

Analyzing theoretical models for predicting thermal conductivity of composite materials for LED heat sink applications

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Abstract

This study investigated the thermal properties of composite materials with respect to their potential to be used for thermal management in light-emitting diode (LED) application. The effect of filler particles size and volume fraction in the composite material thermal conductivity was investigated. Experimental data for copper-filled epoxy composites with copper particles of average size of 5- μm and 150- μm were compared to three theoretical models: Maxwell, Lewis-Nielsen, and percolation theory. Our study shows that all three theoretical models underestimate the thermal conductivity compared to the experimental results for composites with 150- μm copper particles up to 0.54 filler volume fraction. In the filler volume fraction region that is of interest to LED thermal management applications, between 0.40 and 0.54, the percolation model underestimates the experimental data by $\sim 40\%$, and the Lewis-Nielsen model underestimates by $\sim 67\%$. However, the theoretical models show closer agreement with the experimental data for composites with 5- μm copper particles up to 0.51 filler volume fraction. The percolation model overestimates the experimental data by 18% and the Lewis-Nielsen model underestimates by 12%. According to the estimated values from the theoretical models, there is no filler particle size effect on the composite thermal conductivity. However, the experimental data show that the composites with 5- μm copper particles tend to have lower thermal conductivity than the composites with 150- μm copper particles.

Introduction

The study presented here investigated the potential of composite materials to be used for LED heat sink applications. At present, LED is the preferred technology for many lighting applications due its lower energy demand and lower maintenance cost compared to other traditional lighting technologies. LED luminaire prices are continuously reducing, and manufacturers are looking for lower cost components to integrate with their luminaires to remain competitive in the market. The LED heat sink is typically one of the higher cost components in a luminaire and it is usually made of aluminum, which has high thermal conductivity ($\sim 200 \text{ W}/(\text{m}\cdot\text{K})$). High thermal conductivity heat sinks are used to keep the junction temperature of LEDs low to improve their lifetime and reliability [1]. Previous studies have shown that materials with thermal conductivity of 20-40 $\text{ W}/(\text{m}\cdot\text{K})$ may be sufficient to keep the LED junction temperature low [2, 3]. Therefore, using aluminum for LED heat sinks may be excessive. One potential heat sink cost reduction approach is using composite polymer materials, which could lower material costs.

Composite polymer materials can be created by adding fillers of higher thermal conductivity to polymer materials, which results in composite materials with higher thermal conductivity than that of polymers alone. For such materials to be viable in LED thermal management applications, their thermal conductivity needs to be sufficient to keep the LED junction temperature low.

Understanding what factors affect the thermal conductivity of composite materials and having the ability to predict it are crucial to designing and manufacturing such materials.

Several theoretical models have attempted to explain the behavior of thermal conductivity in composite materials. In the late 19th century, Maxwell was one of the first scientists to provide an analytical solution to the effective conductivity of a heterogenic medium, which was part of his famous *Treatise on electricity and magnetism* [4]. He considered spherical particles of thermal conductivity k_f embedded in a continuous matrix of thermal conductivity k_m at small enough volume fractions so that the spheres were significantly farther from each other compared to their size [5]. Maxwell's formula for the effective thermal conductivity of a composite material is shown below:

$$k_{eff} = k_m \frac{k_f + 2k_m + 2\varphi(k_f - k_m)}{k_f + 2k_m - \varphi(k_f - k_m)} \quad \text{Eq. 1}$$

where k_{eff} is the composite thermal conductivity and φ is the filler volume fraction. There have been various modifications to this model to include the effect of filler particle shape, the interfacial thermal resistance, and more than one type of filler in the host material, but they all have one thing in common, according to the literature: they are accurate only at low filler volume fractions (up to about 25%) [6].

Another model that is commonly encountered in the literature regarding the thermal conductivity of composite materials is the Lewis-Nielsen model. Equations for this model are borrowed from the theory of the elastic moduli of composite materials. Predictions of this model are generally accurate up to about 40% filler volume fraction, but at higher filler concentrations the predicted effective thermal conductivity increases rapidly and becomes negative once the maximum filler packing fraction is achieved [6, 7]. The maximum filler packing fraction is a parameter that is assigned to the composite material depending on the pattern of packing. The advantage of this model is that it allows for a range of particle shapes. The Lewis-Nielsen equations for the thermal conductivity of two-phase composite materials are:

$$\frac{k_{eff}}{k_m} = \frac{1 + AB\varphi}{1 - B\psi\varphi} \quad \text{Eq. 2}$$

$$A = k_E - 1 \quad \text{Eq. 3}$$

$$B = \frac{k_f/k_m - 1}{k_f/k_m + A} \quad \text{Eq. 4}$$

$$\psi = 1 + \left(\frac{1 - \varphi_m}{\varphi_m^2} \right) \varphi \quad \text{Eq. 5}$$

k_{eff} is again the composite thermal conductivity, k_m and k_f are the thermal conductivities of the matrix and the filler, and φ is the filler volume fraction. Constant A depends on the shape of the dispersed particles, their aspect ratio, and how they are oriented with respect to the direction of heat flow [8]. It is dependent upon the generalized Einstein coefficient k_E and its values are available in tables for many shapes [8]. Constant B takes into account the ratio of the components' thermal conductivities. Constant ψ is determined by the maximum packing fraction of the filler particles φ_m , which is dependent upon their packing pattern and is given in tables as well [8].

