Effective Thermal Conductivity of Graded Nanocomposites with Interfacial Thermal Resistance

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Abstract. This work employs the self-consistent method to investigate the effective thermal conductivity distribution in functionally graded materials (FGMs) considering the Kapitza interfacial thermal resistance. A heat conduction solution is first derived for one spherical particle embedded in a graded matrix with a prefect interface. The interfacial thermal resistance of a nanoparticle is simulated by a new particle with a lower thermal conductivity. A novel self-consistent formulation is developed to derive the averaged heat flux field of the particle phase. Then the temperature gradient can be obtained in the gradation direction. From the relation between the effective flux and temperature gradient in the gradation direction, the effective thermal conductivity distribution is solved. If the gradient of the volume fraction distribution is zero, the FGM is reduced to a composite containing uniformly dispersed nanoparticles and a explicit solution of the effective thermal conductivity is provided. Disregarding the interfacial thermal resistance, the proposed model recovers the conventional self-consistent model. Mathematically, effective thermal conductivity is a quantity exactly analogous to effective electric conductivity, dielectric permittivity, magnetic permeability and water permeability in a linear static state, so this method can be extended to those problems for graded materials.

Keywords: functionally graded materials; nanocomposites; interfacial thermal resistance; self-consistent method; effective thermal conductivity

INTRODUCTION

In recent years, functionally graded particulate nanocomposites have been proposed to reduce the thermal stress in thermal barrier coatings simultaneously carrying the benefits of light weight of the structure and excellent physical properties of nanomaterials [1-4]. In the literature, most traditional works have considered the idealized case of perfect interface so that continuous boundary conditions along the interface between particles and matrix are employed in the calculation of averaged fields [5, 6]. As a result, the size effect of particles has no contribution to the effective material behavior.

However, with decrease of particle size, the surface-to-volume ratio of the particles increases. Therefore the interface between a particle and the surrounding matrix plays an important role in the effective materials behavior, especially for nanocomposites. It has been observed that the Kapitza interfacial thermal resistance greatly changes the

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effective thermal conductivity for nanocomposites [7, 8]. When particle radius is smaller than a certain value, the contribution of the particles to the effective thermal conductivity is dominated by interfaces instead of the particle's conductivity. Hasselman and Johnson [9] and Benveniste [10] respectively obtained the same formulation [11] for the effective thermal conductivity of uniformly dispersed particulate composites. However, for nanoFGMs, no applicable model exists in the literature.

This work proposes a multi-scale model to investigate the effective thermal conductivity distribution in FGMs considering the Kapitza interfacial thermal resistance. A fundamental solution for one spherical particle embedded in a graded matrix is derived with the interfacial thermal resistance. Consider a two-phase FGM placed between two parallel platens (Figure 1(a)). The platens have different fixed temperatures, so a steady state heat flux is induced in the FGM. In microscopic scale, discrete particles embedded in a continuous matrix can be observed. For an arbitrarily chosen particle, because particle is much smaller than the size of the FGM, using the self-consistent method, one can obtain the particle averaged heat flux from the solution of one particle in an unbounded FGM. From the relation between the averaged heat flux and temperature gradient, the effective thermal conductivity distribution is derived in the gradation direction of the FGM. Some special cases of the model are discussed and validated with experimental data.

