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Investigation of nanoparticle agglomeration on the effective thermal conductivity of a composite material

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ABSTRACT

The viability of organic phase change materials (PCMs), such as paraffin wax, for passive thermal management of portable electronics improves if the PCM overall bulk thermal conductivity is increased through the addition of highly conducting nanoparticles. Previous work has suggested the possibility of increasing the bulk thermal conductivity of composite materials through the controlled agglomeration of nanoparticles, yet no theoretical study has been performed to investigate the conditions under which the bulk thermal conductivity enhancement is achieved. Therefore, this study examines the influence of both spherical clustering and linear percolation network formation on the resultant bulk conductivity. This approach uses effective medium and percolation theories for unpercolated and percolated areas, respectively. Theoretical approaches are shown for spherical clustering and a 1-d conduction model of linear percolation networks, and finite element analysis (FEA) is used for a 2-d conduction model of linear percolation networks. The results for herringbone graphite nanofibers (HGNFs) in a paraffin matrix suggest that spherical clustering and linear agglomeration tend to reduce the bulk thermal conductivity, which agrees with experimental observations. However, linear percolation networks may enhance the effective thermal conductivity when large inclusion–matrix to inclusion–inclusion Kapitza resistance ratios are used.

1. Introduction

The complexity for effective thermal management of portable electronics increases with the increasing demand for higher power portable electronics and smaller electronic envelopes. Although active cooling techniques, such as fans and blowers, are extremely effective, they draw valuable power from the battery. Passive cooling techniques, such as thermosyphons, heat pipes and phase change materials (PCMs), require no additional energy. Ideally, a thermal management design system should cool the components inside the electronic device while keeping the outer shell at a comfortable handling temperature. Most processors in electronics have a maximum operational temperature of 85–90 °C, but ergonomically the outer casing should be kept below 45 °C for handheld electronics [1]. Fortunately, solid–liquid PCMs with a melting point near 50–60 °C can not only provide passive cooling that will protect processors from overheating, but PCM use also maintains a safe handling temperature. Therefore, this study considers the use of PCMs for thermal management.

One viable PCM candidate for passively cooling portable electronics is paraffin wax. Paraffin has a latent heat near 200 kJ/kg [2] and melt temperature in the range of 30–64 °C [2], [3]. Paraffin is also non-corrosive, which is important for electronic applications. However, paraffin has a thermal conductivity of roughly 0.25 W/m K [2] at room temperature, which inhibits the energy storage capability of the PCM due to a lack of thermal diffusion. Therefore, researchers have studied the addition of carbon nanoparticles that contain a thermal conductivity possibly greater than 2000 W/m K [4] to improve the thermal properties of PCMs [5–8]. Herringbone graphite nanofibers (HGNFs) are considered here as a less-expensive alternate to carbon nanotubes and graphene platelets for conductivity enhancement [5,6,8,9]. When produced with a 45° crease angle, the HGNFs have a predicted cross-plane thermal conductivity of 27 W/m K, a transverse thermal conductivity of 263 W/m K in the direction of the crease, and a transverse thermal conductivity of 1500 W/m K normal to the crease [10].

One major issue with nano-enhanced PCMs is controlling the nanoparticle dispersion in the liquid PCM state. Nanoparticles tend to agglomerate during the solid–liquid phase transition in PCMs, especially in paraffin [5,11,12]. As the liquid solidifies after a single thermal cycling period, clusters of nanoparticles are observed