One of the more recent theories for thermal conductivity of composite materials is based on the idea that when increasing the amount of filler per unit volume, one eventually reaches a point at which filler particles begin to come into contact [6]. Increasing the amount of filler further creates chains of connected conductive particles, which cause a significant increase in the composite thermal conductivity. This principle is shown in Figure 1 where high-conductivity particles, forming a continuous path from source to sink, are shown in black. Low thermal conductivity particles are shown in white and high-thermal conductivity particles that do not form continuous chains are hatched. This percolation phenomenon becomes visible when the effective thermal conductivity plotted against the filler volume fraction changes its slope from shallow to very steep. The point of filler concentration at which this occurs is called percolation threshold [9].

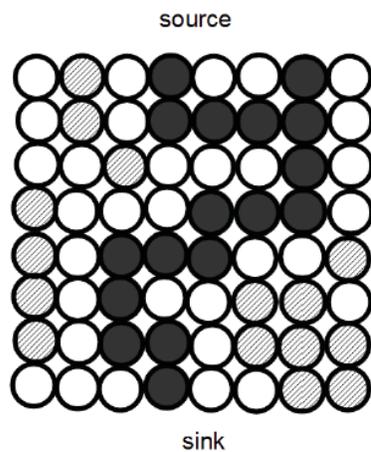


Figure 1 (adopted from [6]). A diagram of thermal conductivity enhancement in a composite material, due to percolation.

A probabilistic numerical model based on the resistor-network approach that takes into account the percolation principle and allows for prediction of the thermal conductivity of composite materials was proposed by Phelan et al. [10]. The composite material is assumed to be made of cubic blocks whose sides are equal to the filler particle size. The block may be either a matrix block or a filler particle block. A grid with a random distribution of filler particles, based on their volume fraction, is generated and the effective thermal conductivity is calculated based on the thermal conductivities of the host and filler materials and the dimensions of the grid. This procedure is repeated multiple times and the average thermal conductivity is taken as the composite thermal conductivity to compensate for the random generation of the grid. Although this model requires significant computational power, it is still rather simple compared to other more complex 3-dimensional numerical simulations that can account for various filler packing patterns, multimodal particle size distributions, and particle shapes other than spherical [11-18]. According to Pietrak et al., and Devpura et al., the Phelan model is also more accurate beyond the percolation threshold than the classical Maxwell and Lewis-Nielsen models that either underestimate thermal conductivity or provide non-real negative thermal conductivity in that region [6, 7].

Various experimental studies have been conducted to investigate how the thermal conductivity of composite materials changes with various parameters. Increasing filler volume fraction,

considering the filler has higher thermal conductivity than the host material, generally increases the composite thermal conductivity. However, if the filler particle size is too low, the interfacial thermal resistance may become critical and the overall thermal conductivity will decrease with filler volume fraction [19]. It has also been shown that the composite thermal conductivity increases with filler and host material thermal conductivities [20,21] and with filler particle size [22-27]. Increasing the aspect ratio of filler particles [20,28], performing filler surface treatment [29-32], and usage of hybrid filler have also been shown to help increase the composite thermal conductivity [33,34].

The goal of the study presented here was to identify and verify a model that can accurately predict the thermal conductivity of a composite material.

Methods

Theoretical predictions

The three theoretical models discussed in the introduction were used for the theoretical predictions of the thermal conductivity in this study: Maxwell, Lewis-Nielsen, and percolation models.

For the Maxwell model predictions, the thermal conductivities of filler and host materials and the filler volume fractions were needed. For the Lewis-Nielsen model predictions, the shape factor A was chosen to be 1.5 corresponding to spherical particles. The maximum packing fraction of filler particles φ_m was chosen to be 0.601 for random loose type of packing [8]. For both of these models, the known values were plugged into the equations and the composite thermal conductivity was calculated.

For percolation theory simulations, a more complicated process was necessary. The commercial programming software Matlab[®] by MathWorks[®] was used to create a model which simulated the resistor-network approach proposed by Phelan et al. [10]. First, an m by n grid with random distribution of filler particles is generated, based on the filler volume fraction, filler size (l), which was the size of the host material particles as well, and boundary temperatures $T_1 = 1K$ and $T_0 = 0K$ at the top and bottom of the grid, an example of which is shown in Figure 2. The length of the grid is $L = n \cdot l$ and the height of the grid is $H = m \cdot l$, while the ratio L/H is set to be 2 to eliminate edge effects.

