A method to calculate the effective thermal conductivity of spherical particle-laden composite

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Abstract. Accurate calculation of the effective thermal conductivity of composite is of great importance. Based on the heat conduction differential equation, the effective thermal conductivity of spherical particle-laden composite was derived in this paper. To validate the calculation method, comparisons with the Maxwell model, Karayacoubian model, Lewis-Nielsen model, Woodside-Messmer model, and the experimental data were conducted. It was found that the results of the present method are closer to the experimental data. Further validation was conducted with comparison to the finite-element simulations. The effective thermal conductivity of a matrix cube with a spherical particle was simulated and calculated. The results obtained by these two methods were consistent with the relative error within 7%.

Key words: Effective Thermal Conductivity; Composite; Spherical Particle.

1. Introduction

Composite material is a kind of new material which is composed of filling other materials in one matrix material. It makes the composite material have the properties of both matrix material and filling material. Its comprehensive performance is superior to that of raw materials and meets various performance requirements. It has been widely used in aerospace, military, energy, electronics, chemical industry and other fields. It is used to enhance heat. The effective thermal conductivity of conductive functional composites is related to their processing methods, filling particle volume fraction, particle morphology and distribution structure [1]. Accurate calculation of thermal conductivity of composites is very important for research and improvement of composites. In numerical simulation, composites are usually divided into a large number of lattices, and the distributed filled particles are also assumed to be rectangular (2-D) or cubic (3-D) unit lattices to simplify the calculation. There are two shortcomings in this simplification: one is that the effect of shrinkage or diffusion is not considered, and the other is that the thermal conductivity is generally too large.

To correct the errors caused by the square particle hypothesis, many models have been developed to calculate the equivalent thermal conductivity of spherical particles in a cubic lattice. The model assumes that the particles are not in contact with the particles, so it is suitable for low volume fraction cases. Many researchers have proposed different improved models based on Maxwell model, considering particle shape, interfacial thermal resistance and coating.
Karayacoubian et al. [1] calculated the upper and lower limits of the equivalent thermal conductivity of the composites with spherical particles. The upper limit corresponds to the case where the isotherm is perpendicular to the direction of the heat flow and the lower limit corresponds to the case where the adiabatic line is parallel to the direction of the heat flow. Woodside and Mesmer [5] established the equivalent thermal conductivity model of two-phase porous materials, Lewis and Nielsen [6] studied the composite materials from the mechanical point of view, and also gave a formula for calculating the thermal conductivity. Song Sihong [7] et al. used the minimum thermal resistance method to calculate the thermal conductivity of spherical materials. The thermal conductivity of the composites was predicted and the results were in good agreement with the experimental data at low volume fraction. Bicheng et al. [8] The gas-solid coupling method was used to solve the three-dimensional ordered nanoporous materials at room temperature, and the equivalent thermal conductivity was obtained.

In this paper, the accurate temperature field is obtained by solving strictly the temperature conduction equation of a cubic unit lattice containing spherical particles, and then the equivalent thermal conductivity is calculated.

| Table 1. various models for calculating effective thermal conductivity of spherical particles |
|---------------------------------------------|
| model                        | formula                                                                 |
| Maxwell model [3]             | $\lambda_{eff} = \lambda_m \frac{2\lambda_m + \lambda_f - 2\phi(\lambda_m - \lambda_f)}{2\lambda_m + \lambda_f + \phi(\lambda_m - \lambda_f)}$ |
| Woodside and Mesmer model [5] | $\lambda_{eff} = \lambda_m \frac{1+1.5B\phi}{1-B\phi(1+0.895\phi)}, B = \frac{\lambda_f/\lambda_m - 1}{\lambda_f/\lambda_m + 1.5}$ |
| Lewis and Nielsen model [6]   | $\lambda_{eff} = \lambda_m \left(\frac{1 - \pi a^2}{2} + 2\pi I_{ub}\right)$ |
| Karayacoubian model lower limit [1] | $I_{ub} = \frac{1}{2(1-\lambda_m/\lambda_f)} \ln \left[ \frac{1}{1-2a(1-\lambda_m/\lambda_f)} \right] - \frac{a}{1-\lambda_m/\lambda_f}$ |
| Karayacoubian model upper limit [1] | $I_{ub} = \ln \left[ \frac{\pi(\lambda_f/\lambda_m - 1)}{2\pi(\lambda_f/\lambda_m - 1)a^2 + 1} \right]$ |

* $a$ is the radius of the filling particle and the volume fraction.

