 *Comparison with Other Models*

4.2.1 Analytical Models

To check the efficacy of the proposed model, the predictions were also compared with other models available in the literature. For brevity, we considered classical models relevant to our study. Maxwell [13] investigated the conduction of two-phase composites analytically by considering the dilute suspension of non-interacting spherical particles. An exact expression for the ETC of composites was obtained by solving the Laplace equation. Jeffrey [53] extended the Maxwell model and considered the interaction between the pairs of spheres to determine the expression of ETC of composite and also introduced a parameter that accounts for the role of Kp/Km ratio on the ETC. The results were also found to be dependent on how pairs of spheres are distributed with respect to each other. The ETC expressions for these models are written as follows:

$$\text{Maxwell [13]: } k_e = k_m \frac{k_p + 2k_m + 2V_f\left(k_p - k_m\right)}{k_p + 2k_m - V_f\left(k_p - k_m\right)} \tag{23}$$

$$\text{Jeffrey [53]: } k_e = k_m + 3k_m V_f \left[1 + V_f \frac{\sigma_1(2k_m + k_p) + \left(k_p - k_m\right)}{2k_m + k_p}\right] \frac{\left(k_p - k_m\right)}{2k_m + k_p} \tag{24}$$

σ_1 is parameter depending on the thermal conductivity ratio.

4.2.2 Numerical Models

Several numerical models are also available to determine the ETC of particulate composites. For example, ETC of random two-phase composite materials was obtained by considering the shape, spatial distribution, thermal contact resistance, and particles V_f ([23],[54],[39] and references therein). Experimental and numerical ETC of polymer matrix filled with metallic spheres were presented by Karki et al. ([41]). The effects of the filler concentrations, the ratio of thermal conductivities of filler to the matrix material and the Kapitza resistance of the contact

inclusion/matrix on the ETC were investigated. For more details and updated review/references on the numerical modeling of the ETC of particulate composites, please see [41]. To the best knowledge of the authors, there is only one model available in the literature that considered the effect of the embedding highly conductive inclusions in a less thermally conductive matrix on the ETC of particulate composites. Zhang et al. [34] proposed a randomly mixed model to compute the ETC of particulate composites numerically with respect to the V_f of the particles and the ratio of the thermal conductivity of the particle to that of the matrix. The cubic shape particles of uniform size were generated randomly using a computer program. The steady state heat equation was solved by the finite difference method directly for the composite with appropriate boundary conditions.

4. Results and Discussion

Results obtained from the proposed finite element homogenization, analytical solutions, and micromechanical models were compared with already published experimental data and other models in literature which were developed explicitly for highly conductive particulate composites.

We investigated the effect of conductivity mismatch on the ETC of the particulate composites. The effect of the different distribution of the inclusions were analyzed, and four RVE of proposed micromechanical models with detailed microstructure were considered. Figure 6 shows how the ETC of composite increases with respect to the matrix conductivity with the increase of the conductivity mismatch (Kp/Km ratio). It was found that one particle model showed the lowest increase in the overall thermal conductivity while the percolation model showed the highest values. At lower volume fraction there was not a significant difference in the increase of the ETC for all models. However, as the volume fraction was increased beyond 0.3, then the ETC of the percolation models was increased with respect to the matrix material conductivity. As per the

percolation theory [36], after V_{fc} beyond 0.3, the particles were increased, and the formations of the conductive chains thus played a dominant role in the increase of ETC.

