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Effective thermal conductivity of three-component composites containing spherical capsules

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ABSTRACT

This paper presents detailed numerical simulations predicting the effective thermal conductivity of spherical monodisperse and polydisperse core–shell particles ordered or randomly distributed in a continuous matrix. First, the effective thermal conductivity of this three-component composite material was found to be independent of the capsule spatial distribution and size distribution. In fact, the study established that the effective thermal conductivity depended only on the core and shell volume fractions and on the core, shell, and matrix thermal conductivities. Second, the effective medium approximation reported by Felske (2004) [21] was in very good agreement with numerical predictions for any arbitrary combination of the above-mentioned parameters. These results can be used to design energy efficient composites, such as microencapsulated phase change materials in concrete and/or insulation materials for energy efficient buildings.

1. Introduction

In 2010, building operations accounted for 41% of total US primary energy resource consumption [1]. Approximately, half of this energy was consumed for heating, ventilation, and air conditioning (HVAC) [1]. A common strategy to improve building energy efficiency is to use materials with a large thermal mass, *e.g.*, concrete or brick [2,3]. While these materials can store large amounts of energy per unit mass, they operate passively, demonstrating only a sensible heat response [2,3]. To add an active or temperature sensitive dimension to the thermal behavior of building materials, there is interest in embedding phase change materials (PCMs) in building elements [4–7]. By reversibly undergoing solid–liquid phase transitions in relation to the temperature of their local environment, PCMs are able to actively and adaptively absorb and release latent heat required to induce phase transitions. These actions further enhance the thermal inertia of building systems. As such, if properly implemented, PCMs embedded in building materials can limit thermal exchange through exterior walls, reducing the need and cost for HVAC operations, and thus improving building energy efficiency.

The incorporation of PCMs (*e.g.*, paraffin waxes, hydrated salts, or fatty acids) in building composites is facilitated by encapsulating the PCMs in a polymeric shell [6,5,4,7]. This serves to isolate the PCM from high pH chemical environments common to building materials, thus enhancing durability and limiting contamination [4–7]. When PCMs are embedded in a cementitious material, the resultant composite consists of three distinct components in the form of matrix (often cement-based), shell (often polymer-based), and PCM (often organic in nature). Clearly, this is a complex three-component composite material whose effective thermal properties must be predicted accurately to estimate heat transfer across composite building walls.

This study aims (1) to rigorously predict the effective thermal conductivity of three-component core–shell composite materials (2) to identify the controlling design parameters and (3) to derive design rules for composite walls. The results of this study could also be applicable to other multicomponent composites including self-healing microcapsule-doped polymers [8] and hollow glass microsphere-embedded syntactic foams [9], to name a few.

2. Background

Numerous models have been derived to predict the effective thermal conductivity of two-component composites as reviewed by Progelhof et al. [10], for example. Comparatively, few models

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Nomenclature

A	parameter in Eq. (4)
A_c	cross-sectional area, m^2
B	parameter in Eq. (4)
C_D	centroidal distance between two proximal capsules, µm
D	diameter, µm
k	thermal conductivity, W/m K
L	unit cell length, µm
N	number of unit cells
$\mathbf{n}$	normal unit vector
p	number of spherical capsules in a unit cell
r	radius, µm
q''_x, q''_y, q''_z	heat flux along the x-, y-, and z-directions, W/m^2
$\bar{q}''_x$	area-averaged heat flux along the x-direction, W/m^2
t_s	thickness of capsule shell, *i.e.* $t_s = (D_s - D_c)/2$, µm
T	temperature, K
T_o, T_L	temperature at $x = 0$ and $x = L$, K

Greek symbols

β	parameter in Eq. (9)
Δx	minimum mesh size, µm
δ	ratio of shell diameter to core diameter, $\delta = D_s/D_c$
ϕ_i	volume fraction of phase “i” in the composite structure
$\phi_{c/s}$	volume fraction of core in the capsule, $\phi_{c/s} = \phi_c/(\phi_c + \phi_s)$
ϕ_{c+s}	volume fraction of capsules in the composite structure, $\phi_{c+s} = \phi_c + \phi_s$
ϕ_{max}	volume fraction of closely-packed capsules
Θ_N, Θ_D	numerator and denominator of the Felske model (Eq. (3))

Subscripts

c	refers to core
$c+s$	refers to core–shell particle
cr	refers to the critical thermal conductivity ratios
eff	refers to effective properties
m	refers to matrix
s	refers to shell

exist for three-component composites [11–21]. Several models were developed for liquid and gas phases in a porous solid matrix such as building materials or soil [17,18]. Other models require prior knowledge of the temperature gradients in each component of the composite to determine the effective thermal conductivity [13,14]. The most practical models provide explicit analytical expressions for the effective thermal conductivity of three-component composites based on the constituent thermal conductivities and on the geometric parameters of the composite structure such as core and shell diameters and/or volume fractions.