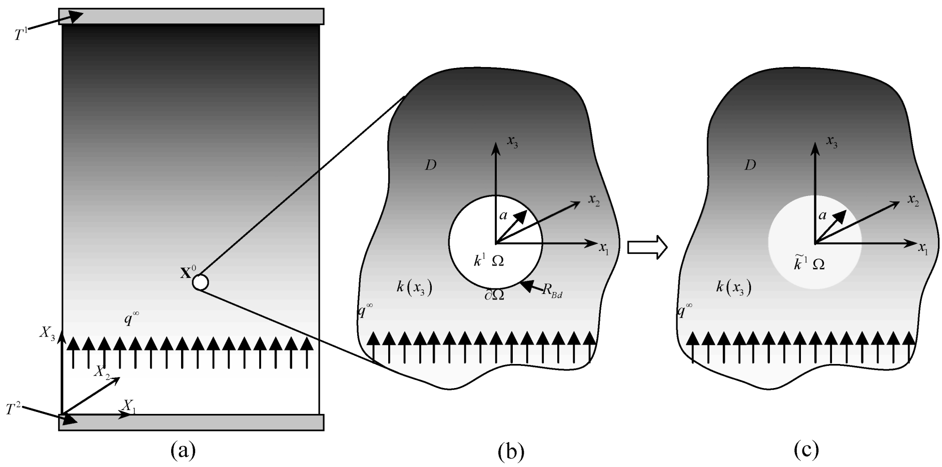

FIGURE 1. Illustration of a self-consistent model for FGMs. (a) An FGM at the macroscale; (b) one particle embedded in the FGM itself with an interfacial thermal resistance, and; (c) a new particle embedded in the FGM with a perfect interface and a lower thermal conductivity.

FORMULATION

Consider an FGM containing two phases, discrete particle *A* and continuous matrix *B*, whose volume fractions gradually change in the gradation direction X_3 (Figure 1(a)). To test the effective thermal conductivity distribution in the X_3 direction, two different temperatures $\left(T^{2} > T^{1}\right)$ are applied in the lower and upper platens, so that a steady state heat flux field is induced. Macroscopically, because the material is homogeneous at each $X_1 - X_2$ layer and the heat flux is conservative for each layer, the averaged heat flux should be uniform, written as q^{∞}. Microscopically, because the microstructure of the material is represented by discrete particles dispersed in continuously graded matrix, the local field is severely inhomogeneous.

For an arbitrary material point $\mathbf{X}^0$ in Figure 1(a), the effective material behavior can be represented by the averaged response of two material phases. Under steady conditions and without the presence of heat sources, the overall averaged heat flux and temperature gradient can be written as [11]:

$$\langle \mathbf{q} \rangle_D \left(X_3^0\right) = \phi\left(X_3^0\right) \langle \mathbf{q} \rangle_\Omega \left(X_3^0\right) + \left[1 - \phi\left(X_3^0\right)\right] \langle \mathbf{q} \rangle_M \left(X_3^0\right), \tag{1}$$

$$\langle \mathbf{H} \rangle_D \left(X_3^0\right) = \phi\left(X_3^0\right) \left[\langle \mathbf{H} \rangle_\Omega \left(X_3^0\right) + \mathbf{J}\left(X_3^0\right)\right] + \left[1 - \phi\left(X_3^0\right)\right] \left[\langle \mathbf{H} \rangle_M \left(X_3^0\right)\right], \tag{2}$$

where the angle brackets with subscripts D, Ω, and M denote the volume averages over the whole RVE, particle phase, and matrix phase of the material point; $\mathbf{q}$ and $\mathbf{H}$ represent the heat flux and temperature gradient. It is noted that because the normal heat flux across the interface between particle and matrix $\partial\Omega$ is continuous, the averaged heat flux only includes two terms from two material phases; whereas due to a temperature discontinuity existing across the interface, an additional term $\mathbf{J}$ represents the contribution of the temperature jump across $\partial\Omega$.

The temperature jump is proportional to the normal heat flux across the interface as the following relations:

$$\Delta T = -R_{Bd} \mathbf{q}^n \tag{3}$$

where R_{Bd} denotes the interfacial thermal resistance [7]. Because Kapitza [12] first quantitatively measured the interfacial thermal resistance between helium and some solids, the physical quantity is also named as Kapitza resistance.