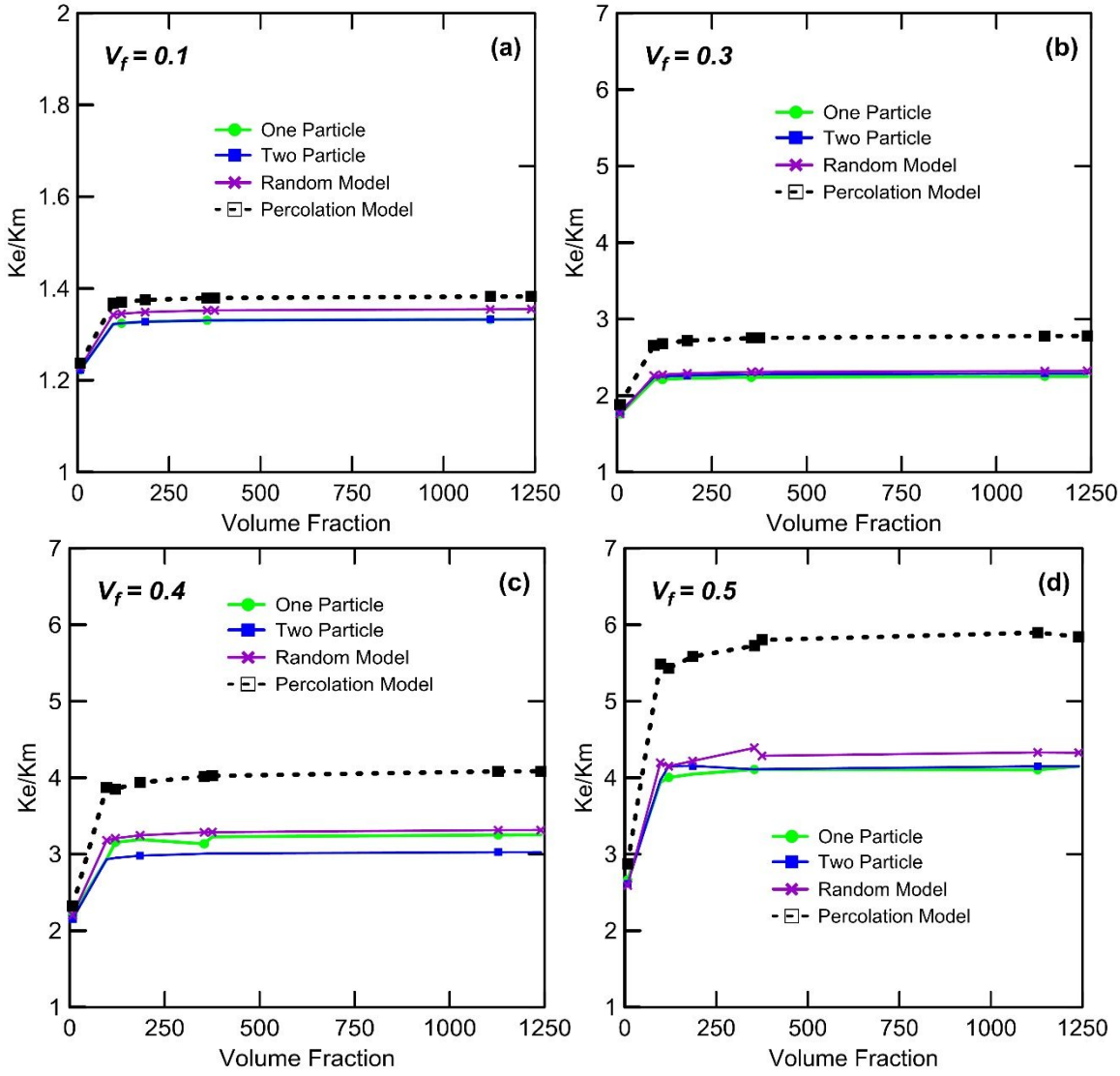
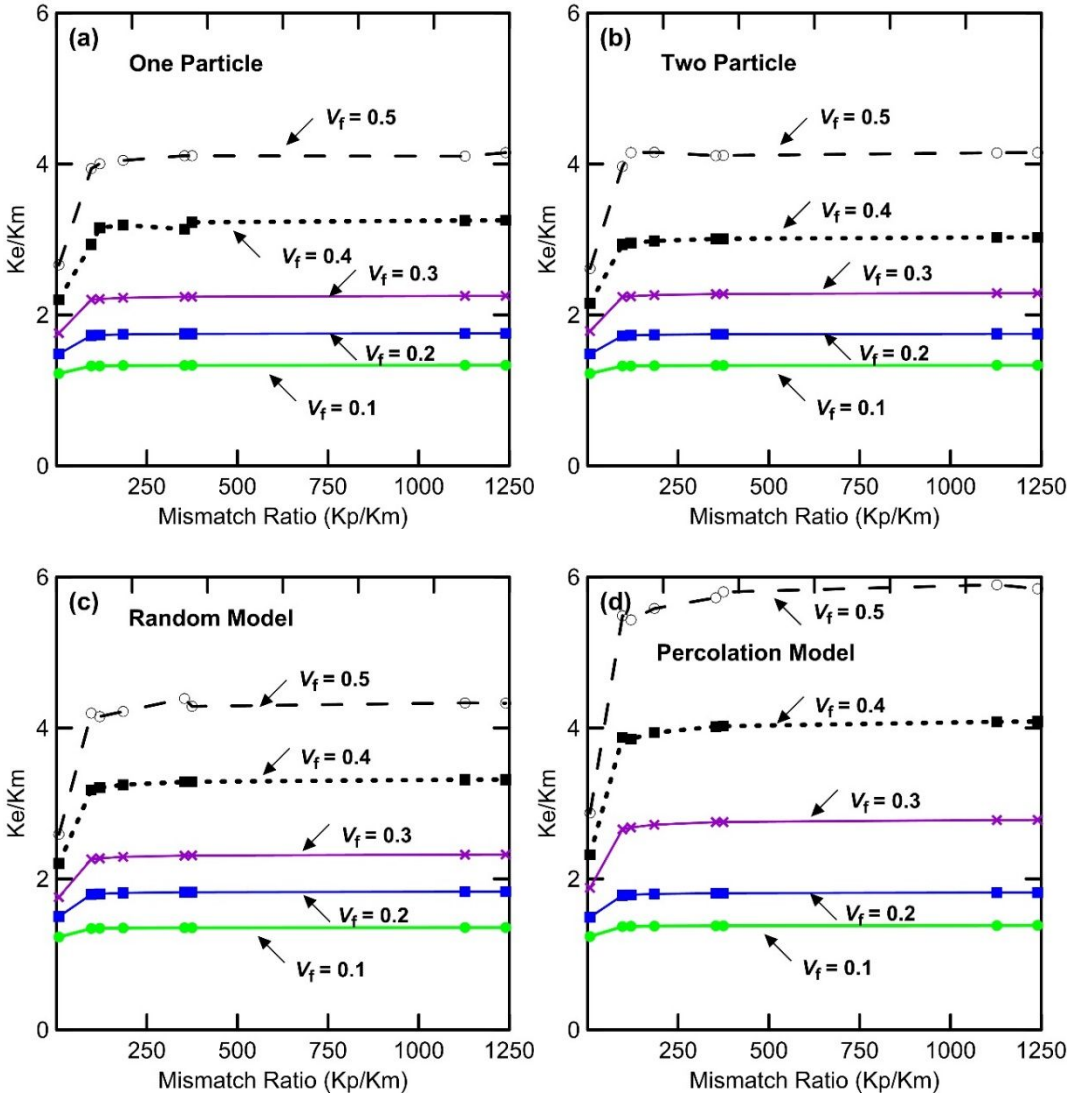


