Development of MgO-Epoxy Composites with Enhanced Thermal Conductivity

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Abstract- *Heat dissipation is an important issue for electronic devices. In present work, MgO-epoxy composites with high thermal conductivity were successfully prepared by infiltrating magnesium oxide(MgO) powder into epoxy resin. The thermal conductivity of MgO-epoxy composites has been studied at variable percentage of filler. Test result indicated an increase of thermal conductivity of the composites at 20 wt% of MgO. The MgO-epoxy composites demonstrate much higher thermal conductivity, up to 0.207 W/(mK), compared with neat epoxy. This study demonstrates a potential route to manufacture epoxy-based composites with extremely high thermal conductivity.*

Key Words: Thermal conductivity (TC), Thermal interface material (TIM), Thermally conductive fillers, Epoxy resin.

1. INTRODUCTION

Polymeric materials when used as the base matrix perform as the platform, whereas the conductive fillers adhere to and further propagate the thermal conductance. Epoxy, a versatile thermoset resin has been widely utilized as a packaging and insulating material in electronic and electrical industries, owing to its high resistivity, low dielectric constant, and excellent processability. However, certain impediments such as low thermal conductivity and high coefficient of thermal expansion (CTE) of these polymers have resulted in thermal failure during its end use application. Moreover, in addition to thermal and electrical properties, mechanical properties of epoxy resin as a substrate and packaging material also play a vital role.

Ceramic filler-reinforced polymer composites have generated considerable interest in the recent years in the areas as of electronic packaging. Relative case of processability and excellent flexibility in these composites has driven them as potential candidates for the development of devices for electric stress control, high storage capability, and high permittivity material. Several investigation pertaining to improving the stiffness and strength in the electronic packaging materials with the use of inorganic fillers such as Al_2O_3, SiO_2, ZnO, and BeO have been suggested. Similar other fillers such as silicon carbide SiC, nitride (AlN and BN) and carbon-based materials are also known to effectively resolve the problems of thermal dissipation.

Packaging electronic devices is a growing challenge as device performance and power levels escalate. As device feature sizes decrease, ensuring reliable operation becomes a challenge. Ensuring effective heat transfer from an integrated circuit and its heat spreader to a heat sink is a vital step in meeting this challenge. For example, die size of the Pentium 2 was 25.4 mm square and the power dissipation was about 33 W. However, the Pentium 4 die is 12.5 mm square and will dissipate up to 80 W. It has been seen that the power dissipation from the CPU are in the range of 20 to 150W. [1]

It is well known that the reliability of an electronic device is exponentially dependent on the operating temperature of the junction, wherein a small increase of 10-15℃ can result in a two times decrease in the lifespan of the device [2,3]. And also the stability of an electronic device will be decreased 10% by per 2℃ rise in temperature when they are working for long time [4]. Thermal management in conventional electronics was difficult because the conducting path between heat sources and heat sinks were made of rigid materials that possess high thermal conductivity, such as metals. It is hard to make a perfect contact between heat sinks and electronics because of the differences in the flatness between the two surfaces because there are only three contact points between two rigid planes, which causes very poor thermal contact [5]. Therefore, there is need of a soft, deformable material capable of forming low-thermal-resistance contacts in the interface between the electronics and heat sinks. The general method to do this is to fill the air gap by using an elastomeric material with high thermal conductivity in the interface between the heat sink and the heated device. Such materials are called thermal interface materials (TIMs). TIMs conduct heat more effectively to the heat sink than air and thus they

reduce the resistance to heat transfer caused by air voids in the device.

1.1 Theory of thermal interface material

Fig (1) shows the location of TIM in typical electronic packaging. Thermal interface resistance is a measure of how well heat is transferred across the interface of two mating rigid surfaces, such as a CPU and a heat sink base plate; for lower value of resistance the heat transfer will be higher.