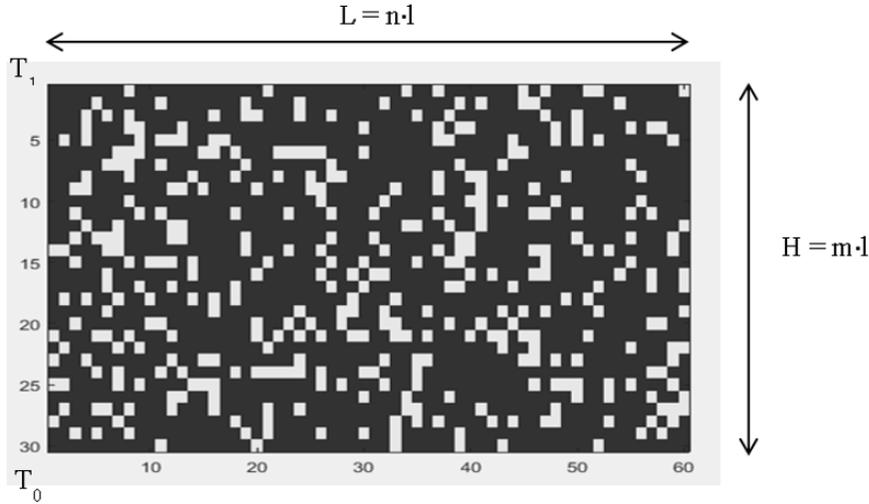


Figure 2. An example of a 30×60 particle grid generated using Matlab®, with gray squares representing the filler particles and black squares representing the host material particles.

After that, four resistances corresponding to the top ($R_{ij,T}$), bottom ($R_{ij,B}$), right ($R_{ij,R}$), and left ($R_{ij,L}$) sides of each particle are assigned depending on the particle's and adjacent particle's thermal conductivities and filler contact conductance (h_c) that is introduced only when the particle is a filler particle.

A series resistance approach is taken for each particle:

$$R_{ij,R} = R_{i,j} + R_{i,j+1} \quad \text{Eq. 6}$$

Assuming the (i, j) particle is a filler particle of thermal conductivity k_f and the (i, j+1) particle is a matrix particle of thermal conductivity k_m (Figure 3):

$$R_{i,j} = \frac{1}{h_c \cdot l} + \frac{1}{2 \cdot k_f} \quad \text{Eq. 7}$$

$$R_{i,j+1} = \frac{1}{2 \cdot k_m} \quad \text{Eq. 8}$$

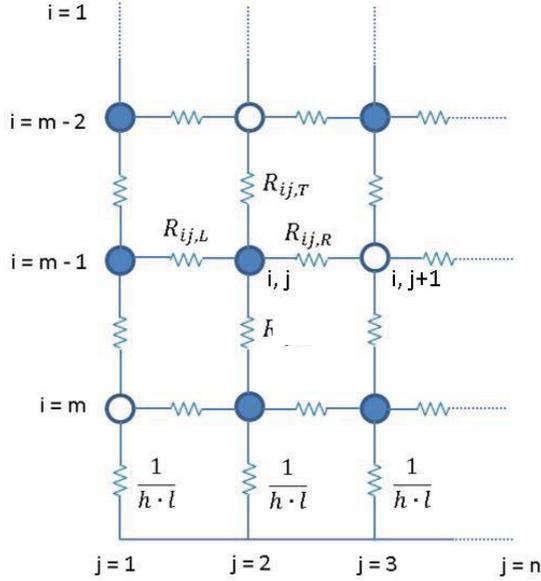


Figure 3 (adopted from [10]). Lower left-hand portion of the thermal resistor network. Filled circles represent filler particles and empty circles represent host material particles.

If the $(i, j+1)$ particle is a filler particle, the filler contact conductance is introduced to $R_{i,j+1}$ as well. For the simulations in this study, the filler contact conductance was assumed to be infinity; thus, the filler contact resistance was ignored, but its effects are discussed later in the paper.

The boundary conditions on the left and right sides of the grid are set to be insulating by making the left-side resistances of the leftmost particles equal to their right-side resistances and conversely, so that no heat flows to the sides. For the temperatures at the top and bottom sides to be uniform, the thermal conductance at those boundaries is assumed to be infinity. The thermal resistance at those boundaries is set to $1/(h \cdot l)$ with h equal to 10^{30} W/(m²·K).

After all the resistances around each particle are set, a system of linear equations is created by setting the net heat flow through each particle to zero, assuming a steady-state condition:

$$\frac{T_{i-1,j} - T_{i,j}}{R_{i,j,T}} + \frac{T_{i,j+1} - T_{i,j}}{R_{i,j,R}} + \frac{T_{i+1,j} - T_{i,j}}{R_{i,j,B}} + \frac{T_{i,j-1} - T_{i,j}}{R_{i,j,L}} = 0 \quad \text{Eq. 8}$$

The system of equations is solved by matrix inversion and the temperature at each node is known. Assuming the thermal resistances at the boundaries and through the composite are in series, the total thermal resistance is:

$$R_{tot} = \frac{1}{2 \cdot h \cdot L} + \frac{H}{k_{eff} \cdot L} \quad \text{Eq. 9}$$

where k_{eff} is the effective thermal conductivity of the sample, which can be expressed as:

$$k_{eff} = \frac{H}{L \cdot R_{tot} - 2/h} \quad \text{Eq. 10}$$

And the total thermal resistance can be found by using the following formula:

$$R_{tot} = \frac{T_1 - T_0}{q} \quad \text{Eq. 11}$$

And q can be found by summing up all the individual heat flows at either the upper or lower boundaries. After the effective thermal conductivity is found for one random configuration of filler particles, the procedure is repeated multiple times and the mean value of thermal conductivity is reported, to account for the probabilistic nature of the procedure. Phelan et. al used 200 iterations, but after conducting a sensitivity analysis, 50 iterations were found to result in a less than 1% error compared to the 200 iterations. Therefore, 50 iterations were performed to reduce the time requirement for simulations.