Figure 6. Conductivity mismatch in a two-phase composite material and its effect on the ETC for various micromechanical models..

For completeness, we studied the conductivity mismatch effect on the ETC for all proposed models. Figure 7 shows the result of conductivity mismatch at different volume fraction for each model. It was found that among all the models considered the linear model produced the highest increase in the ETC with respect to the matrix material thermal conductivity. The simplified micromechanical model showed the lowest increase in the ETC. One particle, two particles, and

random particle model showed a similar trend in the increase of the ETC with respect to the matrix material.



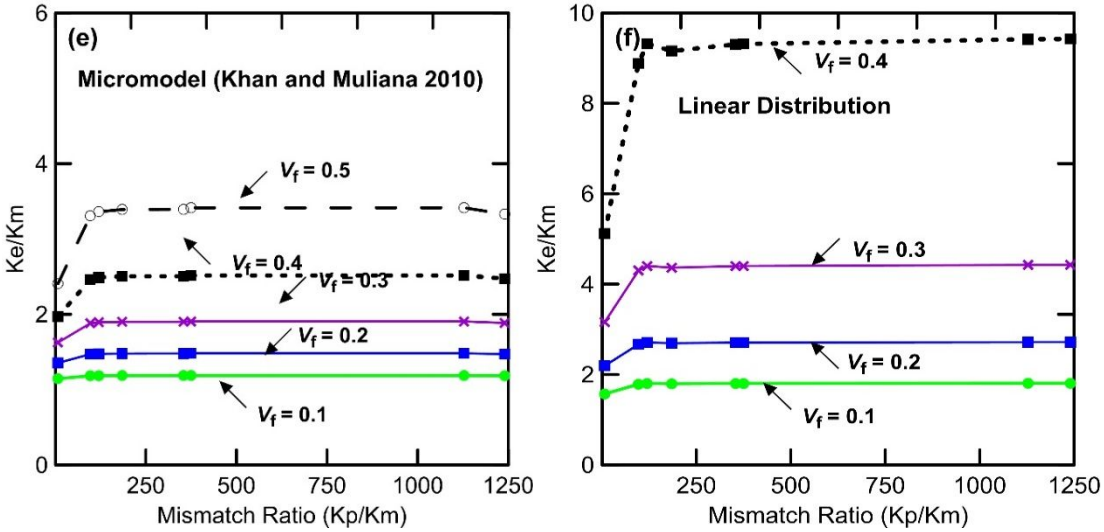


Figure 7. Conductivity mismatch in a two-phase composite material and its effect on the ETC for various micromechanical models.