Lichtenecker [20] proposed an ad hoc expression for the electrical permittivity of a composite consisting of any number of randomly mixed components [22]. Woodside and Messmer [22], among others, have applied this model to the effective thermal conductivity of three-component composites expressed as [20,22,23],

$$k_{eff} = k_c^{\phi_c} k_s^{\phi_s} k_m^{\phi_m} \tag{1}$$

where k_c, k_s, and k_m are the thermal conductivities of the core, shell, and matrix materials, respectively. Similarly, ϕ_c, ϕ_s, and $\phi_m = 1 - \phi_c - \phi_s$, are the volume fractions of the core, shell, and matrix materials, respectively. Woodside and Messmer [22] referred to Eq. (1) as a “geometric mean” and noted that it corresponds to the arithmetic mean of the logarithms of the constituent thermal conductivities. Zakri et al. [23] analytically derived Lichtenecker’s [20] model (Eq. (1)) for the effective electrical permittivity of three-component composites. They concluded that Eq. (1) is “*physically founded*,” despite criticism from Reynolds and Hough [24] who suggested that the model “*lacked a theoretical basis*.” Note that Eq. (1) predicts that k_{eff} vanishes if the thermal conductivity of either the core or the shell vanishes. This is obviously not the case since heat conduction could still take place through the continuous matrix material.

Brailsford and Major [19] developed a model for the effective thermal conductivity of monodisperse homogeneous particles randomly distributed in a continuous matrix. This two-component model was equivalent to the Maxwell–Garnett model for electrical conductivity [25]. Brailsford and Major [19] extended the two-component model to account for monodisperse homogeneous particles made of two different materials randomly distributed in a continuous matrix. Then, the effective thermal conductivity of three-component media was given by [19],

$$k_{eff} = \frac{k_m\phi_m + k_c\phi_c\frac{3k_m}{(2k_m+k_c)} + k_s\phi_s\frac{3k_m}{(2k_m+k_s)}}{\phi_m + \phi_c\frac{3k_m}{(2k_m+k_c)} + \phi_s\frac{3k_m}{(2k_m+k_s)}} \tag{2}$$

Model predictions for two-component media agreed well with experimental data for the effective thermal conductivity of solid glass spheres surrounded by air or water [19]. However, experimental validation was not reported for three-component composite materials.

Felske [21] derived a model, using the self-consistent field approximation [26], to predict the effective thermal conductivity of monodisperse spherical capsules randomly distributed in a continuous matrix. This effort was motivated by the need to estimate the effective thermal conductivity of syntactic foam insulation. The geometry considered in the derivation consisted of a spherical volume of matrix material containing a concentric core–shell particle with volume fractions representative of the overall composite. The model accounted for contact resistance at the shell-matrix interface. An exact series solution of the heat conduction equation was obtained for the temperature distribution in each phase. In absence of contact resistance, the model can be expressed as [21],

$$k_{eff} = \frac{\Theta_N}{\Theta_D} k_m \tag{3}$$

Here, the numerator Θ_N and denominator Θ_D are expressed as [21],

$$\Theta_N = 2(1-\phi_{c+s})A + (1+2\phi_{c+s})B \quad \text{and} \\ \Theta_D = (2+\phi_{c+s})A + (1-\phi_{c+s})B \tag{4}$$

where the parameters A and B are given by [21],

$$A = \left(1+\frac{2}{\phi_{c/s}}\right) - \left(1-\frac{1}{\phi_{c/s}}\right)\frac{k_c}{k_s} \quad \text{and} \\ B = \left(2+\frac{1}{\phi_{c/s}}\right)\frac{k_c}{k_m} - 2\left(1-\frac{1}{\phi_{c/s}}\right)\frac{k_s}{k_m} \tag{5}$$

Here, ϕ_{c+s} is the volume fraction of the composite occupied by the capsule and $\phi_{c/s}$ is the volume fraction of the core with respect to the capsule. They are expressed as $\phi_{c+s} = (D_s/D_m)^3$ and $\phi_{c/s} = (D_c/D_s)^3$ where D_c, D_s, and D_m are the diameters of the core, shell, and matrix domains, respectively. The volume fraction of core–shell capsules ϕ_{c+s} can be written as $\phi_{c+s} = \phi_c + \phi_s$. Pal [12]