Thermo-Gravimetric Analysis of MgO-Epoxy Composite

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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 stability of MgO-epoxy composites has been studied at variable percentage of filler. Test result indicated an increase of filler content, there is decrease in thermal stability of the composites. The MgO-epoxy composites demonstrate much higher thermal stability at lower filler content. This study demonstrates a potential route to manufacture epoxy-based composites with extremely high thermal conductivity.

Key words: Thermally Conductive Fillers, Epoxy Resin, Thermo-Gravimetric Analysis (TGA)

I. INTRODUCTION

Thermal management is critical to the performance, lifetime, and reliability of electronic devices. With the miniaturization, integration of electronics and the development of new filler materials lead to the emergence of new applications such as light emitting diodes, solar PV cells, electronic packaging and in various electronic devices thermal dissipation becomes a challenging problem. Addressing this challenge requires the development of novel polymer-based composite materials with enhanced thermal conductivity [1, 2].

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. [3]

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 [4,5]. 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 [6]. 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 [7]. 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.

II. THERMO-GRAVIMETRIC ANALYSIS

Thermo-gravimetric analysis or thermal gravimetric analysis (TGA) is a method of thermal analysis in which changes in chemical and physical properties of materials are measured as a function of increasing temperature (with constant heating rate), or as a function of time (with constant temperature and/or constant mass loss). TGA can give information about chemical phenomena, such as desolvation (especially dehydration), decomposition and solid-gas reactions (e.g. oxidation or reduction). Similarly TGA can give information's about physical phenomena such as second order phase transition including vaporization, sublimation, absorption and desorption.

TGA can be used to evaluate the thermal stability of a material. It is generally used to investigate the thermal stability of polymers. In a desired temperature range, if material is thermally stable, there will be no observed mass change. Negligible mass loss corresponds to little or no slope in the TGA trace. TGA also gives the upper application temperature of a material. Beyond this temperature the material will begin to degrade.

Many researchers have done TGA analysis in epoxy composites. Some of them have discussed here:

Wang et al. [8] has investigated the thermal stability of pure epoxy and epoxy composites filled with 4.5 wt.% expanded graphite and with 4.5 wt.% (KH550@EG) using TA Q50 TGA system. The sample was heated to 600℃ at the rate of 10°C/min under nitrogen atmosphere. The initial decomposition temperatures were found to be 318℃, 329℃ and 348℃ respectively. They concluded that surface treated samples have better compatibility than untreated samples, which consequently improves the thermal stability.

Gu et al. [9] has investigated the thermal gravimetric (TG) analyses of the samples of Graphene Nano Plates (GNP) and functionalized Graphene Nano Plates (f-GNP) were

carried out at 10 C/min (argon atmosphere), over the whole range of temperature (40–800℃) by STA 449F3. They found that the heat-resisting index is 192℃ (pristine E-51), 195℃ (0.5 wt.% f-GNPs), 199℃ (10 wt.% f-GNPs), 200℃ (20 wt.% f-GNPs) and 203℃ (30 wt.% f-GNPs), respectively and concluded that the thermal stabilities of the f-GNPs/ E-51 Nano composites are gradually improved with the increasing addition of f-GNPs.

Xu et al. [10] has done investigation on TGA analysis of the composites filled with boron nitride and aluminium nitride particles with or without of surface treatment using a PERKIN ELMER TGA7 and measured the weight loss heating up to 600℃ at 20℃/min under nitrogen atmosphere. They found that weight loss of the samples treated with silane agents are higher than the samples which were treated with acetone and acids (nitric and sulphuric acid).

Yu et al. [11] has tested the amount of h-BN in the composites using thermo-gravimetric analyser TG 209 F1 (NETZSCH, Germany) under air atmosphere with a heating rate of 10 °C/ min. The samples were heated up to 700℃ and three significant drops were found from the weight loss curve. The first one, which appeared at around 145 °C-270 °C, attributed to the pyrolysis of the low molecular weight components. The second peak at 270 °C-420 °C should be ascribed to the decomposition of epoxy resin. The last obvious weight loss happened at 420 °C-620 °C due to the combustion of the residual carbon of epoxy resin in air atmosphere.

