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Research paper

Thermal Properties of the Graphene Composites: Application of Thermal Interface Materials

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Abstract

Epoxy mixed with others filler for thermal interface material (TIM) had been well conducted and developed. There are problem occurs when previous material were used as matrix material likes epoxy that has non-uniform thickness of thermal interface material produce, time taken for solidification and others. Thermal pad or thermal interface material using graphene as main material to overcome the existing problem and at the same time to increase thermal conductivity and thermal contact resistance. Three types of composite graphene were used for thermal interface material in this research. The sample that contain 10 wt. %, 20 wt. % and 30 wt. % of graphene was used with different contain of graphene oxide (GO). The thermal conductivity of thermal interface material is both measured and it was found that the increase of amount of graphene used will increase the thermal conductivity of thermal interface material. The highest thermal conductivity is 12.8 W/ (mK) with 30 w. % graphene. The comparison between the present thermal interface material and other thermal interface material show that this present graphene-epoxy is an excellent thermal interface material in increasing thermal conductivity.

Keywords: *Graphene; Thermal Interface Material (TIM); Thermal Conductivity; Graphene Oxide (GO).*

1. Introduction

Thermal management in electronic industry has become crucial especially in removing the heat flux generated from the chip to the heat sink [1]. One of the biggest problem is high thermal contact happen when two surfaces collide under the high thermal input which can affect the reliability of heat sink from overheat and malfunction [2]. There are a lot of countermeasure and research has been done to solve the existing problem. High thermal conductive material is one of the most possible solution to solve the thermal management issue and to depress the reliability of electronic component [3]. Thus, thermal pad was studied to improve the reliability and thermal conductivity of electronic components especially in heat sink and chips application [4]. Investigation of thermal interface material also focus on reducing thermal contact resistance between heat sink and chip in order to reduce the heat flux generated from chip to heat sink [5]. The traditional method to produce thermal interface material usually using polymer (silicon elastomer) or grease cause low thermal conductivity of TIMs [6].

Thermal grease and thermal paste are the types of thermal interface material commonly used in the current market [7]. The base material in thermal grease and thermal paste is generally categorized as a silicone or non-silicone type. This types of material used silicone-based pastes because they are well-established, low-cost, high performing, and very reliable [8]. The filler material in most pastes is metal oxide (ZnO, BN, Al_2O_3), silver, or graphite. In order to increase the thermal conductivity, several high thermal conductive material has been use as a filler such as Cu, Al, AIN etc. [9]. However, by adding these fillers as large as 30-50% didn't give much improvement to thermal conductivity of thermal interface material, it only increases up to 1-5W/ (mK) at the room temperature [10]. Beside that there a lot of problem by using thermal grease and thermal conductive adhesive such as overflow, inconsistent thickness and curing process [11].

However, after the discovery of the graphene which has the highest thermal conductivity, many researchers start to study the effect of filling graphene to the thermal interface material start with study of composite between thermal grease with small portion of graphene. It was proven thermal grease fill with 4.25 vol. % graphene was able to increase 668% thermal conductivity up to 1.047W/mK from 0.156 W/mK [12]. Graphene sheets and alumina particles filler also was used to improve thermal conductivity. Graphene (1 wt. %) was add in thermal grease to increase the thermal conductivity up to 3.45 W/mK [13]. Epoxy has been incorporated with graphene to increase the thermal conductivity because epoxy has poor thermal conductivity (0.15–0.25 W/mK) [14]. Thermal conductivity of thermal interface material increases up to 6.45 W/mK when incorporated epoxy resin with 25 vol. % of graphene [15]. In this research, the effect of graphene with different weight percentage on thermal conductivity are all measured and studied to discover the most suitable graphene filling fraction for graphene-epoxy composite thermal interface material, to improve thermal conductivity in electronic application especially in heat sink application.