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Nomenclature

D	inclusion diameter, m
f	inclusion volume fraction, dimensionless
f_c	percolation threshold, dimensionless
f_{ns}	inclusion volume fraction outside clusters, dimensionless
f_s	inclusion volume fraction within clusters, dimensionless
f_0	overall inclusion volume fraction, dimensionless
F	ratio of volume fraction of inclusions outside clusters to the overall volume fraction of inclusions, dimensionless
g	clustering density ratio, dimensionless
G	conductance, W/K
h_c	inclusion–inclusion contact conductance, W/m^2 K
k_b	effective bulk thermal conductivity, W/m K
$k_{b,0}$	effective bulk thermal conductivity in a composite with uniformly distributed inclusions, W/m K
k_e	effective local thermal conductivity, W/m K
k_i	inclusion thermal conductivity, W/m K
k_m	matrix thermal conductivity, W/m K
k_{ns}	effective thermal conductivity outside clusters, W/m K
k_{pe}	effective percolation backbone thermal conductivity, W/m K
k_1^c, k_2^c, k_3^c	adjusted inclusion conductivities used in effective medium model, W/m K
k_s	effective thermal conductivity within clusters, W/m K
L_d	decay length in inclusion distribution near the boundary, m
L_x	domain width, m
L_i	inclusion length, m
L_y	domain height, m
L_1, L_2, L_3	geometric factors in effective medium model, dimensionless
N	number of nodal temperatures along the top boundary, dimensionless
p	inclusion aspect ratio, dimensionless
q''	heat flux, W/m^2
r_{mesh}	mesh scaling parameter, dimensionless
R_b	inclusion–matrix thermal boundary resistance, m^2 K/W
T_{bot}	temperature at bottom of domain, K
T_{top}	temperature at top of domain, K
v	volume fraction of clusters, dimensionless
α	parameter used in Bruggeman model, dimensionless
β	percolation factor, dimensionless
β_1, β_2, β_3, γ	parameters used in effective medium model, dimensionless
δ	ratio of thermal conductivity inside clusters versus outside clusters, dimensionless
ε	small change in volume fraction or thermal conductivity ratio, dimensionless
Δf_e	variation of elemental inclusion volume fraction, dimensionless
ζ	geometric factor in percolated cylindrical inclusions method
χ	inclusion–inclusion contact Biot number, dimensionless

along the grain boundaries. This agglomeration may be mitigated by introducing high density polyethylene (HDPE) [12,13], but this approach also suppresses favorable Rayleigh–Benard convection currents. Therefore, the agglomeration of nanoparticles must be taken into consideration to be able to examine its influence on the overall thermal conductivity of a composite. Some studies show an improvement in thermal performance with the formation of percolation networks [11,14,15], yet others show a detrimental effect [12], so this influence is not conclusive. Therefore, this study introduces a theoretical means to predict this influence.

In order to predict the composite thermal conductivity of nano-enhanced PCMs, investigators have developed models based on approaches such as an effective medium theory (EMT) or a percolation-based theory. Traditionally, these models take into consideration the thermal boundary resistance (TBR), or Kapitza resistance, between the inclusion and the surrounding matrix [16–18]. However, the majority of these models focus on the influence of the inclusions on the effective thermal conductivity below the percolation threshold. An effective medium theory derived from the well-accepted model by Nan et al. [18] is commonly used to calculate the effective thermal conductivity in this study when the local volume fraction, f, falls below the percolation threshold, which may be defined as [19]

$$f_c = \frac{\pi^2}{16p} \tag{1}$$

where $p = L_i/D$ is the inclusion aspect ratio, where L_i and D are the inclusion length and diameter, respectively. It should be noted that Eq. (1) is one of several percolation threshold models for a composite material containing uniformly distributed and oriented cylindrical or prolate inclusions, and that other models have been proposed [20]. In addition, studies by Gao and Li [21] and Wang et al. [22] have specifically observed the dependence of particle shape on the percolation threshold.

Investigators have also developed approaches, including Monte Carlo (MC) [23] and Lattice-Boltzmann [24] simulations, to predict the thermal conductivity above the percolation threshold. The MC and Lattice-Boltzmann simulations are time consuming and not as general as analytical expressions to predict the effective composite thermal conductivity. Therefore, Wemhoff [19] created a theoretical model for a composite material containing a random distribution of straight cylindrical inclusions above the percolation threshold.

Experiments [11] have indicated a deviation in the bulk composite thermal conductivity when local nanoparticle agglomeration occurs, which leads to the need to model the influence of agglomeration on bulk conductivity. Some studies, such as that by Prasher et al. [14] and Reinecke et al. [15], have successfully combined existing theories such as the Bruggeman [25] and Maxwell [26] models in order to model composites containing percolated chains of spherical particles. This study continues this line of work by, for the first time, providing parameters that directly indicate the level of agglomeration and deducing how they influence the overall bulk composite thermal conductivity, and then applying the method on a composite material with cylindrical inclusions. Furthermore, the study applies a sensitivity analysis to the results to suggest the conditions under which agglomeration may be beneficial for bulk thermal conductivity.

This study explores three separate models of agglomeration of inclusions with the goal of establishing under which conditions agglomeration enhances or reduces the composite's bulk thermal conductivity. The first model explores spherical clustering of inclusions, whereas the remaining two models (1-d resistor network and 2-d FEA) explore the formation of linear percolation networks, keeping in mind that in reality the composite material will contain