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A numerical investigation on the heat conduction in high filler loading particulate composites

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ABSTRACT

Particle-filled composite materials have been widely used as thermal interface materials (TIMs) to reduce the thermal contact resistance. For industrial applications, the particle-filled composite usually has high volume fraction (>50%). However, most of the research on the thermal properties of particle-filled composites has been focusing on low volume fraction composites. In this work, the finite element method (FEM) is adopted to investigate the particle-filled composites with high filler loading. We consider the close-packed simple cubic (SC), face-centered cubic (FCC), and a dual diameter (DD) model with even a higher volume fraction than the FCC. It is found that with a high volume fraction, small increase in volume fraction can lead to a strong enhancement in the overall thermal conductivity. With a certain filler loading and thermal conductivity of the matrix, the effective thermal conductivity first dramatically increases with the thermal conductivity of the filler and then saturates. We show that the effective medium theory based models cannot properly predict the effective thermal conductivity for the close-packed structures. The percolation theory based on the resistance network agrees surprisingly well with our simulation results. Through a careful investigation of the effect of proximity between adjacent particles, it is found that good contact between particles is crucial to the enhancement of the overall thermal conductivity. We also considered the interface thermal resistance between fillers and matrix and compared the simulation results with analytical models. Our analysis provides a better understanding on the heat transfer in the high volume fraction composite materials and is important for the fabrication of high thermal conductivity TIMs.

1. Introduction

With the increasing power density of electronic device, heat dissipation has become one of the most critical challenges [1]. TIMs [2] have been widely used in electronic packaging and cooling system to reduce the contact thermal resistance across jointed surfaces, such as the contact between microprocessors and heat sinks [3]. Particle-filled composite materials made of thermal grease matrix (with typical thermal conductivity of 0.1–0.4 W/m K [3,4]) and highly conductive ceramic or metallic particles are the most widely used commercial TIMs, of which the thermal conductivity values are generally in the range of 2–5 W/m K [4]. Although there are practical concerns such as thermal stability, wetting ability, and viscosity, to further increase the effective thermal conductivity of TIMs is still highly desirable to the electronic packaging industry.

Recent experimental investigations of particle filled TIMs mainly take two different approaches, i.e., increasing the volume fraction and mixing different types of fillers together. For example, Yu et al. [5] reported that the composite of hybrid size alumina fillers with the addition of graphene of 1 wt.% filled in silicone matrix had an effective thermal conductivity of 3.45 W/m K at a filler volume fraction of 63%. Gao et al. [6] found that the alumina with a diameter of 75 μm filled silicone matrix composite had an effective thermal conductivity of 2.25 W/m K at a high volume fraction of 62%. Some commercial TIMs even report thermal conductivity as high as 17 W/m K [7], although the data have not been verified by other sources. It can be seen that the reported effective thermal conductivity of composites with high volume fraction varies significantly in different studies.

Actually, the effective thermal conductivity of composites is determined by many factors [8] other than the volume fraction, such as the particle type and shape, the collective arrangement of particles, and the interfacial thermal resistance between fillers and matrix. It is generally difficult to separate the effect of these

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Nomenclature

T_1, T_2 (K) temperature
k_f (W/m K) thermal conductivity of filler
k_m (W/m K) thermal conductivity of matrix
k_{eff} (W/m K) effective thermal conductivity
Q (W) heat flow rate
A (m^2) cross-sectional area
ΔT (K) temperature difference
L (m) length of the cubic model
D (m) diameter of the particle
d (m) gap distance
f volume fraction
ΔS (m^2) contact area of the cushion
S (m^2) surface area of the particle
t (m) the half of the thickness of the cushion
δ (m) thickness of the interfacial layer
k_c (W/m K) thermal conductivity of the cushion
k_s (W/m K) thermal conductivity of the shell
R_b (m^2 K/W) interfacial thermal resistance
R_s (m^2 K/W) thermal resistance of the shell
R_m (m^2 K/W) the matrix intrinsic thermal resistance corresponding to thickness of δ
Bi Boit number
ζ_2 geometric parameter
β_1 dimensionless parameter
R dimensionless parameter
λ ratio of thermal conductivity of fillers to matrix
α dimensionless parameter
d_0 (m) critical diameter