Gwinn et al. [1] has explained more clearly the concept of TIMs. In the case of the CPU–heat sink interface, the interface resistance (per unit area) R_{int} is defined as
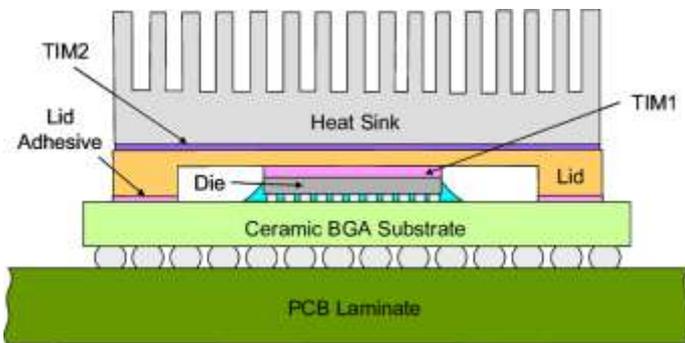
Fig-1: Typical Packaging Architecture for a High Performance Microprocessor, highlighting location of TIM1 and TIM2

$$R_{int} = \frac{T_{cpu} - T_{hs}}{Q} \qquad (1)$$

Where T_{cpu} is the CPU (or electronic packaging) surface temperature, T_{hs} is the heat sink base plate temperature and Q is the power per unit area of heat emitting component.

The interface resistance is generally expressed in K cm²/W (or in K in.² /W). Fig. 2(b) and (c) shows that the CPU and heat sink surfaces being joined contain a combination of surface roughness and surface non-flatness. The micro roughness on a macroscopic non-planar is in the form of a concave, convex or wavy surface, resulting in as much as 99% of the interface area between the heat sink and CPU being separated by air filled gaps unless a TIM is used. Theoretically, the mating surfaces will only contact each other at discrete points and remaining is filled with air. Because air is a very poor thermal conductor (k_{ai}=0.026 W/mK at room temperature), it acts as a thermal barrier preventing efficient heat transfer across the interface.

If no TIM is used to enhance heat transfer across the interface, the total interface resistance, R_{int} is due to contact resistance, $R_{contact}$ as shown in Fig. 2a. Contact resistance, $R_{contact}$, is composed of two parallel resistances: (1) conduction resistance at the points where the mating surfaces contact and (2) conduction and/or radiative resistance across the gaps of non-contacting area. The existence of TIM material adds a thermal conduction resistance across the material of thickness t.

$$R_{int} = R_{contact1} + R_{cond} + R_{contact2} \qquad (2)$$

And $$R_{cond} = \frac{t}{k_{TIM}A} \qquad (3)$$

Where $R_{contact1}$ the contact resistance between the TIM and the CPU surfaces is, $R_{contact2}$ is the contact resistance between the TIM and the heat sink base plate and R_{cond} is the conduction resistance across the thickness of the TIM. k_{TIM} is the TIM bulk thermal conductivity, t is the thickness of the TIM and A is the surface area of the electronic component across which heat transfer occurs.
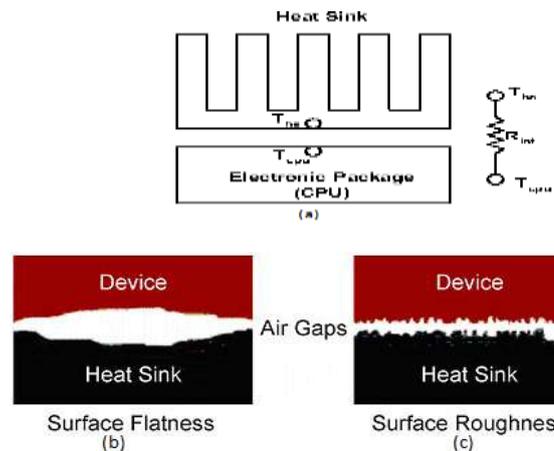
Fig -2: (a) definition of thermal interface resistance, R_{int} (b) heat sink and CPU surface roughness, and (c) poor heat sink and CPU surface flatness.