To solve averaged the heat flux field in particle phase A, the self-consistent method [13, 14] is used as follows:

- For a given point $\mathbf{X}^0$ in the global FGM system as seen in Figure 1(a), we build up a local coordinate system with a particle centered at the origin as seen in Figure 1(b). The thermal conductivity of the graded matrix is assumed to be the same the FGM itself at the global system;
- Because the particle is essentially contacted with the continuous matrix phase B, a constant interfacial thermal resistance exists along the interface between the particle and the matrix as seen in Figure 1(b);
- To simplify the solution for the particle's averaged field, the particle with interfacial thermal resistance is replaced by a new particle with perfect thermal interface as seen in Figure 1(c). Therefore, the particle's averaged heat flux field

is solved by one particle embedded in an unbounded graded matrix under a uniform heat flux at far field.

Although the heat flux field for one particle embedded in an FGM with an interfacial thermal resistance is fairly complex, in a sense of volume average, the inhomogeneous heat flux field over the particle can be replaced by the averaged one written as $\overline{\mathbf{q}}$. Observing from the outside of the particle surface, one can write the averaged temperature gradient as

$$\langle \mathbf{H} \rangle_{\Omega} = \frac{1}{V^{\Omega}} \left[\int_{\Omega} - \frac{\mathbf{q}(\mathbf{x})}{k^1} d\mathbf{x} + \int_{\partial\Omega} \Delta T(\mathbf{x}) \mathbf{n} dS \right] = -\overline{\mathbf{q}} / k^1 - \frac{R_{Bd} \overline{\mathbf{q}}}{a}. \tag{4}$$

where k^1 denotes the thermal conductivity of the particle, and a the radius of the particle. Therefore, no matter how complex the local heat flux field is in the particle domain, observed from the outside of the particle, it is equivalent to a new particle with a perfect interface but a lower thermal conductivity as,

$$\widetilde{k}^1 = k^1 / \left(1 + R_{Bd} k^1 / a\right). \tag{5}$$

Therefore, using the equivalent particle, the averaged temperature gradient in Equation (2) can be written as

$$\langle \mathbf{H} \rangle_D \left(X_3^0\right) = \phi\left(X_3^0\right) \langle \mathbf{H} \rangle_{\Omega} \left(X_3^0\right) + \left[1 - \phi\left(X_3^0\right)\right] \langle \mathbf{H} \rangle_M \left(X_3^0\right)\right], \tag{6}$$

with the constitutive law as

$$\langle \mathbf{H} \rangle_{\Omega} = \langle \mathbf{q} \rangle_{\Omega} / \widetilde{k}^1, \ \langle \mathbf{H} \rangle_M \left(X_3^0\right) = \langle \mathbf{q} \rangle_M / \overline{k}\left(X_3^0\right). \tag{7}$$

The particle's averaged heat flux can be obtained from the solution for one particle embedded in an unbounded FGM with a perfect interface [15], written as:

$$\langle q_3 \rangle_{\Omega} \left(X_3^0\right) = \frac{3\widetilde{k}^1}{3\widetilde{k}^1 - 2\left(1 - a^2 \overline{\alpha}^2\right)\left[\widetilde{k}^1 - \overline{k}\left(X_3^0\right)\right]} \langle q_3 \rangle_D \left(X_3^0\right). \tag{8}$$

where $\overline{k}$ denotes the effective thermal conductivity, which is a function of the coordinate X_3; and

$$\overline{\alpha} = \frac{1}{2\overline{k}} \frac{d\overline{k}}{dX_3}, \tag{9}$$

which reflects the effect of the material gradation.

Using Equations (1), and (6)-(8), one can derive the relation between the averaged heat flux and temperature gradient as

$$\langle q_3 \rangle_D \left(X_3^0\right) = k^B \left[1 - \phi \frac{3\left(\widetilde{k}^A - k^B\right)}{3\widetilde{k}^A - 2\left(1 - a^2\overline{\alpha}^2\right)\left(\widetilde{k}^A - \overline{k}\left(X_3^0\right)\right)}\right]^{-1} \langle H_3 \rangle_D \left(X_3^0\right). \quad (10)$$