Experimental setup

Copper particles with the average size of 5 μm and 150 μm were chosen to prepare the composite materials with epoxy resin as a host material. Copper powder was acquired from the Goodfellow Corporation, and two-part epoxy resin and catalyst 20-3302 were acquired from Epoxies, Etc.

The composite samples were disks of 19.1 mm in diameter and thickness that varied from 0.5 mm to 5 mm. Composites were prepared by mixing copper powder with epoxy resin and catalyst in a silicone mold. The mass of each component was recorded. The samples were cured in an oven for 2 hours at 52°C. They were then polished with sandpaper to ensure flat surfaces. The volume fraction of the copper particles was calculated using the following formula:

$$\frac{V_c}{V} = \frac{m_c}{m_c + \frac{\rho_c}{\rho_e} \cdot m_e} \quad \text{Eq. 12}$$

Where (V_c/V) is the volume fraction of copper in the composite, m_c is the mass of copper, m_e is the mass of epoxy, and ρ_c and ρ_e are the densities of copper and epoxy, respectively.

The experimental setup used for measuring the thermal conductivity of the composite materials is illustrated below in Figure 4.

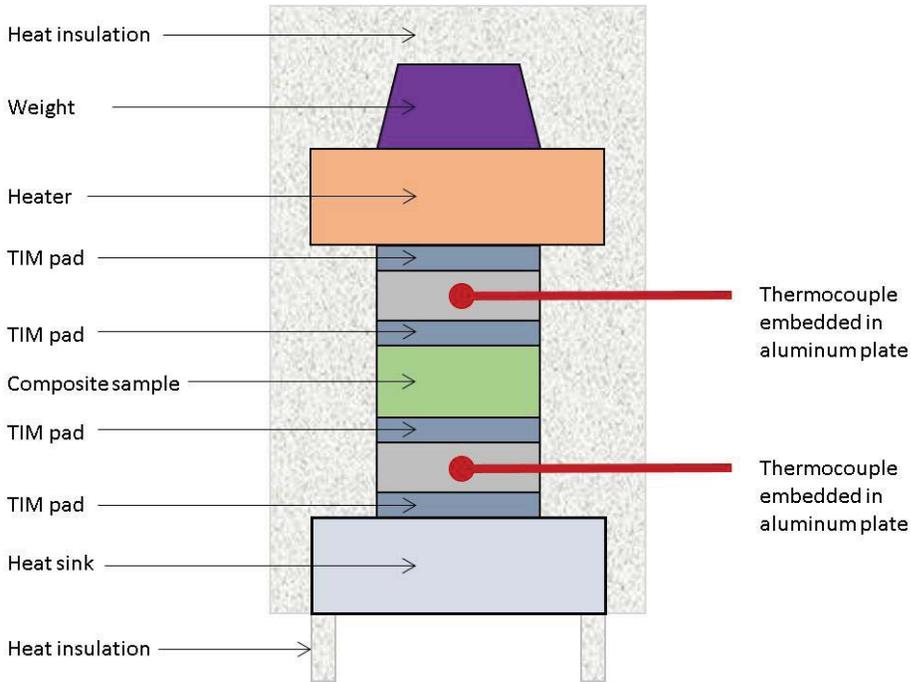


Figure 4. Schematic diagram of the thermal conductivity measurement experimental setup.

The heater was made of aluminum with a resistive heating element embedded in it. The sample was attached to the heater with a thermal interface material (TIM) pad. Thermal insulation was used on the sides of the samples to ensure minimum heat dissipation through the sides. The temperature gradient was measured along the test sample thickness using two calibrated thermocouples, one embedded in the heater very close to the top of the sample, and one embedded in the heat sink very close to the bottom of the sample. A weight of 500 g was placed on top of the setup to maintain a constant load. All thermocouple cables were connected to a thermocouple data logger, which was connected to a computer for data acquisition.

To calculate the thermal conductivity of the composites, samples of three thicknesses for each copper volume fraction were prepared. Each sample was subjected to two temperature gradients by varying the voltage applied to the heater and letting the temperature stabilize. Fourier's law was used for calculations:

$$\dot{q} = \frac{k \cdot A \cdot \Delta T}{l} \quad \text{Eq. 13}$$

In this equation, \dot{q} is the steady-state rate of heat transfer, k is the isotropic bulk thermal conductivity of the medium, A is the cross-sectional area of heat transfer, ΔT is the temperature differential driving the heat transfer, and l is the distance between the locations where the ΔT is measured.

$\dot{q}/\Delta T$ was calculated from the measurements of the power applied to the heater and the temperature gradient measured by the thermocouples. The heat losses were accounted for by first running the calibration samples of known thermal conductivity and using the parallel thermal resistance approach schematically (Figure 5).

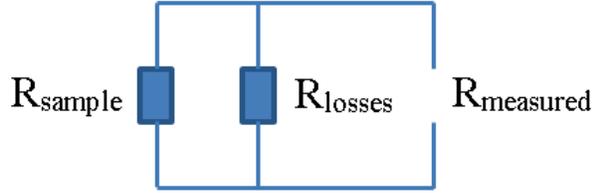


Figure 5. Schematic diagram of parallel thermal resistance approach to thermal conductivity calculations.