2. Methodology

2.1. Preparation of the graphene epoxy composites

The graphene used in this paper is prepared and process after go through liquid-phase exfoliation [16]. In order to obtain superior thermal properties in graphene composite material some modification has been made by using acidification to make sure the epoxy and graphene mixed well [17]. Preparation of acidified graphene need to go through several procedures. First, graphene must be added to a sulfuric acid with 3-4-hour under magnetic stirring process and then undergoes ultrasonic oscillation process. The ratio between these two types of material which are graphene with acid sulfuric was used 1:50 g/ml. After the process of mixing these material finish, nitric acid is added, with 150 °C constant temperature, then acid sulfuric was added with the ratio 3:1 volume ratio of sulfuric acid to nitric acid. Then magnetic stirring was used for 1-2 hours to ensure the mixture is condensed and it using fiber micro porous membrane with the high grade and micro scale pore (pore size of 0.6 μm) until the pH value is 7. After the filtration process finish, the mixer is dried in a vacuum drying oven under temperature 50 °C for a certain period of times (usually for 7-8 hours). After drying process was completed, the graphene and epoxy are stirred under vacuum at temperature at 150 °C for 2-3 hours) in order to remove the air bubbles that still existing in the composites. Lastly, to make these composite into the proper thermal interface material shape, it was pressed into thin films at 150 °C. After compressed process, the graphene-epoxy composite that been converted into thin film shape, it need to be are cooled to room temperature, as shown in Figure 1.

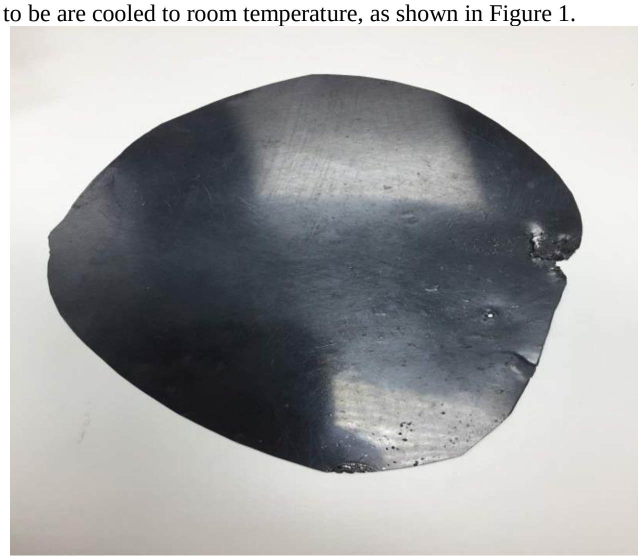


Fig. 1: Samples of graphene-epoxy TIM is formed

2.2. Characterization and measurement

The scanning electron microscope (SEM) was used to show the morphologies of the graphene-epoxy as shown in Figure 2 and 3. The thermal conductivity of composite were calculated by using formula which is $k(T) = \alpha(T) * Cp(T) * \rho$, where, the thermal diffusivity of graphene composites was measured by using the laser flash method, and the specific heat capacity of composites is measured by differential scanning calorimetry (DSC). Minimum of 5 samples was used with the same filler weight percentage are tested in each process, to ensure the result are accurate and precise. The measurement is conducted in a vacuum chamber to reduce the heat loss.