Fillers given that polymers have low TC (generally less than 1.0 W/(mK)), making high-TC polymer-based materials requires introducing high-TC filler particles into polymer matrix [6-8]. Especially in the topic of the Nano fluids [9] and polymers [10]. Also, metal meshes and foams have been used in the thermal storage systems to improve the thermal conductivity of the systems. A few theories have been suggested on the heat transfer enhancement in Nano fluids or composite materials.

Fillers may be in the form of fibers or in the form of particles uniformly dispersed in the polymer matrix. The thermo-physical properties of fiber-filled composites

are anisotropic, except for randomly dispersed fibers; while the thermo-physical properties of particle-filled polymers are isotropic [11]. The size of the fiber or particle may be of the order of Micro or Nano. Different types of fillers used in epoxy composite such as Metallic fillers, Carbon based materials, Ceramic fillers, and Hybrid fillers out of which ceramic fillers used extensively where high thermal conductivity and electrical insulation are required.

Ceramic fillers have been widely studied for thermally conductive and electrically insulating composites due to their inherent properties. They lack free electrons, and the heat transfer is predominantly through phonons. Generally, except for BeO, most metal oxide fillers such as alumina (Al_2O_3) [12, 13] and silica (SiO_2) have a lower TC. Non-oxide fillers, such as aluminum nitride (AlN)[14, 15], boron nitride (BN), silicon nitride (Si_3N_4) [16] or silicon carbide (SiC) [17] have high TC as their strong interatomic bonds and crystal structure can significantly reduce phonon scattering. In particular, BN and AlN fillers are widely used for thermally conductive and electrically insulating composites [18]. However, these fillers also have some limitations, e.g., BN has an extremely low TC in the through-plane direction and AlN particles are easy to hydrolyze [19, 20] (although the moisture reactivity of AlN could potentially be mitigated by surface modification such as silica coating [21]).

2. EXPERIMENTAL DETAILS
2.1 Raw materials

The raw materials and the corresponding specifications for the fillers, epoxy resin and curing agent are given in table 1.

2.2 composite preparation

The composite samples of various compositions are prepared by using hand lay-up technique. Hand lay-up technique is the oldest and simplest technique for composite preparation. The MgO-Epoxy composites are prepared on following steps:

(i) The moulds are prepared with PVC material in order to get disc type specimens (diameter 50mm and thickness 6mm) and coated with releasing agent (silicon high vacuum grease)

(ii) Uncured epoxy resin (Aretite crystal clear) and corresponding hardener (Aretite epoxy hardner) are mixed in a ratio of 10:3 by weight as per recommendation.

(iii) Micro-sized MgO particles are mixed with different proportions i.e. 0%, 10%, 15% and 20% by weight of epoxy and stirred for 30 minutes.

(iv) The uniformly mixed mixtures (epoxy filled with MgO particles) are poured into the PVC moulds.

(v) The castings are then left in the room temperature for about 30 hours and then the PVC moulds are broken and the samples are released.

3. RESULTS AND DISCUSSION

The thermal conductivity of the composites have been tested in the laboratory by using UNITHERM 2022 instrument according to standard ASTM E1530: 2015 test method. The recorded values of thermal conductivity of different composite samples with their filler loadings are shown in the table 2.

Table 1 The raw materials and corresponding specifications of filler, epoxy resin, and curing agent.

Materials	Abbreviation	Purity (%)	Supplier
Magnesium Oxide	Micro-MgO	99%	Merck life science private limited, Mumbai
Epoxy Resin	-	99%	Aretite Godadiya chamicals, Surat, Gujrat
Epoxy Hardner (curing agent)	-	99%	Aretite Godadiya chamicals, Surat, Gujrat
Silicon high vacuum grease	-	98%	Molychem company limited

Table 2 Experimental values of thermal conductivity of MgO-Epoxy composites

Sample no.	Filler loading (wt %)	Thermal conductivity (W/m-K)	K_c/K_m
1	0	0.140	1
2	10	0.171	1.221
3	15	0.180	1.285
4	20	0.207	1.478