In other words:

$$\frac{1}{R_{measured}} = \dot{q}/\Delta T = \frac{1}{R_{losses}} + \frac{1}{R_{sample}} \quad \text{Eq. 14}$$

$$R_{sample} = \frac{l}{k \cdot A} = \left(\frac{1}{R_{measured}} - \frac{1}{R_{losses}} \right)^{-1} \quad \text{Eq. 15}$$

$\left(\frac{1}{R_{measured}} - \frac{1}{R_{losses}} \right)^{-1}$ was plotted against l for three different thicknesses of copper samples (Figure 6). The thermal conductivity was found by dividing the inverse slope of the line by the average surface area of the samples.

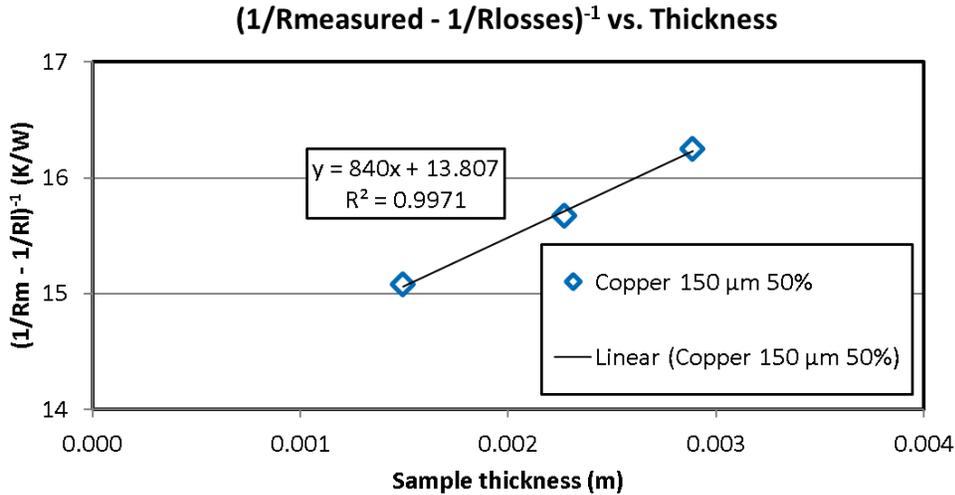


Figure 6. Example of thermal conductivity calculations graph for 150- μ m 50% by volume copper-filled epoxy samples.

Results

Simulations

Predictions of the theoretical models for copper-filled epoxy samples are summarized in Figure 7, where the host material and copper thermal conductivity are taken to be 0.4 W/(m·K) and 400 W/(m·K) respectively, and the size of the filler particles is taken to be 100 μ m for percolation

simulations. The error bars on the percolation simulation graph show the standard deviation of 50 iterations.

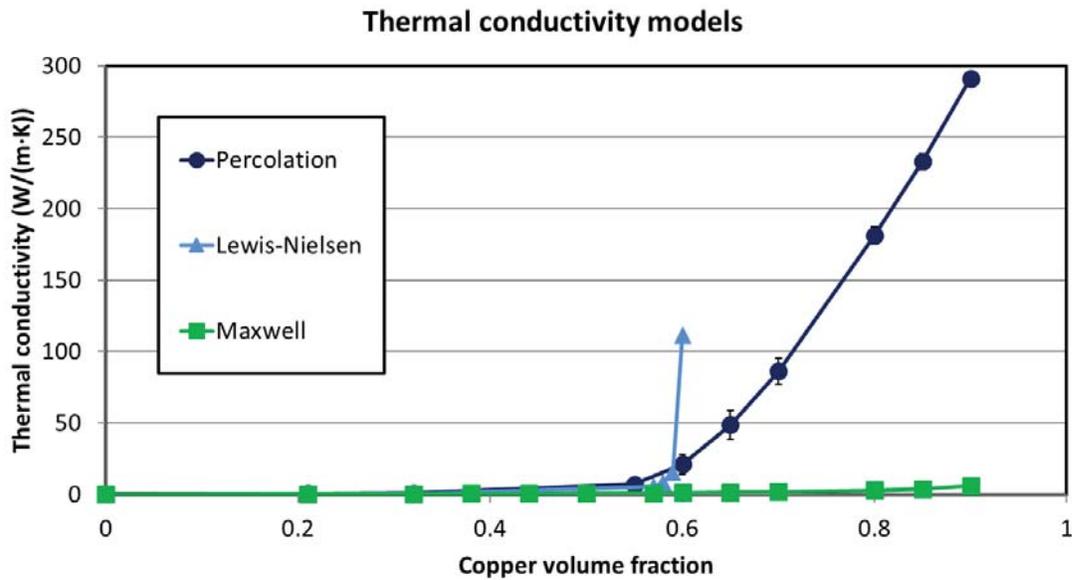


Figure 7. Thermal conductivity against copper volume fraction based on different theoretical models' predictions.