factors by purely experimental approach. Therefore, theoretical understanding is vital to guide the design and fabrication of better composite material for thermal application. There are two main categories of analytical or semi-analytical theories. One is the effective medium theory. The widely used models include the Maxwell [9] and Bruggeman [10] symmetric model. These simple models only consider the effect of the volume fraction and are generally valid for low volume fraction (usually less than 40 vol% [11]). Another approach is based on the Rayleigh's mathematical technique of expressing the temperature field as a multipole expansion in the composites [12]. This technique works for cubic arrangements (including SC, BCC, FCC) of spherical fillers and gives more accurate solutions for larger volume fraction. However, the Rayleigh's original treatment was later questioned by Levine et al. [13] because it just considered low order multipole expansion coefficients and involved the summation of a non-absolutely convergent series. In order to overcome these difficulties, McPhedren et al. [14] and McKenzie et al. [15] extended the Rayleigh's work to take into account multipoles of arbitrarily high order and overcome the non-absolutely convergent difficulty for the case of SC particle arrangement and the case of BCC and FCC, respectively. Sangani et al. [16] modified the multipole expansion by taking advantages of the symmetric particles and the periodicity of the temperature to calculate the effective thermal conductivity for cubic array of spheres (including SC, BCC, FCC) from dilute to nearly touching volume fraction. There are also other approaches that investigate the effective thermal conductivity of composites for cubic array of spheres, such as the boundary integral method developed by Zick [17] and the asymptotic expression derived by Keller [18]. However, these works generally only consider cubic lattice arrangement of particles and are difficult to apply to volume fraction higher than the closed-packed FCC structure. To further include the effect of interface thermal resistance, several extensions of the analytical theories have been developed. For example, Hasselman and Johnson [19] derived an expression based on Maxwell model that includes the effect of interface thermal resistance. With the modification of Bruggeman model, Every et al. [20] proposed an expression that includes a dimensionless parameter Biot number to consider the interfacial thermal resistance between fillers and matrix. Cheng et al. [21] applied the Rayleigh's method to calculate the effective thermal conductivity of periodic arrays of spherical inclusions with interfacial thermal resistance, and an accurate approximation formula was provided. Some of the models are summarized in review articles [22,23].

On the other hand, the direct numerical solution of the heat diffusion equation [24] can obtain detailed information of heat transfer in composites and theoretically consider any type of arrangement of particles. Numerical simulations were performed to study different types of fillers and different arrangements of fillers, such as cubic [25], spherical elliptic fillers [26], and fibers [27], with ordered or random distribution [28]. The lattice Boltzmann method is also employed to calculate the effective thermal conductivity of particulate TIMs in squeeze flow [29] and study the heat transfer in porous media [30]. Most of these studies are focusing on composites with relatively low filler volume fraction as well. There are also some methods developed for high volume fraction, such as the Agari's [31] semi-empirical model which requires experimental data to obtain the fitting curve as the function of filler concentration. Some other methods are based on the assumption that the composites can be treated as resistance networks. For example, the effective unit cell model (EUCM) [32] uses the effective medium theory to determine the value of the resistance, and another model uses the FEM [33] to obtain the value of the resistance. The percolation model proposed by Devpura et al. [34] models the composite material as a binary compound of cubic units with the same size and then uses the resistance network to obtain the effective thermal conductivity of the composite. It needs an empirical correction factor to be applied to spherical fillers. These models for a high volume fraction either need fitting parameters, or the assumption that the system can be modeled as a resistance network. In contrast, a direct numerical simulation of the high filler loading composite materials does not require any assumptions. It can help to develop a better understanding of the heat transfer mechanism in the high filler loading composite and also to validate the numerous models that have been proposed in literature.

In this work, a numerical simulation of particle-filled composites is carried out, focusing on high filler loading composites model. A few high volume fraction unit cell models are established. On the premise of neglecting the microscale effect, the FEM is employed to investigate the effects on the effective thermal conductivity of these cells. The factors, such as volume fraction, thermal conductivity of the filler, the proximity effect of closely packed fillers, and the interfacial thermal resistance between fillers and matrix, have been analyzed. The results are also compared with different models and discussed.

2. Model description and simulation details

To model high volume fraction composite materials, ideally one should build a large simulation domain and have many particles randomly placed in the domain [35]. However, for the high volume fraction configurations, many particles will be close to each other. As will be shown later, the proximity effect between adjacent