2. Mathematical Modeling

In order to solve the thermal conductivity of composites, the whole composites are divided into continuous N identical lattices, each lattice containing 1 or 0 particles. Further, assuming that the particles and the matrix material are isotropic, the thermal conductivity of the particles and the matrix material is recorded as $\lambda_F$ and $\lambda_m$, respectively. A series of micro-elements (cubes) are linked together, each of which consists of a dispersed phase monomer in the matrix material. Since a micro-element is very small for the composite material as a whole, the heat flow in the element can be considered to be linear. The two-dimensional calculation model in this paper is shown in Figure 1. Suppose a three-dimensional element with an edge length of $L$ is assumed. In the cubic element, a spherical particle with a radius is symmetrically distributed, and the heat flow is transmitted along the X direction. The temperature field of the matrix material is expressed by $T_m$, and the temperature field of the particle phase is expressed by $T_f$. The steady-state heat conduction equation in the element is in the form of Laplace equation.
\[
\n\nabla^2 T_i = \frac{\partial^2 T_i}{\partial x^2} + \frac{\partial^2 T_i}{\partial y^2} + \frac{\partial^2 T_i}{\partial z^2} = 0, \quad i = f, m \tag{1}
\]

The general solution of formula (1) is given by Hasselman and Johnson\[9\], that is,

\[
\begin{align*}
T_f &= rA\cos \theta \\
T_m &= \Delta T \cdot r \cos \theta + (B / r^2) \cos \theta
\end{align*} \tag{2}
\]

Where \(\Delta T\) is the temperature gradient, \(A\) and \(B\) are undetermined coefficients. \(r\) and \(\theta\) are spherical coordinates, and \(\theta\) is the angle between the radius \(r\) and the temperature gradient.

\[
\begin{align*}
\lambda_f (\partial T_f / \partial r) &= \lambda_m (\partial T_m / \partial r) \\
T_f - T_m &= -(\lambda_f / h_c)(\partial T_f / \partial r)
\end{align*} \tag{3}
\]

Considering the contact thermal resistance between the particle phase and the matrix material, the \(h_c\) in equation (3) is the contact thermal conductivity between the particle phase and the matrix material. If the contact thermal resistance is neglected, the temperature field is continuous on both sides of the interface.

By solving the formula (2) and formula (3), the temperature field of the element can be obtained.

\[
\begin{align*}
T_f &= \Delta T \cdot r \cos \theta - \frac{2(\lambda_m / \lambda_f)}{\lambda_m / \lambda_f + \lambda_m / (2h_c) + 1} \\
T_m &= \Delta T \cdot r \cos \theta + \frac{\Delta T \cdot \alpha^3 \cos \theta \lambda_m / \lambda_f - 1}{r^2 \lambda_m / \lambda_f + 1}
\end{align*} \tag{4}
\]

\[\text{Figure 1. two dimensional computation model}\]

When \(h_c \rightarrow \infty\), the \(T_f\) and \(T_m\) of the particle and matrix phases can be reduced to the following form of Rayleigh solutions.

\[
\begin{align*}
T_f &= \Delta T \cdot r \cos \theta - \frac{2(\lambda_m / \lambda_f)}{\lambda_m / \lambda_f + 1} \\
T_m &= \Delta T \cdot r \cos \theta + \frac{\Delta T \cdot \alpha^3 \cos \theta \lambda_m / \lambda_f - 1}{r^2 \lambda_m / \lambda_f + 1}
\end{align*} \tag{5}\]
After obtaining the temperature field in the element body, the method for calculating the effective thermal conductivity is given below. For convenience, the spherical coordinate system of Eq. (4) is transformed into Cartesian coordinate system, then Eq. (4) is written as

\[
\begin{align*}
T_j &= \Delta T \cdot x \cdot \frac{2(\lambda_n / \lambda_j)}{\lambda_n / \lambda_j + \lambda_n / (2h) + 1} \\
T_m &= \Delta T \cdot x + \frac{\Delta T \cdot a^3 x}{(x^2 + y^2)^{3/2}} \cdot \frac{\lambda_n / \lambda_j + \lambda_n / (ah) - 1}{\lambda_n / \lambda_j + \lambda_n / (ah) + 1}
\end{align*}
\] (6)

In Fig. 1, by differentiating the Y direction, one of the strip elements dy is investigated. According to Fourier's law, the heat flow through the strip element dy is obtained.