Figure 8 was used to analyze the percent increase in the ETC that was obtained with respect to the matrix materials. Again the maximum ETC was predicted using the linear model. As a comparison, we showed the results at two volume fraction for all models. We observed that at V_f =0.1, almost all the micromechanical models yielded very low values except the linear model which gave nearly 80% higher ETC values than the matrix conductivity. While at V_f =0.3, the linear model provided 350% higher than the matrix conductivity as compared to the percolation model which yielded 180% higher than the matrix conductivity.

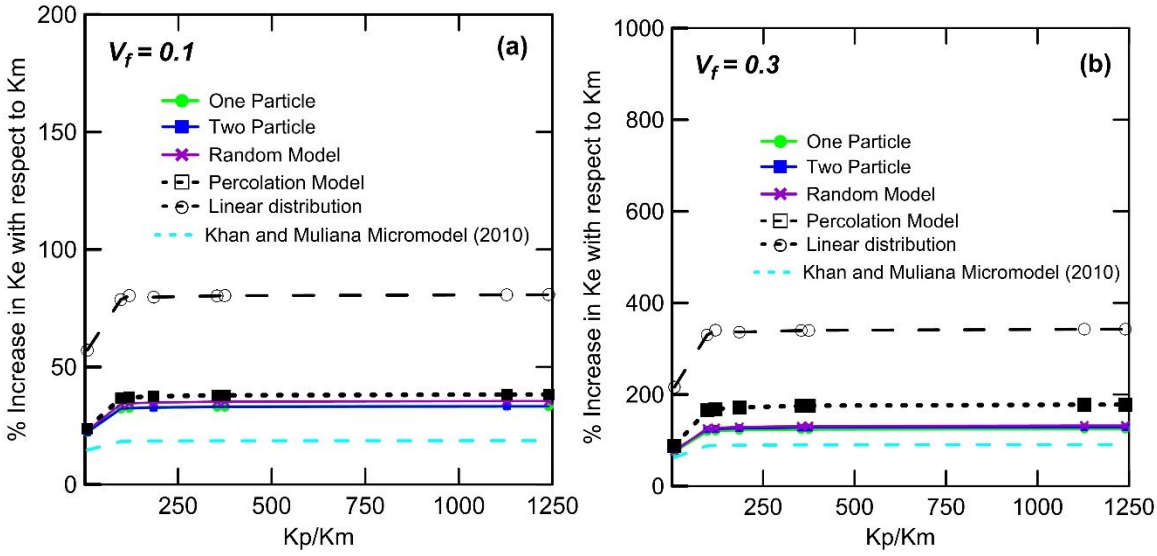


Figure 8. Conductivity mismatch in a two-phase composite material and its effect on the ETC for various micromechanical models.

4.1 Comparison with Experimental Data

The ETC of epoxy filled with silica (Kp/Km=7.7), alumina (Kp/Km=185) and silica-coated aluminum nitride (SCAN, Kp/Km=1128) particulates was experimentally studied by Wong and Bolampally [7]. The average particle size of the fillers used was 12–15 microns. The thermal conductivities of all materials used in this study are given in Table 1. Figure 9 showed the comparisons of the experimental and predicted ETC for ratio Kp/Km=7.7. For all Kp/Km, the models observed with reasonable estimates but linear and Jeffrey models overestimated the ETC.

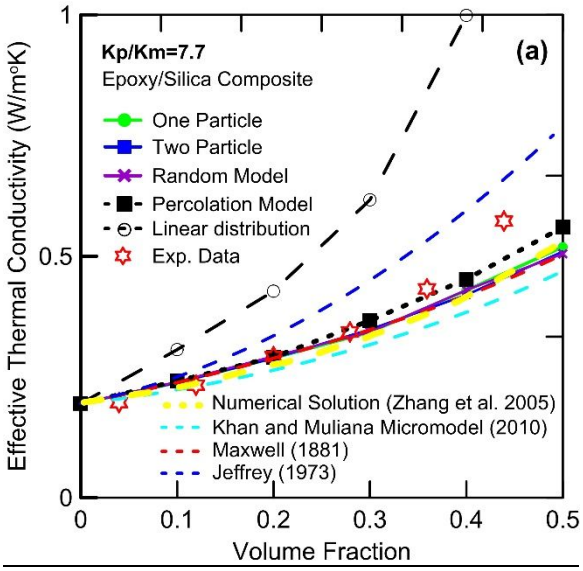


Figure 9. Conductivity mismatch in a two-phase composite material and its effect on the ETC for various micromechanical models.

