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A New Approach to Predict the Thermal Conductivity of Composites with Coated Spherical Fillers and Imperfect Interface

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An examination of the concept of a microgeometry proposed by Benveniste reveals that the thermal conductivity of the concentric sphere adopted by generalized self-consistent model (GSCM) is equal to that of the composite. It is also noted that the thermal conductivities of the composite with spherical fillers predicted by GSCM and modified Eshelby model (MEM) are the same. These equivalencies enable to propose a simple and alternative approach for determining the thermal conductivity of the composite with multiply coated spherical fillers by applying MEM repeatedly. The present result is compared and shows the exact agreement with the results from literatures.
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1. Introduction

In composites, the third layer is developed in the form of a coating, chemical reaction layer, and interfacial gap at the interface between the matrix and reinforcement regardless of being intended or not. Its thermo-mechanical properties are different from those of the basic constituents and are generally known to severely influence the thermo-mechanical properties of composites. The thermal conductivity of composites is drastically lowered due to the existence of a thermal barrier resistance between the matrix and reinforcement which is experimentally confirmed.[1] This resistance is induced by an interfacial gap which is developed by the combined effect of an imperfect mechanical or chemical adherence at the interface and a mismatch in the coefficients of thermal expansion.[2] In order to improve the thermal conductivity of composites, the reinforcement is intentionally coated with the third highly conductive material. Models have been proposed to examine the effect of the coating[3–9] on the thermal conductivity of composites together with the interfacial thermal barrier.[10–14] One class of these models[4–6,9,11–14] is to solve Laplace equation to obtain the temperature field of composites which is similar to the generalized self-consistent model (GSCM).[15] The other[3,10] is to employ Esehlby's theory[16] modified with Mori-Tanaka's mean field approach[17] which is called modified Eshelby model (MEM).

Benveniste and Miloh[6,18,19] introduced the concept of a microgeometry for determining the thermal conductivity of composites and it is embedded in the effective medium having composite property. The microgeometry consists of the matrix and reinforcement whose shape and thermal conductivity are analytically determined not to disturb the original temperature field and heat flux of the composite. The obtained conductivity is thus the exact conductivity of the composite. By using the microgeometry, Benveniste and Miloh computed the thermal conductivities of composites[18] and cracked bodies[19] and coated short-fiber composites.[6] The shape of the microgeometry for spherical fillers is exactly the same as that of the concentric sphere, spherical filler surrounded by spherical matrix, employed by GSCM and the thermal conductivities predicted by both methods are consistent.[19] Benveniste[20] examined the effective thermal conductivity of composites with a thermal contact resistance between the constituents by using GSCM and MEM and concluded that both methods, distinctly different in their approach, result in the same closed-form simple expression for the effective thermal conductivity.

Based on these investigations, it is noted that the thermal conductivity of the microgeometry and concentric sphere can be derived by MEM. This result is extended to determine the thermal conductivity of the composite with coated spherical fillers and imperfect interface. The representative models to predict the thermal conductivity of this composite are classified as the two groups, but their derivations are rather complicated. In this letter, a simple and alternative approach to determine the thermal conductivity of the composite is proposed by applying MEM repeatedly. The thermal conductivity predicted by the present model is compared with other results from literatures.[13]

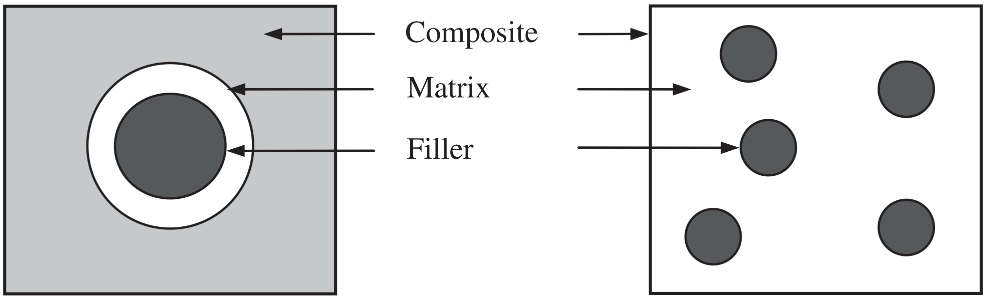
2. Model

2.1 Basic formulation

The thermal conductivity of the composite is computed by using MEM, where the spherical fillers are assumed to be perfectly bonded to the matrix. Let's first consider the composite consisting of isotropic matrix and spherical fillers as shown in Fig. 1, where Fig. 1(a) and (b) represent the models for predicting the effective thermal conductivity of the composite with spherical fillers by GSCM[13,14,20] and MEM,[20] respectively. The thermal conductivities of the composite, k_{eff}^{mf}, predicted by both GSCM and MEM are proved to be the same and given by

$$\frac{k_{eff}^{mf}}{k_m} = \frac{2(1-f_{mf}) + (1+2f_{mf})k_{fm}}{(2+f_{mf}) + (1-f_{mf})k_{fm}}, \tag{1}$$

where k, f, and subscripts and superscripts m and f stand for the thermal conductivity, filler volume fraction, and the matrix and filler, respectively. The detailed derivations by