It can be seen from Figure 7 that according to the Maxwell model, the composite material thermal conductivity never increases beyond a low value comparable to that of the host material due to disregard of thermal interaction between particles. Lewis-Nielsen model predicts a rapid increase in thermal conductivity when the filler volume fraction approaches the maximum packing fraction close to 0.60. However, since 0.601 is the maximum volume fraction up to which the model predicts the thermal conductivity, the model cannot be used for composite materials with higher filler volume fractions. On the other hand, while the percolation model predicts a similar rapid increase in thermal conductivity around 0.60 volume fraction, the increase continues beyond that volume fraction, according to the simulations. Since for the composite materials to be an option for LED heat sinks the effective thermal conductivity needs to be around 20-40 W/(m·K), the region of 0.40-0.60 filler volume fraction, where the thermal conductivity increases from about 1 W/(m·K) to 25 W/(m·K), is of interest.

Experiment

The experimental data for copper-filled epoxy samples is shown against the theoretical predictions of all three models in Figure 8, where horizontal error bars represent error due to preparation of samples and vertical error bars show the variability of thermal conductivity values between the samples.

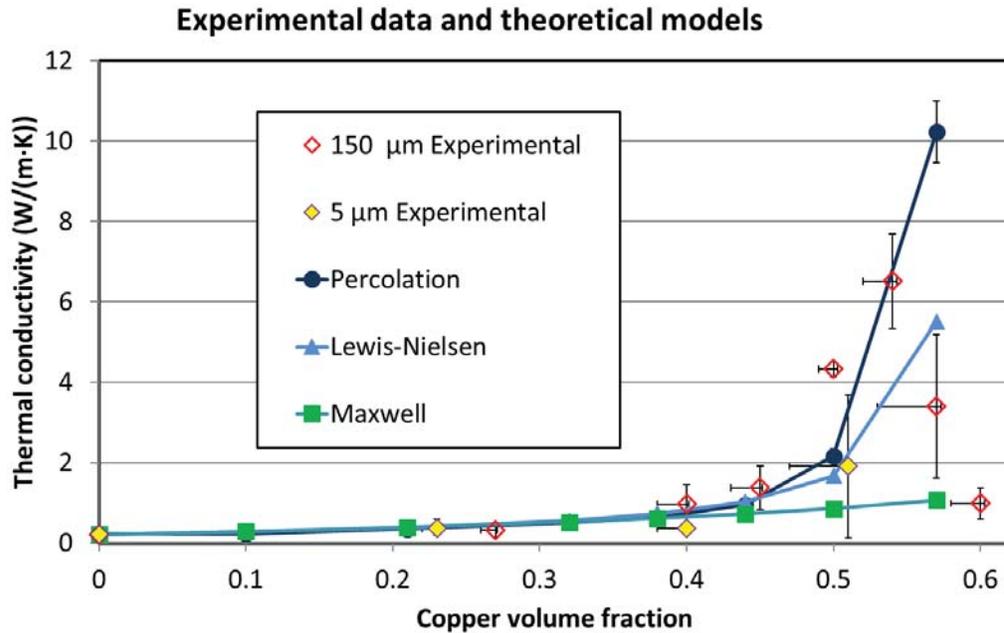


Figure 8. Experimental and theoretical values for thermal conductivity of copper-filled composites as a function of copper volume fraction.

Discussion

Plotting theoretical predictions against experimental data, as shown in Figure 9, demonstrates that all three theoretical models underestimate the thermal conductivity compared to the experimental results for composites with 150- μm copper particles up to 0.54 filler volume fraction. In the filler volume fraction region that is of interest to LED thermal management applications, between 0.40 and 0.54 filler volume fraction shown in Figure 9, the percolation model underestimates the experimental data by $\sim 40\%$, and the Lewis-Nielsen model underestimates experimental data by $\sim 67\%$. However, the theoretical models show closer agreement with the experimental data for composites with 5- μm copper particles up to 0.51 filler volume fraction (Figure 10). The percolation model overestimates the experimental data by 18% and the Lewis-Nielsen model underestimates by 12%. According to the estimated values from the theoretical models, there is no filler particle size effect on the composite thermal conductivity. However, the experimental data show that the composites with 5- μm copper particles tend to have lower thermal conductivity than the composites with 150- μm copper particles.

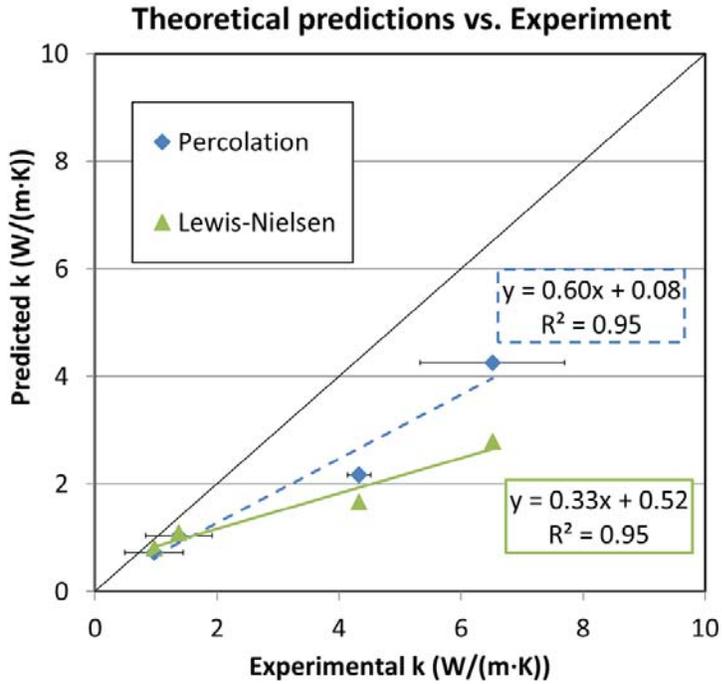


Figure 9. Comparison of the theoretical predictions to the experimental results for composites with 150- μm copper particles in the copper volume fraction range between 0.40 and 0.54.