The comparison of the experimental, the predicted ETC of epoxy filled with alumina (Kp/Km=185) and silica-coated aluminum nitride (SCAN, Kp/Km=1128) particulates with different models are shown in Figure 10. For, Kp/Km=185, the percolation model and Jeffrey model were observed with reasonable estimation. Maxwell model, micromechanical model, one particle, two particles, and random model were found with reasonable estimates until V_f <20%. They showed an underestimation of the ETC at higher V_f. The linear model and numerical solution overestimated the ETC, but after V_f >30% both models showed a similar trend of increase in the ETC with the increase of V_f. For Kp/Km=1128, all the models deviated from the experimental data after V_f >20%; while linear distribution gave acceptable estimates. The numerical model was observed with a reasonable estimate until V_f <30%, and after that, the results have deviated significantly.

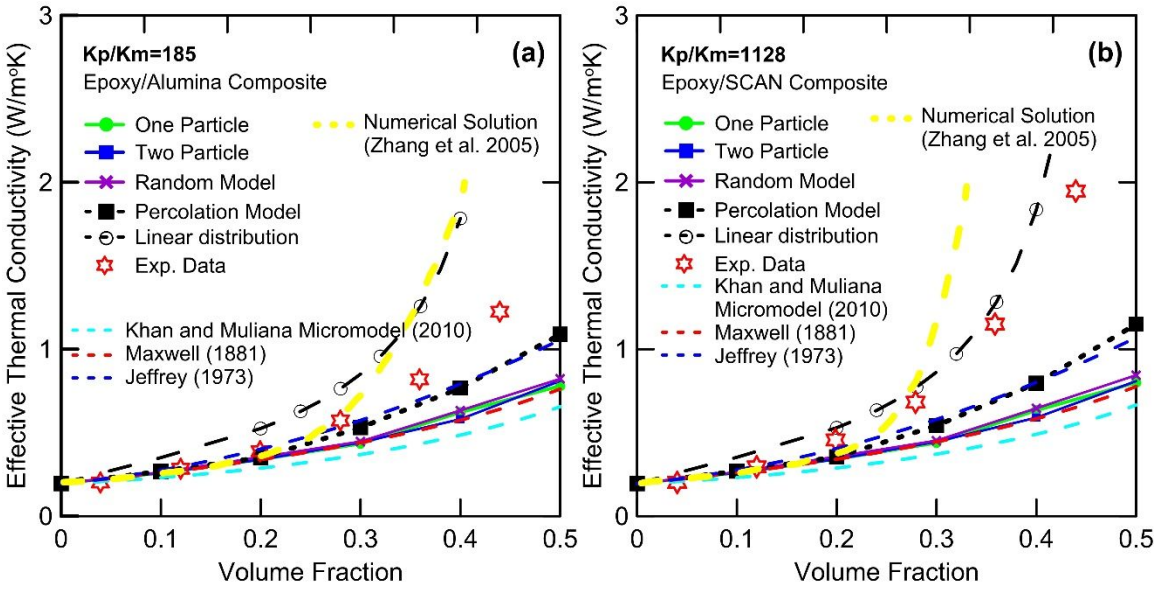


Figure 10. Comparison of the experimantal data and predicted ETC for a) Epoxy/Alumina b) Epoxy/silica coated aluminum nitride composites.

Comparison of the experimental data and the predicted ETC of polystyrene filled with CaO (Kp/Km=97) and MgO (Kp/Km=354) particulates are presented in Figure 11. The experimental data were obtained by Sundstrom and Lee [5]. The calcium oxide and magnesium oxide were powders with particles approximately spherical in shape and particle size in the range of 62-125 microns. For both cases, the Jeffrey model observed with the best estimates of the ETC. Both the numerical model and percolation model were providing a consistent trend of increase in the ETC with the increase of V_f. Maxwell model, micromechanical model, one particle, two particles, and random model were observed with reasonable estimates until V_f <10%; but they were providing underestimated values of the ETC at higher V_f. In both cases, the linear model overestimated the ETC as compared to all the other models.