Liang et al. [12] have prepared Nano composite samples with 1 wt% loading of either SiO_2 fillers or Al_2O_3 fillers and micro-composite samples with 20 wt% loading of either SiO_2 fillers or Al_2O_3 fillers. Nano-micro (hybrid) composite samples (either use SiO_2 or Al_2O_3 fillers) were prepared by dispersing 1 wt% nano fillers and 20 wt% micro fillers in the epoxy resin. The TGA analysis was performed on Perkin-Elmer TGA7 to obtain information on thermal stability. The TGA analysis was done under the environment of Nitrogen gas. During the test, the temperature in test chamber increased from 30°C to 800°C at a heating rate of 10°C/min. From 800°C to 850°C, atmosphere change from nitrogen to air and the heating rate was 10°C/min. They found that the weight loss rate of micro-composite and nano-micro composite is much lower compare to neat epoxy and the SiO_2 filled specimens displayed a very similar pattern to Al_2O_3 filled specimens.

III. EXPERIMENTAL DETAILS

A. Raw materials

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

B. 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:
1) 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)
2) 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.
3) 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.
4) The uniformly mixed mixtures (epoxy filled with MgO particles) are poured into the PVC moulds.
5) 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.

IV. RESULTS AND DISCUSSION

In the present research work the thermo-gravimetric analysis of MgO-Epoxy composite samples are measured using TGA Q50 instrument ASTM-D 1868:2016 standard test method. In this analysis the test samples are heated 800 ℃/min at constant heating rate 10℃/min under the environment of Nitrogen gas. In the thermo-gravimetric analysis (TGA), all the samples of MgO-Epoxy composite are heated up to 800℃ at constant heating rate 10℃/min of Nitrogen gas. Nitrogen gas is used here to create inert environment which prevents the undesirable reactions. The TGA curve is commonly referred as thermogram, which shows the change in mass or weight as well as in the rate of weight loss with increase in temperature. The thermograms of all MgO-Epoxy composite samples of different filler loading are attached here (fig. 1-4).

In every thermogram certain amount of material (which is shown in the upper left corner of the Fig.) is taken for analysis and heated upto certain temperature 600-800 ℃ with constant heating rate of 10℃/min. The weight of composite decreases with increase in temperature. A derivative weight loss curves are also being used to tell the point at which weight loss is most apparent.

In a desired temperature range, if material is thermally stable, there will be no observed mass change or Negligible mass loss corresponds to little or no slope in the TGA trace. TGA also gives the upper application temperature of a material. Beyond this temperature the material will begin to degrade. Initial decomposition temperature is the point where material starts disintegrating and is the measure of thermal stability of that material. The thermal stability of all MgO-Epoxy composites are evaluated from their respective thermograms and are listed below in the table 2.

Here we see that the thermal stability of MgO-Epoxy composites decreases with increase in filler loading (table 2). Every thermogram of epoxy composites contains 3 or 4 peaks in derivative curves. The first peak on derivative curves may be due to the loss of moisture content of composites. The second and third peaks stand for decomposition of composite material and at the last peak the entire composite is converted into residue i.e. ash. The area under the derivative curve qualitatively determines the amount of that component decomposed.

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

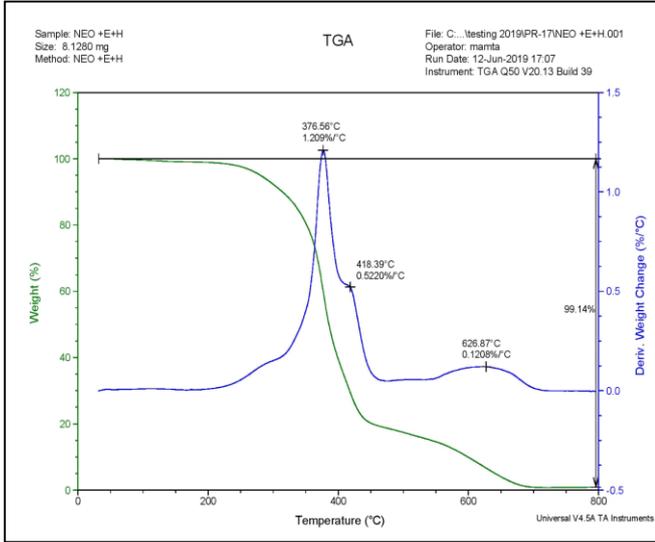Table 1: The raw materials and corresponding specifications of filler, epoxy resin, and curing agent.

