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Experimental and numerical study of the effective thermal conductivity of nano composites with thermal boundary resistance

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ABSTRACT

The thermal interface resistance at the macro scale is mainly described by the physical gap between two (inter) faces and constriction resistance due to this gap. The small gaps and surface geometry mismatch between the two material faces makes up the majority of thermal interface resistance (R_c) at the macro scale. There are various models to predict R_c at macro scale. Although R_c represents thermal resistance accurately for macro size contacts between two metals, it is neither suitable nor accurate to describe interface resistance of a modern composite Thermal Interface Material (TIM) containing micron to nano-sized particles. The thermal discontinuity at a perfectly bonded interface of two dissimilar materials is termed as thermal boundary resistance (R_b) or Kapitza resistance. It is necessary to understand feasibility of using nanoparticles in composite TIM by having better understanding of thermal boundary resistance at that scale. The phenomenon of thermal boundary resistance is an inherent material property and arises due to fundamental mechanisms of thermal transport. For metal–matrix particulate composites, R_b plays a more important role than R_c. The free flowing nature of the polymer would eliminate most of the gaps between the two materials at their interface. This means almost all of the thermal resistance at particle/matrix interface would occur due to R_b. Here, the thermal boundary resistance for silica nanoparticles embedded inside epoxy resin is studied. The bulk conductivity of the sample is measured, and R_b is back calculated using the Hasselman–Johnson's (H–J) equation. The numerical validation of the equation is also presented, including extrapolation study to predict effective conductivity of the nanocomposite TIM.

1. Introduction to TIM and thermal boundary resistance

The materials used between electronics components (e.g., between silicon chip and a heat sink) are designated as Thermal Interface Materials (TIMs). There are various types of TIMs available in the market, which are normally in the form of elastomers or gels. These materials are electrically insulative but have higher thermal conductivity than polymeric resin. The elastomers are useful in applications where leakage is a main concern. The gel type materials cannot be used effectively for thick TIMs, and they do not possess a high electrical break down voltage. For the high performance embedded electronics, elastomers are more suitable to achieve desirable thickness in order to dissipate the heat.

Both, pure silicon and epoxy based materials, possess low thermal conductivity and are normally mixed with high thermally conductive particles like copper, alumina or diamond to achieve better effective conductivity. There are various theoretical models to predict the thermal conductivity of such particulate composites [1–4]. The rule of mixtures has been used in the past to predict the composite thermal conductivity but it does not yield accurate results. The micro mechanics type approaches [2] are able to better predict the thermal conductivity of composite media, but even then there have been some discrepancies between predictions and experimental results. One of the reasons affecting the accuracy of the prediction involves consideration of thermal interface (or thermal boundary) resistance. This piece of information concerning the thermal resistance across the interface between the particle and host medium is required in all micro mechanics models. In an ideal situation, the interfacial contact between the particle and host medium would cause little temperature discontinuity. However, due to imperfect particle-matrix interfaces, a temperature discontinuity exists for steady state heat transfer.

The thermal interface resistance at the macro scale is mainly described by the physical gap between two interfaces. Fig. 1 shows a typical interface between two materials at macro scale.

The small gaps between the two material faces makes up the majority of thermal interface resistance at the macro scale. Thus, most of the studies have been focused on characterizing and modeling effect of surface geometry and material properties to thermal interface resistance. This resistance is more widely known as

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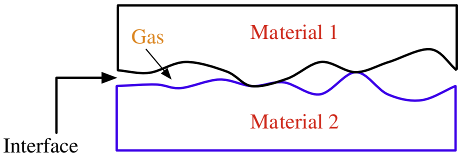

Fig. 1. Contact between two materials at macro scale.

thermal contact resistance, also represented with R_c [5,6]. These models predict thermal resistance R_c for two given materials by utilizing their bulk thermo mechanical properties. These theories predict total nominal contact area under thermal and mechanical loads. The deformation of contact surfaces under these loads can be modeled in terms of various properties like surface roughness, microhardness, young's modulus, yield strength, conductivity etc. The detailed analyses of these theories are available in the literature [5,7,8,9,10].

Although R_c represents thermal resistance accurately for macro size contacts between two metals, it is not suitable to describe interface resistance of particles in modern particulate composite TIMs. The particles inside recently available TIMs are micrometer size. In an effort to further increase surface area, this particle size is approaching nano scale. At this small scale, R_c does not accurately predict thermal interface behavior, as it is very difficult to characterize the surface topography. Also, phonon velocity (lattice vibrations) plays a very important role as the interface dimension approaches nano scale.