(a) (b)

Fig. 1 Models for predicting the thermal conductivity of composites with spherical fillers; (a) generalized self-consistent model, and (b) modified Eshelby model.

both models are omitted here. The thermal conductivity ratio of the constituents is defined as

$$k_{ij} = \frac{k_i}{k_j}, \tag{2}$$

where subscripts i and j represent the constituent materials, respectively. The volume fraction of the fillers is f_{mf} defined as

$$f_{mf} = \frac{V_f}{V_m + V_f}, \tag{3}$$

where V denotes the volume.

According to GSCM,[13–15,20] the thermal conductivity of a composite is derived by embedding a concentric sphere into an infinite medium having that of the composite. The concentric sphere is the spherical filler surrounded by the same shape of the matrix, where the volume fraction of the filler is the same as the volume fraction of the filler in the entire composite under investigation. Since the thermal conductivity of the concentric sphere represents that of the composite itself[19] and the thermal conductivities of the composite predicted by GSCM and MEM are the same as mentioned in the above,[20] the thermal conductivity of the concentric sphere can be easily computed by MEM.

2.2 Thermal conductivity of coated filler

Let's focus on the determination of the effective thermal conductivity of a coated filler only. According to the model for GSCM shown in Fig. 1(a), the coated filler can be simulated to be embedded into the composite having the conductivity of the coated filler. Based on the analogy between GSCM and MEM shown in Fig. 1, Fig. 1(a) is converted into Fig. 1(b) for the computation of the thermal conductivity by MEM. In Fig. 1(b), the composite is treated as that the matrix is coating material and the reinforcement is filler. The thermal conductivity of the coated filler, k_{eff}^{cf}, can be obtained by replacing k_m, f_{mf}, and the subscript m in Eq. (1) with k_c, f_{cf}, and the subscript c and is expressed as

$$\frac{k_{eff}^{cf}}{k_c} = \frac{2(1 - f_{cf}) + (1 + 2f_{cf})k_{fc}}{(2 + f_{cf}) + (1 - f_{cf})k_{fc}}, \tag{4}$$

where f_{cf} and subscript c represent the filler volume fraction in the coated filler and coating material, respectively and f_{cf} is related with the volume of the coating material and fillers, V_c and V_f:

$$f_{cf} = \frac{V_f}{V_c + V_f}. \tag{5}$$

The thermal conductivity of the coated filler so derived is consistent with the result by Benveniste.[6]

2.3 Thermal conductivity of coated filler with imperfect interface

Once the thermal conductivity of the coated filler is given, the thermal conductivity of the coated filler with an imperfect interface is obtained. Three interfacial conductance such as fiber-matrix contact, heat transfer through gas filling any interfacial gap, and heat transfer by radiation can be simulated as a thin layer between the filler and matrix. It is assumed for simplicity of analysis that the layer is a material placed between the matrix and filler and whose effective thermal conductivity and thickness are k_i and t_i, respectively. The thermal contact conductance h is related with k_i and t_i:

$$k_i = ht_i. \tag{6}$$

The coated fillers with the imperfect interface are considered as a composite consisting of the coated fillers and interfacial layers. Following the aforementioned procedure, it can be schematically represented for the computation of the thermal conductivity by GSCM which is shown in Fig. 2(a). Figure 2(a) is further transformed to Fig. 2(b) by using the analogy between GSCM and MEM. In Fig. 2(b), the composite is treated as that the matrix is the interface material and the reinforcement is the coated filler. The thermal conductivity of the coated fillers with imperfect interface, k_{eff}^{icf}, is computed by replacing k_m, k_f and f_{mf} in Eq. (1) with k_i, k_{eff}^{cf} in Eq. (4) and f_{icf}, respectively. It is represented as

$$\frac{k_{eff}^{icf}}{k_i} = \frac{2(1 - f_{icf}) + (1 + 2f_{icf})k_{eff}^{cf}/k_i}{(2 + f_{icf}) + (1 - f_{icf})k_{eff}^{cf}/k_i}, \tag{7}$$

where f_{icf} stands for the volume fraction of the coated fillers in the composite and is defined as

$$f_{icf} = \frac{V_c + V_f}{V_i + V_c + V_f}. \tag{8}$$

After the rearrangement of Eq. (7) with Eq. (4), the closed-form solution of the thermal conductivity k_{eff}^{icf} is expressed as