Figure 1: Thermogram of MgO-Epoxy composite with filler loading 0%.
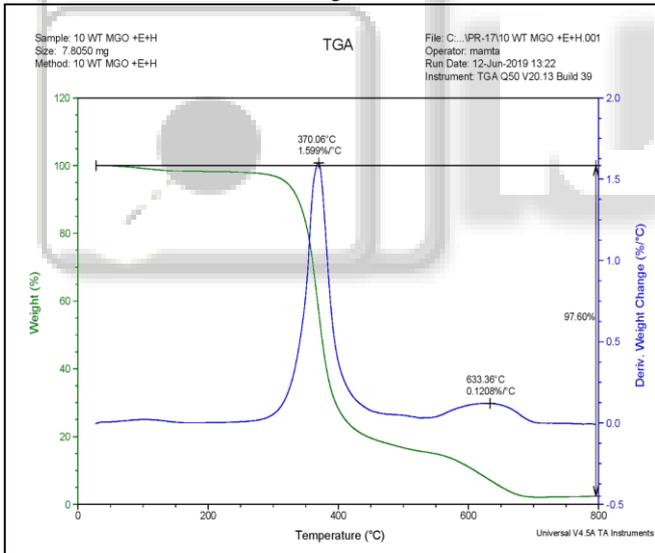

Figure 2: Thermogram of MgO-Epoxy composite with filler loading 10%
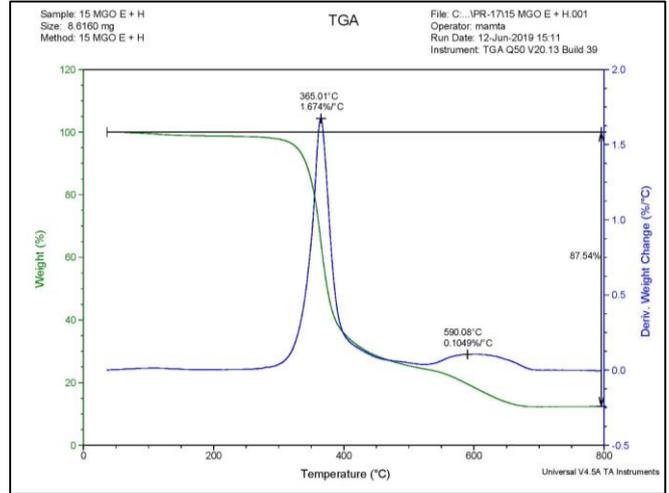

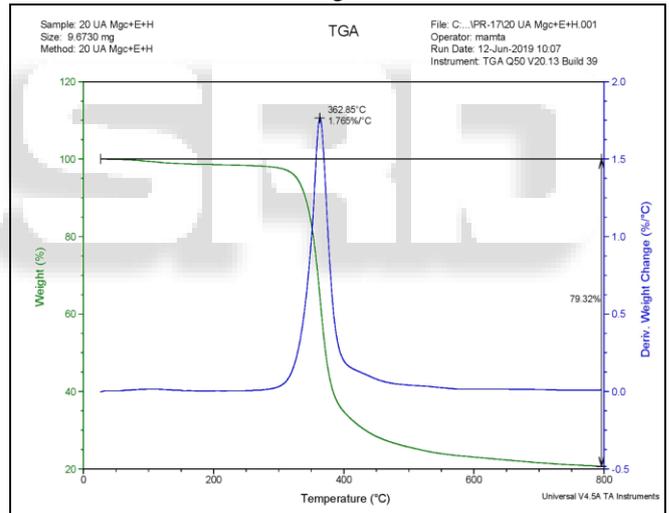Figure 3: Thermogram of MgO-Epoxy composite with filler loading 15%