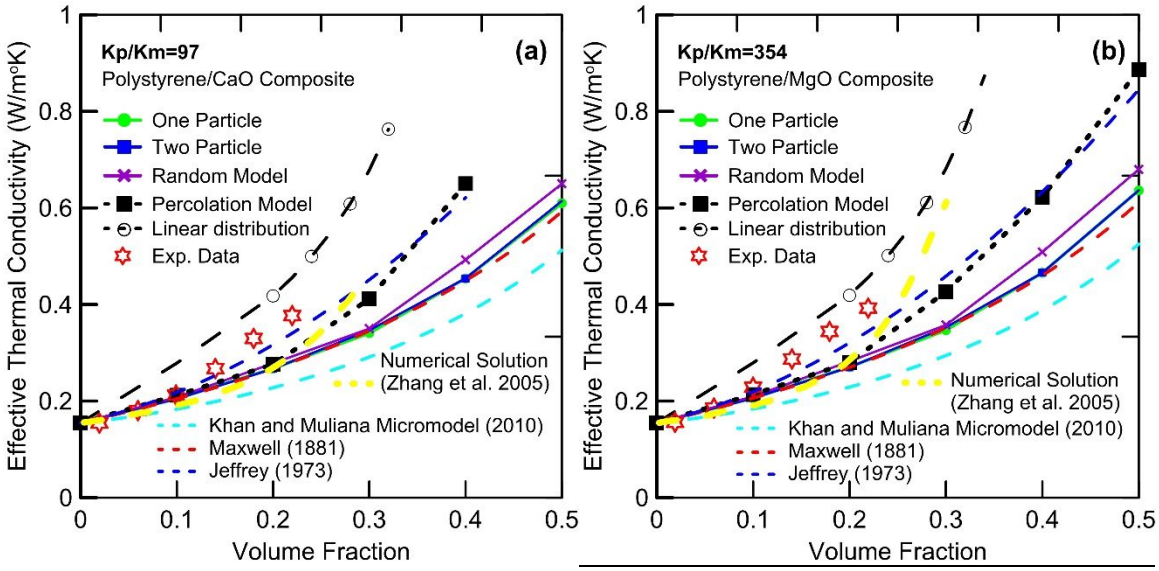


Figure 11. Comparison of the experimantal data and predicted ETC for a) Polystyrene/CaO b) Polystyrene/MgO composites.

Kumlutas et al. [44] and Tavman [6] experimentally studied the ETC of high-density polyethylene filled with tin (Kp/Km=120) and aluminum (Kp/Km=375) inclusions. The metallic fillers of tin and aluminum used in the form of fine powder with particles approximately spherical in shape and particle size in the range of 20-40 and 40–80 microns, respectively. The comparison of the experimental data and the predicted ETC with different models are shown in Figure 12

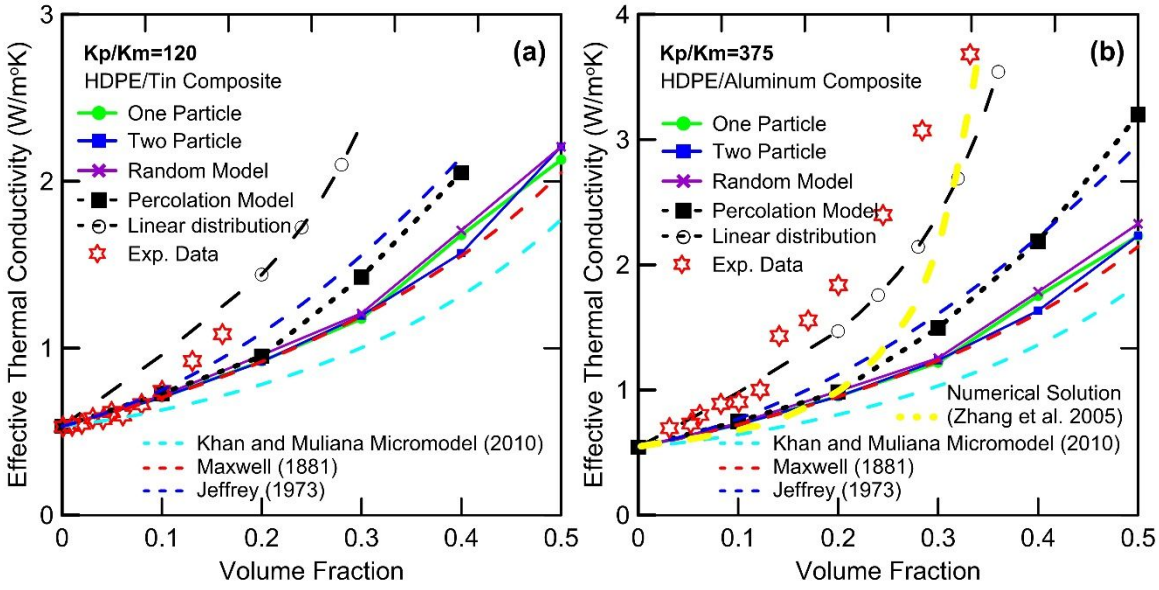


Figure 12. For Kp/Km=120, Jeffrey and percolation model were with the best estimates and provided trends in the increase of ETC. All other models underestimated the predictions except the linear model. However, for Kp/Km=375, linear distribution was observed with quite reasonable estimates of the trend in the increase of ETC with the increase of V_f of the inclusions. However, all other models underestimated the ETC at this ratio.