The experimental values are also represented in a form of graph(Fig. 3)

Fig. 3 Effect of filler loading on thermal conductivity
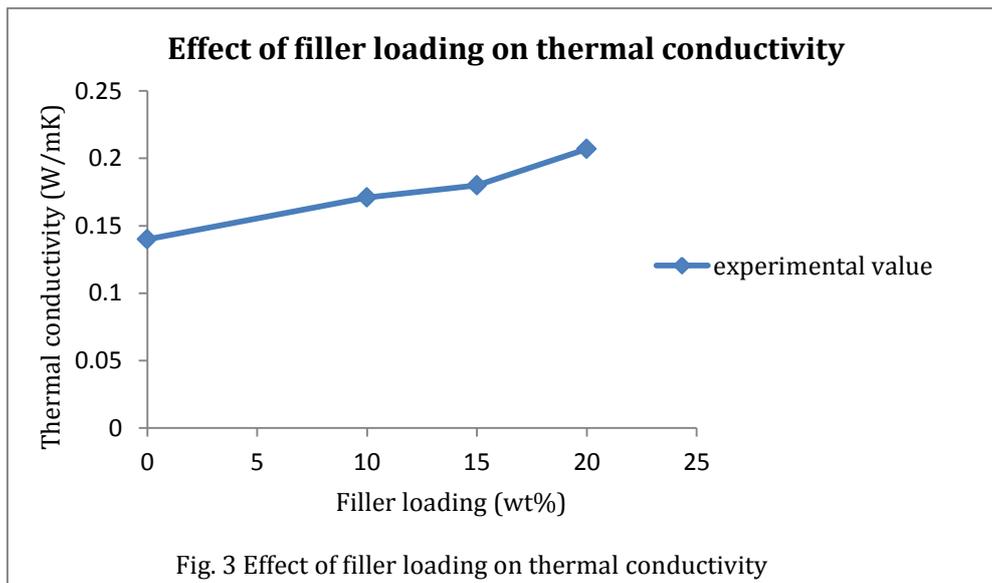
There are various types of theoretical thermal conductivity models like Rule of mixture model, Geometric mean model, Lewis and Nielsen model, Maxwell-Garnett model, Bruggeman model etc. but only one model is considered for validation because of the similarity in results with experimental values. The Maxwell–Garnett (MG) model is considered into account for comparison of the experimental results with the results calculated by using Maxwell–Garnett (MG) model. Maxwell–Garnett [22] derived the expression for the conductivity k for suspension of spherical randomly distributed and non-interacting particles, as defined in equation (4)

$$K_c = K_m \left\{ 1 + \frac{3\varphi(\delta-1)}{2+\delta-\varphi(\delta-1)} \right\} \qquad (4)$$

Where, K_c, K_f and K_m are the thermal conductivities of the composites, filler and polymer matrix respectively, φ be the volume fraction of filler into epoxy composite and $\delta = K_f / K_m$.

Comparison of values of effective thermal conductivity obtained from Maxwell-Garnett model with experimentally measured values is shown in Table 3. For more depth of interpretation the tabulated data is also plotted below in the form of graph in fig.4

Table 3 Comparison of values of effective thermal conductivity obtained from Maxwell-Garnett model with experimentally measured values