Figure 4: Thermogram of MgO-Epoxy composite with filler loading 20%

S.No.	Filler loading (wt%)	Thermal stability temperature(in ℃)	Derivative weight change (in %/℃)
1	0	376.56	1.209
2	10	370.06	1.599
3	15	365.01	1.674
4	20	362.85	1.765

Table 2: Thermal stability temperatures for MgO-Epoxy composite

On comparing all the thermograms we found that the maximum derivative weight change per ℃ rise in temperature or rate of decomposition increases with increase in filler loading which shows that the addition of MgO fillers into epoxy matrix make the epoxy matrix to be unstable at higher temperature. The maximum rate of decomposition temperature are evaluated and listed below in table 3

S.No.	Filler loading (wt%)	Maximum decomposition rate (in %/°C)
1	0	1.209
2	10	1.599
3	15	1.674
4	20	1.765

Table 3: Thermal stability temperatures for MgO-Epoxy composite

V. CONCLUSION

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

- The thermal stability temperatures are recorded as 376.56℃, 370.06℃, 365.01℃ and 362.85℃ for MgO-Epoxy composite having filler loading 0%, 10%, 15% and 20% by weight respectively.
- The thermal stability of for MgO-Epoxy composite decreases with increase in filler loading.
- The maximum derivative weight change per℃ rise in temperature increases with increase in filler loading.

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REFERENCES

[1] Moore AL, Shi L. Emerging challenges and materials for thermalmanagement of electronics. Mater Today 2014;17:163–74.

[2] Tong XC. Advanced materials for thermal management of electronicpackaging. New York: Springer; 2011, 616 pp.

[3] J.P. Gwinn, R.L.Webb, Performance and testing of thermal interface materials. Microelectronics Journal 34 (2003) 215–222.

[4] Kisang Ahn, Kiho Kim, Joohen Kim, Thermal conductivity and electric properties of epoxy composites filled with TiO2-coated copper nanowire. Polymer 76 (2015) 313-320.

[5] L.C. Sim, S.R. Ramanan, H. Ismail, K.N. Seetharamu, T.J. Goh, Thermal characterization of Al_2O_3 and ZnO reinforced silicone rubber as thermal pads for heat dissipation purposes, Thermochim. Acta 430 (2005) 155-165.

[6] Wenhui Yuan, Qiangqiang Xiao, Li Li, Tao Xu, Thermal conductivity of epoxy adhesive enhanced by hybrid graphene oxide/AlN particles. Applied Thermal Engineering 106 (2016) 1067–1074.

[7] Farhad Sarvar, David C. Whalley, Paul P. Conway, Thermal Interface Materials - A Review of the State of the Art, Electronics System integration Technology, IEEE (2006) 1292-1302.

[8] Zhaofu Wang, Rong Qi, Jin Wang, Shuhua Qi, Thermal conductivity improvement of epoxy composite filled with expanded graphite, Ceramics International (2015).

[9] Junwei Gu, Xutong Yang, Zhaoyuan Lv, Nan Li, Chaobo Liang, Qiuyu Zhang, Functionalized graphite nanoplatelets/epoxy resin nanocomposites with high thermal conductivity, International Journal of Heat and Mass Transfer 92 (2016) 15–22.

[10] Yunshang Xu, D.D.L. Chung, increasing the thermal conductivity of boron nitride and aluminium nitride particle epoxy matrix composite by particle surface treatments, Composite interfaces, vol 7, no. 4 (2006) 243-256.

[11] Cuiping Yu, Jun Zhang, Zhuo Li, Wei Tian, Liangjie Wang, Jie Luo, Qiulong Li, Xiaodong Fan, Yagang Yao, Enhanced through-plane thermal conductivity of boron nitride/epoxy composite, journals of composites (2017).

[12] Mu Liang, K L Wong, Study of mechanical and thermal performances of epoxy resin filled with micro particles and nanoparticles, Energy Procedia 110 (2017) 156–161