Thus, the effective thermal conductivity can be obtained as

$$\overline{k}\left(X_3^0\right) = k^B \left[1 - \phi \frac{3\left(\widetilde{k}^A - k^B\right)}{3\widetilde{k}^A - 2\left(1 - a^2\overline{\alpha}^2\right)\left(\widetilde{k}^A - \overline{k}\left(X_3^0\right)\right)}\right]^{-1}. \quad (11)$$

Notice that the above expression is implicit because the right side includes $\overline{k}\left(X_3^0\right)$ itself. Equation (11) can be ultimately simplified into a quadratic equation with two roots. One of them that is in the range between $\widetilde{k}^A$ and k^B should be the physical solution for $\overline{k}\left(X_3^0\right)$. However, because $\overline{\alpha}$ is still unknown, we need solve the above equation in a recursive method, in which a boundary condition is typically implied as:

$$\overline{k}(0) = k^B, \quad (12)$$

because the volume fraction of the particle phase A is zero. For particle volume fraction does not start from 0%, the modified boundary condition of $\overline{k}(0)$ can be still obtained with the aid of the uniform composite model as seen in Equation (13).

RESULTS AND DISCUSSION

Equation (11) provides a prediction for the effective thermal conductivity distribution in an FGM. When the material gradation is zero, the effective thermal conductivity will only depend on the volume fraction, so Equation (11) will be reduced to

$$\overline{k} = k^B \left[1 - \phi \frac{3\left(\widetilde{k}^A - k^B\right)}{3\widetilde{k}^A - 2\left(\widetilde{k}^A - \overline{k}\left(X_3^0\right)\right)}\right]^{-1}. \quad (13)$$

Every et al. [7] tested the effective thermal conductivity for diamond/ZnS composites with two radii, i.e. $a = 250nm$ and $2.0\mu m$. The other material constants are used as: $k^{diamond} = 600W/mK$, $k^{ZnS} = 17.4W/mK$, and $R_{Bd} = 6\times10^{-8} m^2K/W$. In Figure 2(a), for the case of $a = 2.0\mu m$, the effective thermal conductivity increases with the volume fraction of the diamond particles due to the reinforcement of the particles with much higher thermal conductivity; whereas for the case of $a = 250nm$, the effective thermal conductivity decreases because the interfacial thermal resistance plays a dominant role at this size. The present model predicts the tendency of the experimental data well though some difference is found for the case of $a = 250nm$ due to the irregular particle shape and nonuniform size of particles.

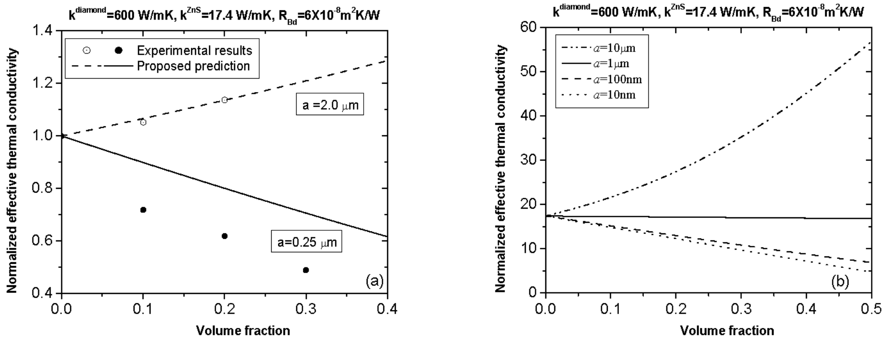

FIGURE 2. Effective thermal conductivity versus volume fraction for (a) uniform diamond/ZnS composites, and (b) FGMs with different particle sizes

Figure 2(b) shows the effective thermal conductivity distribution for linearly distributed FGMs. Here the thickness of the FGMs is set as 1mm. Four particle sizes are illustrated as $a = 10, 1, 0.1, 0.01\mu m$. It is found that the particle size has dramatic effect on the effective thermal conductivity behavior of FGMs.

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