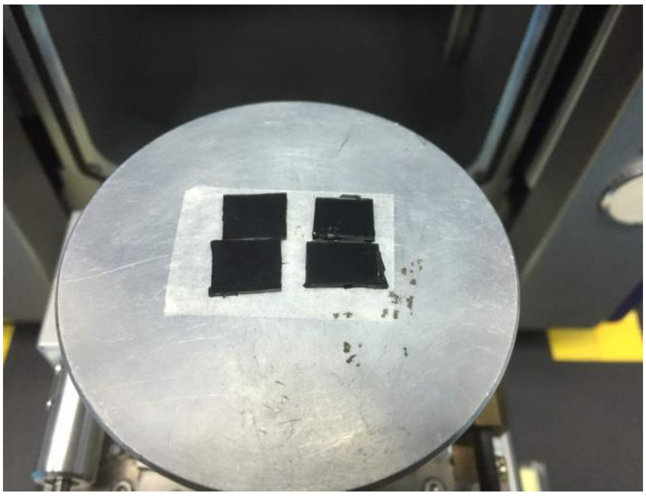
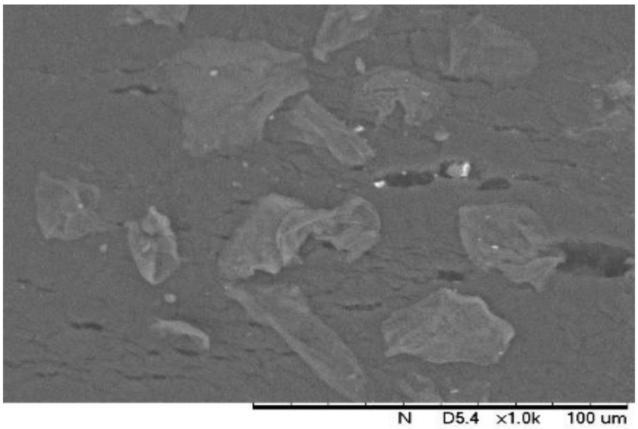
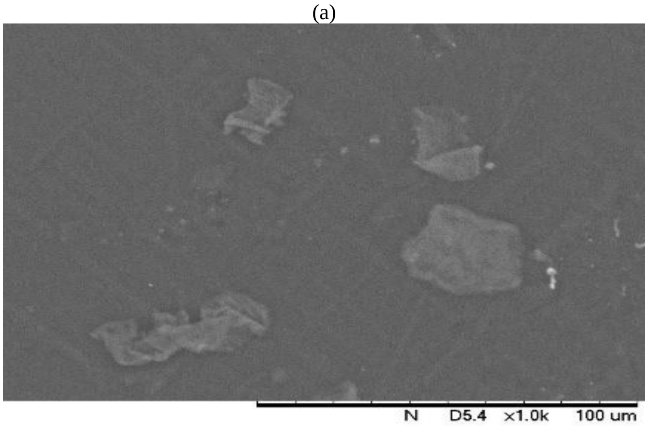


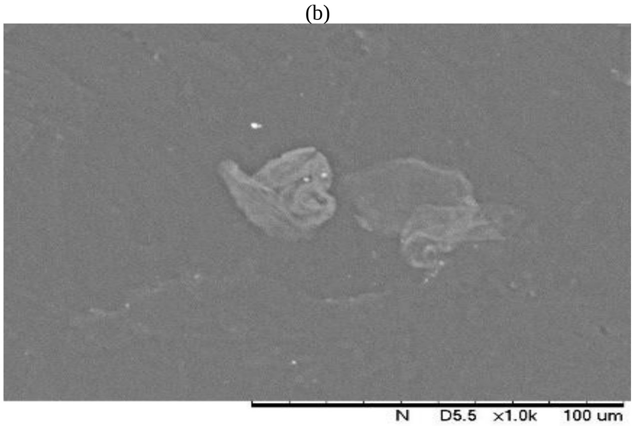
Fig. 2: Samples of graphene composite during SEM process



(a)



(b)



(c)

Fig. 3: Samples of graphene composite during SEM process; a) 30 wt. % of grapheme; b) 20 wt. % of graphene; c) 10 wt. % of graphene

3. Results and discussion

The thermal conductivity of graphene-epoxy composite samples as function of weight percentage of graphene are measured and shown in Figure 3. The calculation of thermal conductivity of graphene composite based on formula as shown below. In order to verify the accuracy of measured thermal conductivity, the modified Maxwell–Garnett effective medium approximation (EMA) [18] and Xie's model [19] are applied to compare with the experimental The experimental results are obtained at room temperature 20 °C and epoxy was used as based material and has thermal conductivity of 0.3 W/(mK).