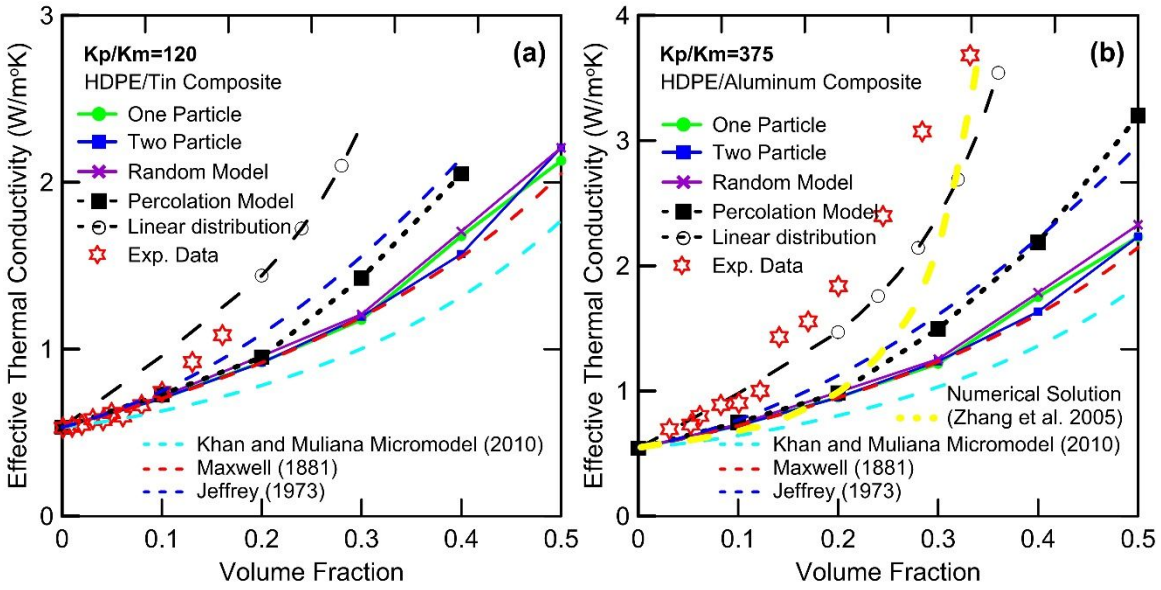


Figure 12. Comparison of the experimantal data and predicted ETC for a) HDPE/Aluminum b) HDPE/Tin composites.

Agari and Uno ([3], [4]) studied Graphite based composites and observed that at higher V_f the graphite flakes agglomerated and formed the conductive chain and as a result ETC of the composite was considered to increases drastically [24]. The average particle size of the fillers used was 44–149 microns. Figure 13 shows the comparison of the experimental data and the predicted ETC. The linear distribution provided the best estimates. Jeffrey and percolation models provided a reasonable estimate and showed the same tendency as the experimental data. All other models underestimated the ETC. The results covered the commonly used range of the thermal conductivity ratio of matrix to the inclusions, i.e., Kp/Km=10 – 1240.

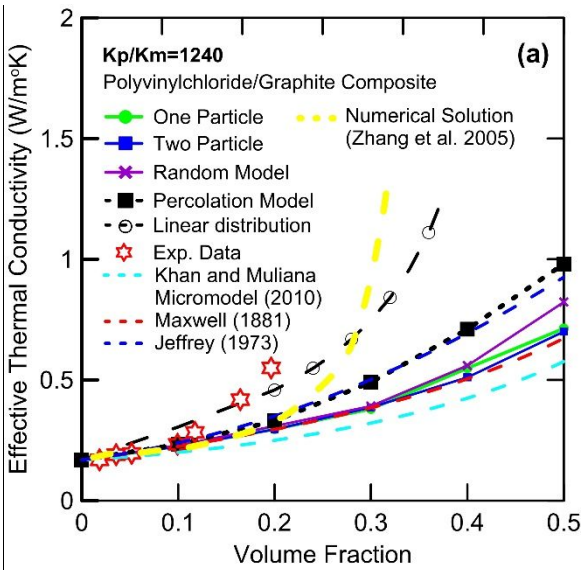


Figure 13. Comparison of the experimantal data and predicted ETC for a) Polyvinylchloride/Graphite b) Polyethylene/Graphite composites.