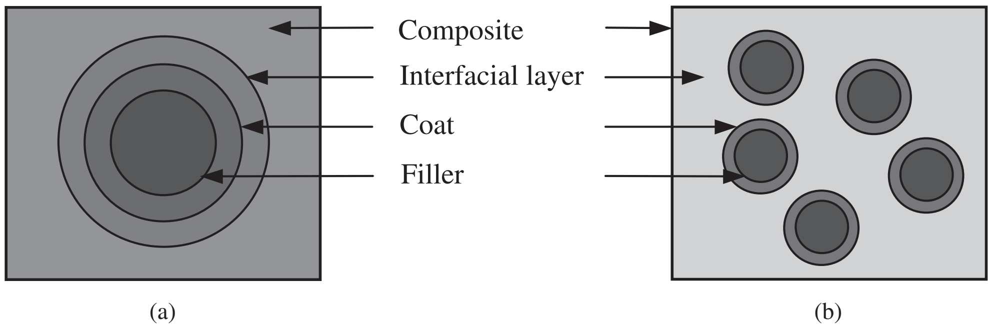


Fig. 2 Models for predicting the thermal conductivity of the coated spherical fillers with the interfacial layer; (a) generalized self-consistent model, and (b) modified Eshelby model.

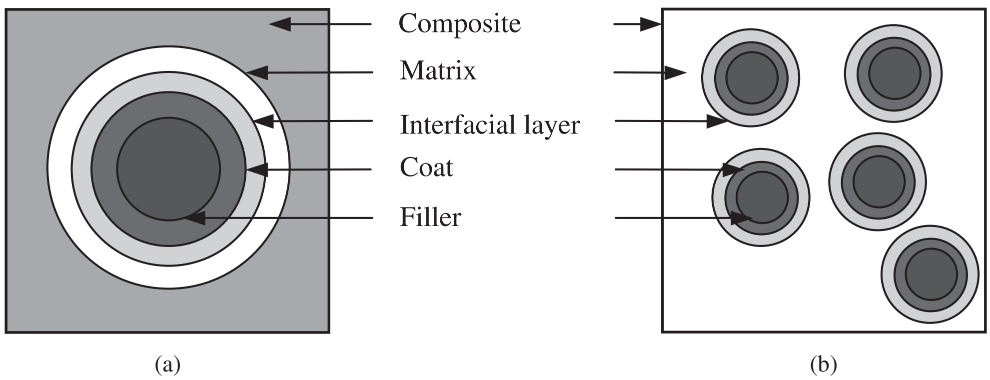


Fig. 3 Models for predicting the thermal conductivity of the composites with coated spherical fillers and interfacial layer; (a) generalized self-consistent model, and (b) modified Eshelby model.

$$\frac{k_{eff}^{icf}}{k_i} = \frac{2(1-f_{icf})[(2+f_{cf})k_i + (1-f_{cf})k_{fc}k_i] + (1+2f_{icf})[2(1-f_{cf})k_c + (1+2f_{cf})k_f]}{(2+f_{icf})[(2+f_{cf})k_i + (1-f_{cf})k_{fc}k_i] + (1-f_{icf})[2(1-f_{cf})k_c + (1+2f_{cf})k_f]}. \tag{9}$$

2.4 Thermal conductivity of composite

Finally, the thermal conductivity of the composite consisting of the matrix and the coated spherical fillers with the imperfect interface is simply derived using the above result k_{eff}^{icf}. The composite can be schematically represented for the computation of the thermal conductivity by GSCM which is shown in Fig. 3(a). Figure 3(a) is further reduced to Fig. 3(b) by using the analogy between GSCM and MEM. In Fig. 3(b), the composite is treated as that the matrix is the matrix material and the reinforcement is the coated fillers with the imperfect interface. The thermal conductivity of the composite, k_{eff}^{micf}, is computed by replacing k_f and f_{mf} in Eq. (1) with k_{eff}^{icf} in Eq. (7) and f_{micf} in Eqs. (4) and (5), respectively and given by