Kapitza reported a thermal discontinuity between helium and solids at very low temperature [11]. This was then investigated further for other materials in perfect contact, and similar results were obtained at low and high temperatures. This thermal discontinuity at perfectly bonded interface of two dissimilar materials is termed as thermal boundary resistance (R_b) or Kapitza resistance. The macroscopic assumptions that thermal discontinuity is only due to gaps and surface geometry leads to substantial error in determining interface thermal properties at micron and nano scale. The thermal boundary resistance is an inherent material property and arises due to fundamental mechanism of thermal transport. For metal-matrix particulate composites, R_b plays more important role than R_c as the free flowing nature of the polymer would eliminate most of the gaps between the two materials at their interface. This means almost all of the thermal resistance at particle/matrix interface would occur due to R_b.

Due to uncertainties involved in many numerical MD (Molecular Dynamics) simulations and theoretical calculations for R_b [11,12,13], it is best advised to use empirical or experimental approach at this stage. The bulk conductivity of particulate composite can be experimentally measured, and using one of the micromechanics equations [15], thermal boundary resistance at the matrix-particle interface can be back calculated. The accuracy of micromechanics equation can be verified with a finite element model.

2. Introduction to the Hasselman–Johnson equation for composite conductivity prediction

From [14,15] the thermal conductivity, K_{eff}, of a particulate composite can be given as:

$$\frac{K_{eff}}{K_m} = \frac{[K_p(1+2\alpha)+2K_m]+2v_f[K_p(1-\alpha)-K_m]}{[K_p(1+2\alpha)+2K_m]-v_f[K_p(1-\alpha)-K_m]} \qquad (0)$$

where, subscript m and p represent matrix and particle respectively, v_f is the volume fraction of particles and dimensionless parameter α is given by

$$\alpha = \frac{R_b K_m}{a} \qquad (1)$$

where, a is the radius of the particles and R_b is a thermal boundary resistance.From the Eq. (1), we can calculate the critical particle size at which composite K is the same as K_m:

$$a_0 = \frac{R_b K_m}{1-\frac{K_m}{K_p}} \qquad (2)$$

As an example, consider a particulate TIM consisting of 401 W/m-K and matrix conductivity of 0.143 W/m K. The boundary resistance is assumed to be 0.0148 cm^2 K W^{-1}. This gives critical particle size a_0 as 2e−7 m. This means that adding particles of diameter less than 2e−7 m (0.2 micron) will make the thermal conductivity of TIM lower than that of the matrix. This particle size represents the threshold for this given composite TIM. Fig. 2 shows the variation of relative composite thermal conductivity with respect to particle size for two volume fractions of copper in silicone elastomer. This figure can be used as a guideline for designing particulate composite TIMs. A sharp decrease in conductivity due to small particle sizes can be avoided.

3. Experimental study of boundary resistance for composite with nanoparticles

The thermal conductivity of particulate composites consisting of silica nanoparticles in epoxy resin was measured to characterize thermal boundary resistance. The resin, Epoxy SC-79, was obtained from Applied Poleramic, Inc. Silica nanoparticles were made using sol-gel process, which ensures uniformly distributed silica nanocomposite. The sol–gel process also functionalizes silica and nanoparticles are treated with coupling agent to ensure even distribution in the epoxy resin. The 5 and 10% weight fractions (3.1% and 5.2% volume fractions) of silica nanocomposites were manufactured with two particles sizes (20 nm and 45 nm) to study the size effect.

Thermal conductivity measurements were performed at Oak Ridge National Lab (ORNL) in the High Temperature Materials Lab (HTML). The thermal conductivity was measured using the transient plane source (TPS) technique, also called as hot disk technique, which was developed by Gustafsson [16]. It is a modified hot wire technique, which has been used for a long time for measuring

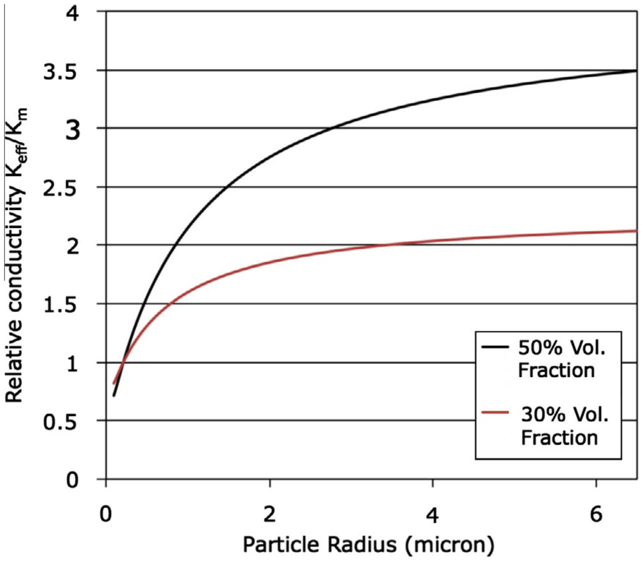

Fig. 2. Relationship of size of conductive particle (copper) as a function of relative conductivity of the TIM (matrix = silicone elastomer).