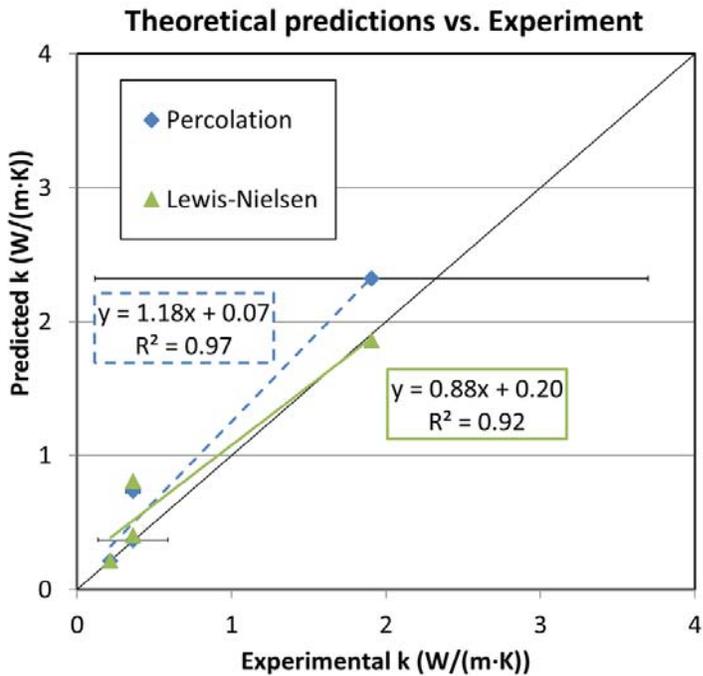


Figure 10. Comparison of the theoretical predictions to the experimental results for composites with 5- μm copper particles in the copper volume fraction range between 0 and 0.51.

The samples with filler volume fraction of 0.57 and 0.60 do not follow the predicted trend of increasing thermal conductivity, which may be due to several factors. One possible reason may be that at higher volume fractions, the material becomes very thick and filler contact resistance with the host material becomes large, which significantly reduces the overall thermal conductivity. The percolation model used in this study allows for accounting of filler contact resistance that was assumed to be infinite for the previous simulations. Introducing this parameter to the simulations at 0.57 volume fraction brings the thermal conductivity value of the material down (Figure 11).

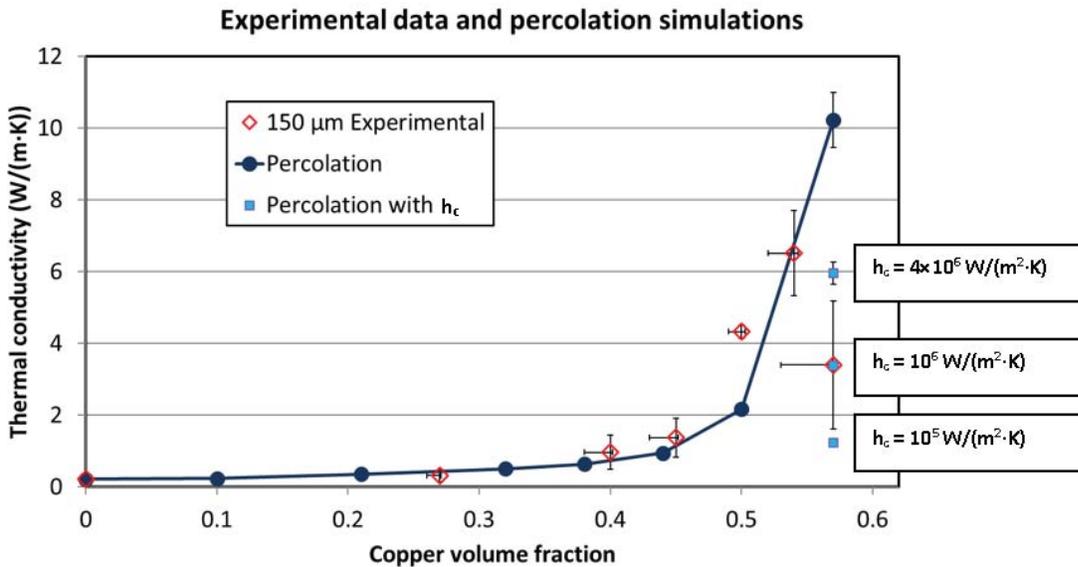


Figure 11. Experimental and percolation model data for copper-filled epoxy samples up to 0.57 filler volume fraction and percolation predictions with the inclusion of the filler contact thermal resistance at 0.57 filler volume fraction.