Sample no.	Filler loading (wt %)	Effective Thermal conductivity (W/m-K)	
		Experimental values	Maxwell-Garnett model
1	0	0.140	0.140
2	10	0.171	0.185
3	15	0.180	0.212
4	20	0.207	0.242
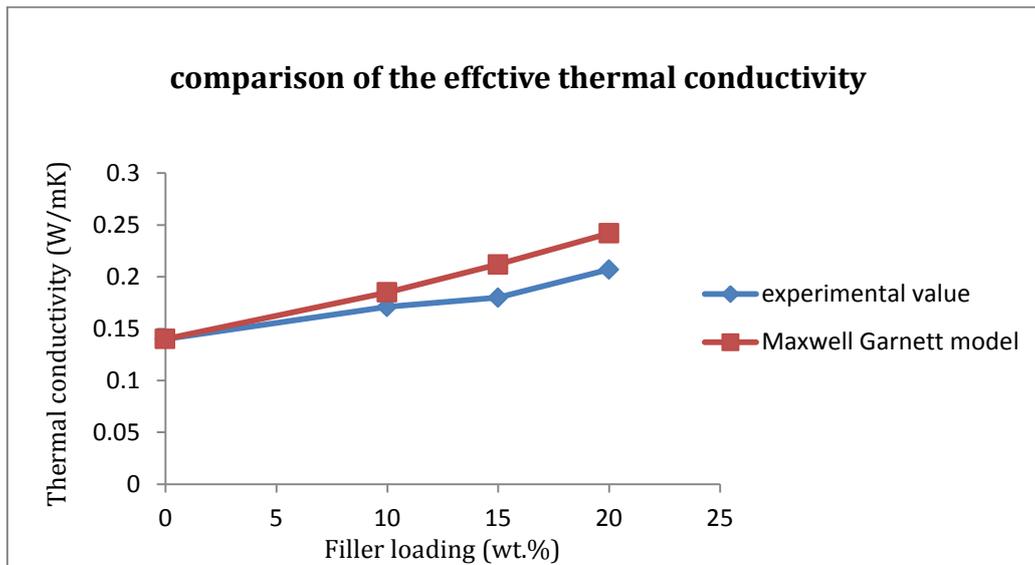
Figure 4 Comparisons of experimental result with the results obtained by theoretical models

Here we see that the experimental result follows the same pattern with the results obtained by TC model. The thermal conductivity of MgO-Epoxy composites increases with increase in filler loading from 0 to 20 % in both methods, but there is decrease in thermal conductivity of composite after a threshold limit. The possible reason behind this happening may be percolation threshold. For every experimental method, there is a limit beyond which the addition of filler material i.e. increase in filler loading into composite is not desirable. The addition of filler materials beyond the percolation threshold into composites may cause uneven changes in the property of composites like difficulty in processing the composite, loss of mechanical property, abrupt changes or not improvement or even sometimes decrease in thermal conductivity. Chen et al. [22] has described in their review that high filler loading above the percolation threshold is usually needed to achieve high TC. High filler loading will normally cause a significant increase on the viscosity of composites and make it more difficult for composites to be processed by conventional polymer processing techniques like hand lay-up. The rise of viscosity arises from the interfacial friction between fillers, and between fillers and polymers. This rise in viscosity may damage TC property of material. McNamara et al. [23] has also described in his review that the impact of increasing the percolation threshold causes decrease in the expected thermal conductivity of the composites. Some discrepancies with the experimental and theoretical results are mainly due to the differences in theoretical assumptions with the actual conditions. The theoretical models is designed for spherical filler with filler loading in vol.%, but the experimental result is on the basis of filler loading in wt.%

and also the non-intersecting condition is difficult to achieve through experimental technique in case of particle filled composites. At last, experimental results closely follow the MG model and the highest value of thermal conductivity in experimental method is found to be 0.207 W/m-K at 20% filler loading, which is 1.478 times higher than that of neat epoxy.

CONCLUSIONS

Following conclusions can be drawn from present investigation on MgO- Epoxy composites:

1) Magnesium oxide powder possesses reinforcing potential to be used as a filler material epoxy matrix composite.

2) Incorporation and fabrication of MgO particles into epoxy matrix is successfully done by using simple hand lay-up technique.

3) The thermal conductivity of MgO- Epoxy composite increases with increase in filler loading up to percolation threshold. The highest value of thermal conductivity of epoxy-Mg composite is found 0.207 W/m-K at 20wt%, which is 1.478 higher than the neat epoxy matrix.

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