4.2 Discussion

It can be realized that for all the available experimental data, the trend shows an increase of ETC with the increase of V_f of the inclusions. All analytical models show good predictions of the overall ETC when the V_f is relatively small, but the predictions tend to deviate as the V_f increases. However, in most of the analyzed cases, linear distribution, Jeffrey and Percolation models effectively predict the trend in the increase of ETC with the increase of V_f of the inclusions.

If we recall the assumption of Maxwell's equation: there is no interaction among the particles. Jeffrey [53] extended the Maxwell model and considered the interaction between the pairs of spheres to determine the expression of ETC of the composite. On the other hand, an analytical model is based on the linear distribution of the dispersed particles which effectively considered the interaction between the pairs of spheres to determine the expression for the ETC of a two-phase composite as a function of V_f and the thermal conductivity of the constituents. The micromechanical model computes the ETC based on the unit cell representing a dilute suspension

of non-interacting spherical particles. The possibility of the explicit formation of particle chains is not considered in all these models.

Since we used the Digimat software to create the FE percolation models, so we do not have enough control over the algorithm of particle generation. The software gives us FE models with limited percolation effects, for example, a maximum of 20-30 percolated chains are usually formed depending on the volume fraction. After investigating the location of spheres, it was found that the chain of the highly conductive particles, i.e., high conductivity percolation paths, consists of maximum 4 spheres touching each other (2D Illustration is shown in Figure 1). This limited chain length was found insufficient in getting the enhancement of the ETC, and it was the primary reason for significant differences between estimated and experimental ETC at higher volume fractions and the higher mismatch between particle and matrix thermal conductivity. However, the percolation models were found competitive in describing the sphere interaction mechanism and almost reproduces the results of Jeffrey's model [53].

We observed that the proposed analytical model with linear distribution worked very well for most of the particulate composites. For such highly conductive composites, we recommended that experiments should be performed to find the real 3D spatial distribution and connectivity of the inclusions. We plan to conduct experiments by ourselves to acquire the 3D spatial distribution of the clusters and conductive chains of particles using micro CT to generate real microstructure of particulate composites. Nevertheless, we still believed that the percolation modeling approach could be used to predict the ETC of particulate composites provided several long conductive chains should connect the opposite faces of the matrix, which is not the case in the proposed model due to software limitations. To resolve this issue, we also plan to develop our algorithm to generate

spatially controlled and architected chains of spheres to study their effect on the ETC of the particulate composites.

It should be realized that the discrepancies in predictions in comparison with the experimental data are due to a number of other reasons: 1- The model does not account for the effect of particle shape, 2- size, orientation distribution, interfacial thermal resistance and the effect on particle agglomeration on the enhancement of the ETC of a composite were not considered. For such work, we refer to the work of Nan et al. [24], Prasher et al. [25], Hong et al. [55] and more recently by Wemhoff [31] and Wemhoff & Webb [32].

4. Conclusions

Two micromechanical modeling approaches were presented to predict the ETC of two-phase composites containing inclusions with very high thermal conductivity than the matrix. The first modeling approached introduced four micromechanical models based on the various spatial distribution. Finite element homogenization was performed to compute the ETC. In the second approach, a simplified micromechanical model with one particle embedded in a matrix was considered. The micromechanical model used four particle and matrix subcells. Micromechanical relations were formulated in terms of incremental average heat flux and temperature gradient, in the subcells. The first order homogenization scheme was applied, and ETC was formulated by imposing heat flux and temperature continuity at the subcells' interfaces. The predictions obtained from both modeling approaches were compared with the published experimental data and other models available in the literature. It was found that the random network percolation models gave reasonable estimates and showed a rational trend in the increase of ETC for the high Kp/Km ratio as compared to the ones obtained from the other micromechanical models.

Moreover, it was found that the analytical solution based on the linear distribution of the inclusions provided reasonable estimates with the increase of ETC for the high Kp/Km ratio. We concluded that conductive chain mechanism could be adequately represented by the micromechanical model based on random network percolation models and Jeffrey model in which the inclusions were distributed randomly in a matrix but form limited connections and interaction among inclusions. It was also concluded that with further improvement of the percolation models such as introducing more conductive particle chains and use of smaller size particles with clustering effects may improve the prediction of the ETC.

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2019-04-10
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https://doi.org/10.1177/0021998319843329
Khan K, Hajeri F, Khan M. Analytical and numerical assessment of the effect of highly conductive inclusions distribution on the thermal conductivity of particulate composites. Journal of Composite Materials, Volume 53, Issue 25, 2019, pp. 3499-3514

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