$$\frac{k_{eff}^{micf}}{k_m} = \frac{2(1-f_{micf}) + (1+2f_{micf})k_{eff}^{icf}/k_m}{(2+f_{micf}) + (1-f_{micf})k_{eff}^{icf}/k_m} = \frac{N}{D}, \tag{10}$$

where f_{micf} stands for the volume fraction of the coated fillers with interfacial layers in the composite and is defined as

$$f_{micf} = \frac{V_i + V_c + V_f}{V_m + V_i + V_c + V_f}, \tag{11}$$

$$\begin{aligned} N = {} & 2(1-f_{micf})(2+f_{icf})[(2+f_{cf}) + (1-f_{cf})k_{fc}]k_m \\ & + 2(1-f_{micf})(1-f_{icf})[2(1-f_{cf})k_c + (1+2f_{cf})k_f]k_{mi} \\ & + 2(1+2f_{micf})(1-f_{icf})[(2+f_{cf})k_i + (1-f_{cf})k_{fc}k_i] \\ & + (1+2f_{micf})(1+2f_{icf})[2(1-f_{cf})k_c + (1+2f_{cf})k_f], \end{aligned} \tag{12}$$

$$\begin{aligned} D = {} & (2+f_{micf})(2+f_{icf})[(2+f_{cf}) + (1-f_{cf})k_{fc}]k_m \\ & + (2+f_{micf})(1-f_{icf})[2(1-f_{cf})k_c + (1+2f_{cf})k_f]k_{mi} \\ & + 2(1-f_{micf})(1-f_{icf})[(2+f_{cf})k_i + (1-f_{cf})k_{fc}k_i] \\ & + (1-f_{micf})(1+2f_{icf})[2(1-f_{cf})k_c + (1+2f_{cf})k_f]. \end{aligned} \tag{13}$$

Since the thickness of the interfacial layer approaches 0, the third terms in both equations, Eqs. (12) and (13), vanish and the second terms in these equations can be further simplified as

$$\lim_{t_i \to 0} \frac{1-f_{icf}}{k_i} = \lim_{t_i \to 0} \frac{(r_c+t_i)^3 - r_c^3}{(r_c+t_i)^3} \frac{1}{ht_i} = \frac{3}{hr_c}, \tag{14}$$

where r_c is the radius of coated filler and Eq. (6) is used. After the rearrangement of Eq. (10) with Eq. (14), the closed-form solution of the thermal conductivity k_{eff}^{micf} is expressed as

$$\frac{k_{eff}^{micf}}{k_m} = \frac{2(1 - f_{micf})\Psi + \beta\Theta_N}{(2 + f_{micf})\Psi + \beta\Theta_D}, \tag{15}$$

where $\beta = hr_c/k_c$,

$$\Psi = [2(1 - f_{cf}) + (1 + 2f_{cf})k_{fc}], \tag{16}$$

$$\begin{aligned}\Theta_N = {} & 2(1 - f_{micf})[(2 + f_{cf}) + (1 - f_{cf})k_{fc}] \\ & + (1 + 2f_{micf})[2(1 - f_{cf})k_{cm} + (1 + 2f_{cf})k_{fm}],\end{aligned} \tag{17}$$

$$\begin{aligned}\Theta_D = {} & (2 + f_{micf})[(2 + f_{cf}) + (1 - f_{cf})k_{fc}] \\ & + (1 - f_{micf})[2(1 - f_{cf})k_{cm} + (1 + 2f_{cf})k_{fm}].\end{aligned} \tag{18}$$

The thermal conductivity of the composite with the coated spherical fillers and imperfect interface derived by successively using MEM, Eq. (15), is exactly the same as the results from the literature.[13] The symbols in the present study, f_{cf}, f_{micf}, k_m, k_{fc}, k_{fm}, and k_{cm} correspond to the symbols in the reference,[13] $1/v_{f3}$, v_f, k_1, k_{32}, k_{21}, and k_{31}, respectively. It can be inferred from the present result that instead of solving the complicated Laplace equation for the composite with multiply coated spherical fillers and imperfect interface, its thermal conductivity can be easily predicted by the present approach.

3. Conclusions

The observed equivalency of the thermal conductivities of the concentric sphere and the composite itself adopted for GSCM and the same result predicted by both GSCM and MEM enable the simple and alternative approach to predict the thermal conductivity of the composite with the coated spherical fillers and imperfect interface. The present model predicts the thermal conductivity by repeatedly using MEM and its closed-form solution is shown to be the same as the other results from literatures. It is clear that the present approach can be easily extended to composites with multiply coated spherical fillers and imperfect interfaces.

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