Another reason may be sedimentation of copper particles at higher volume fractions. If copper particles sediment during the preparation of the samples, few or no heat paths form throughout the sample, which results in reduction of the composite material thermal conductivity.

Lastly, higher volume fraction composite solutions showed higher viscosity before curing and had a higher chance of trapped air bubbles. During the curing process due to buoyancy these air bubbles would be trapped closer to the top of the sample. In the experiment, this side of the samples was always oriented away from the heater.

Finite-element analysis of a computer-aided design model with similar dimensions to the composite material samples used in the study revealed that the air gap spanning the circular cross section of the sample did reduce the thermal conductivity. It also showed the location of this air gap also affected the thermal conductivity. The air gap further along the axial direction of the sample away from the heater resulted in a lower thermal conductivity compared to the air gap closer to the heater.

Since preparation of higher filler volume fraction samples was an issue when using copper as the filler material, other materials were examined for their potential to be better fillers for achieving higher filler volume fractions. Although copper has very high thermal conductivity, it has very

high density as well. Therefore, a very high weight fraction of copper (>0.90) is needed to achieve ~ 0.60 volume fraction (Figure 12). Aluminum, with high thermal conductivity of ~ 200 W/(m·K) could be an alternative to copper, since only 0.79 weight fraction is sufficient to achieve 0.60 volume fraction, which may potentially make the preparation of samples easier.

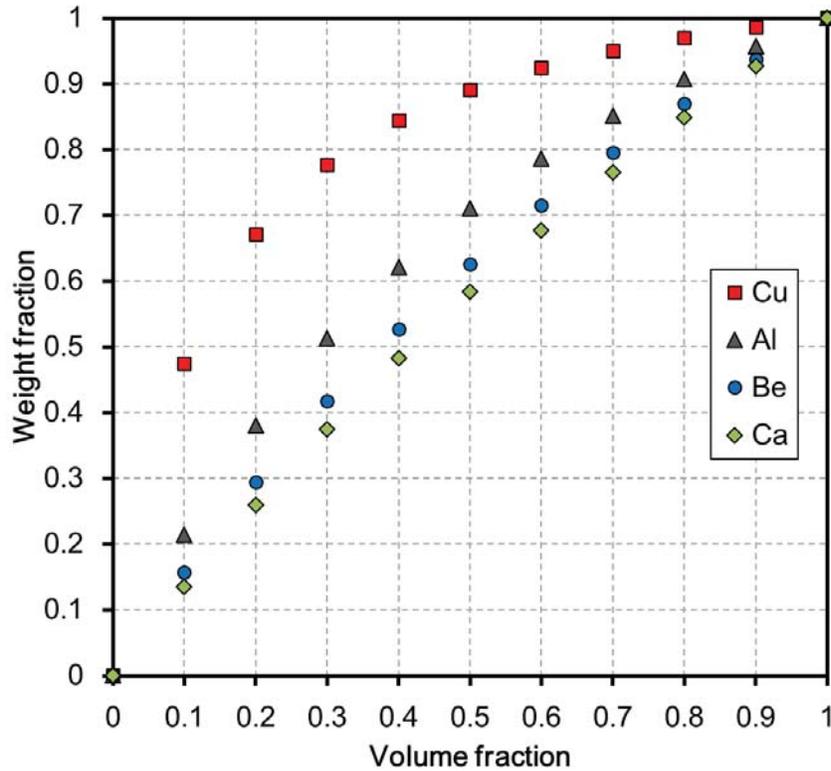


Figure 12. Weight fraction of copper (Cu), aluminum (Al), beryllium (Be), and calcium (Ca) against their volume fraction in epoxy.

One of the main limitations of the percolation model used in this study is that it is two-dimensional, while there may exist variation in filler particles distribution along the third dimension as well. Similar to the resistor-network approach followed in this study, other percolation models take the third dimension into consideration, but these were not investigated in this study.

Manual preparation of samples leading to large dispersions and limitations on preparing higher concentration composite samples are other limitations of this study.

Conclusions

Based on our experimental data, none of the theories examined predicted the thermal conductivity of copper-filled epoxy composite materials with 150- μm copper particles well in the region of interest to LED heat sink manufacturing. For composite materials with 5- μm copper particles, the Lewis-Nielsen model was the most accurate at predicting the effective thermal conductivity up to 0.51 filler volume fraction, underestimating it on average by 12%. The percolation model overestimated the thermal conductivity in this region by 18% and the Maxwell model underestimated by 70%. The models' accuracy beyond that filler volume fraction for

composite materials with 5- μm copper particles was not analyzed due to inability to make samples of higher filler volume fraction.

Although no filler particle size effect on the composite thermal conductivity was predicted by any of the models, the experimental data show that composites with 5- μm copper particles tend to have lower thermal conductivity than composites with 150- μm copper particles.

Overall, the inability to make samples of high enough filler volume fraction, at which the desired 10-20 W/(m \cdot K) for LED heat sinks were predicted to be achieved, hindered identification of which model is the most accurate in that region.

Using a different filler material with lower density may allow for preparation of samples of higher filler volume fraction than were achieved in this study.

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