\[
dQ = -\lambda_{dy} \frac{\partial T}{\partial x} \bigg|_{x=0}
\] (7)

Equivalent thermal conductivity of ribbon microelement dy

\[
\lambda_{dy} = \frac{dQ}{\left[ T_m \big|_{x=0} - T_m \big|_{x=0} \right]}
\] (8)

By integrating the formula (7) in the interval \([-l/2, l/2]\), the equivalent thermal conductivity of the unit is obtained.

\[
\lambda_{eff} = \frac{2\lambda_n (1-C)}{l} \int_0^{l/2} \frac{(y^2 + l^2 / 4)^{3/2}}{(y^2 + l^2 / 4)^{3/2} + a^3 C} dy
\] (9)

And

\[
C = \frac{\lambda_n / \lambda_j + \lambda_n / (ah) - 1}{\lambda_n / \lambda_j + \lambda_n / (ah) + 1}
\] (10)

Equation (9) gives a case in which a spherical particle is filled in a cubic element. For other shapes of particles, the approximate simplified calculation can be carried out by using spherical particles. Assuming the volume of non-spherical particles is V, the equivalent radius is given and the solution can be substituted by Equation (9).

3. Results and Discussions
Using phenolic resin (lambda = 0.2 W / (m K)) as matrix material and spherical aluminum particles (lambda = 203 W / (m K)) as filling material, the results of equivalent thermal conductivity of composites with different volume fractions calculated by the present method and the models listed in Table 1 are shown in Figure 2. The results of this method are between the upper and lower bounds of Karayacoubian model, higher than those of Maxwell model and Lewis-Nielsen model. When the volume fraction is more than 47%, the equivalent thermal conductivity increases sharply. The reason may be that the particles contact each other and form each other when the volume fraction is higher, forming a heat conduction channel.
Lin et al. [10] studied the equivalent thermal conductivity of copper oxide particles filled in epoxy resin matrix by transient method. The results of Maxwell model and Karayacoubian model are compared with the experimental results of Lin et al. The results of this paper are shown in Figure 3. All the results are between the upper and lower bounds of the Karayacoubian model, which indirectly proves the correctness of the Karayacoubian model. As can be seen from Figures 2 and 3, the results of the Maxwell model are small and only suitable for the case of low volume fraction.

In order to further verify the proposed method, the finite element method is used to simulate the temperature field of a cubic element containing spherical particles. The matrix material is epoxy resin ($\lambda = 0.2 \, \text{W} / (\text{m} \, \text{K})$) and the filler particle is copper oxide ($\lambda = 1067.9 \, \text{W} / (\text{m} \, \text{K})$). The temperature field of the element is obtained by partial equation, and the heat flux $Q$ in the X direction is obtained. The equivalent thermal conductivity of the element containing spherical particles is solved by Fourier law.

$$\lambda_{\text{eff}} = \frac{q}{\Delta T / \Delta x} = \frac{ql}{\Delta T}$$

(11)
The results of the finite element simulation of the temperature field are shown in Fig. 4. From the heat flow direction, it can be seen that the heat tends to pass through the particles of the highly conductive material, the isotherm is not perpendicular to the original heat flow line, and the adiabatic line is not parallel to the direction of the heat flow, so this situation is contained in the upper and lower bounds of the Karayacoubian model. The equivalent thermal conductivity of a cube containing spherical particles is obtained by the radius of the particle (i.e. volume fraction), as shown in Fig. 5. The relative error between the results obtained by the present method and the finite element method is 7%.

![Finite element simulation results of elements containing spherical particles conductivity calculated by finite element](image1)

**Figure 4.** Finite element simulation results of elements containing spherical particles conductivity calculated by finite element

![Comparison of equivalent thermal method and finite element method](image2)

**Figure 5.** Comparison of equivalent thermal method and finite element method

### 4. Conclusion
Based on the differential equation of heat conduction, the equivalent thermal conductivity of composite materials with spherical particles is derived. In order to verify the correctness of this method, this method is compared with the traditional Maxwell, Karayacoubian, Lewis-Nielsen models and experimental data. It is found that this method is in better agreement with other models. The equivalent thermal conductivity of the composites is obtained by finite element simulation, and the relative error is 7%. This method can be used to calculate the equivalent thermal conductivity in the element body, and can be used to replace the cubic particle hypothesis in the traditional algorithm.

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