A few procedure has been implement in order to enhance the thermal conductivity of graphene-epoxy composite. Initially, the experimental process starts with 10 wt. % graphene, in order to increase the thermal conductivity of thermal interface material to 5.2 W/(mK). In addition of 10 wt. % of graphene to the based material it was proven successfully increase the thermal conductivity of TIM more than ten times. Then, the weight percentage of graphene increase to 20 wt. %, the thermal conductivity of TIM composites is 8.8 W/(mK), increase 69.2% compare with 10 wt. %. The highest thermal conductivity is up to 12.8 W/ (mK) when the weight percentage of graphene increase to 30 wt. %. This happen because high filling volume of graphene increase the internal thermal conductivity due to gap between particles is decrease. The present of graphene make the bonding between particles is stronger and highly increase the properties of thermal conductivity. The graphene is diffused in matrix materials isotropously and uniformly, the equations of the two models are as follows:

$$k = k_p \left[\frac{3k_m + 2f(k_p - k_m)}{(3-f)k_p + k_m f + \frac{R_B k_m k_p f}{H}} \right]. \quad (1)$$

Xie's model:

$$k = \frac{1}{6}\left\{ (3-5f)(k_m - k_p) + \sqrt{(3-5f)^2 (k_m - k_p)^2 + 12k_m \left[3k_m + f(k_p - k_m)\right]} \right\} \quad (2)$$

where f is the volume fraction of graphene, k, kp, km are the thermal conductivity of the composites, filled graphene and matrix material respectively. Besides that, the thermal boundary resistance (RB) at the graphene/matrix interface is taken into account in the modified

Maxwell–Garnett EMA, H is the thickness of the multilayer graphene. Table 1 shows the comparison between present studies with others in order to show the improvement has been made.

Table 1: Comparison of Thermal conductivity between different Thermal Interface Material

Filler	Thermal Conductivity W/(mK)	Fraction	References
GNP	6.44	25 vol.%	[15]
Graphite	5.1	10 vol.%	[18]
Graphene- MLG	3.6	20 vol.%	[19]
Graphene	5.6	10 vol.%	[16]
Graphene*	12.8	30 vol.%	Present Study

From Table 1 show that between all types of filler used, graphene is the most suitable and enable to increase the thermal conductivity of TIM. Graphene has been known worldwide which has the highest thermal conductivity and very compatible in TIM application [12]. Besides that, graphite also proven to enhance the thermal conductivity and properties of TIM but it is not as high as graphene. The different between graphite and graphene are 9.6% although they are using the same number of fraction. It is because the structural of graphite are slightly different and the fabrication process of graphite in producing TIM are not very compatible compare with graphene. The graphene also been used as composite component in producing TIM, example multilayer graphene (MLG) and mix with other material such as ZnO, BN, Al_2O_3. Although some adjustment and modification has been made but the thermal conductivity of the TIM are not improving [9, 19]. From Table 1, it shows that thermal conductivity of multilayer graphene (MLG) are lower than single layer by 144.4% with same amount of filler used. Thermal conductivity of TIM more suitable and effective by using single layer of graphene due to the less interference and thermal resistance during heat transfer and dissipation process [14-15]. This present study is using 30 vol. % of fraction as improvement of [16] and was able to increase 128.6% when filler increase 20%. The increase volume filler of graphene was proven to increase thermal conductivity because the increase amount of graphene particle enables to upgrade the structural bonding between composite particles. The stronger and more compatible particle was able to boost the internal thermal properties and enhanced thermal properties of TIM.

4. Conclusion

Graphene–epoxy composite material are proven to be good combination to improve thermal conductivity of TIM. The most important thing to the increase the thermal conductivity in TIM is by using suitable material that match the based material and they need to be very compatible to each other. Further investigation need to be made by using different weight percentages of graphene, the following conclusions can be obtained.

1. The thermal conductivity increased with the increase in the portion of graphene. The filled graphene gives great enhancement in the thermal conductivity of TIM composites. In this study the highest filled graphene only up to 30 wt. %, so it is more appropriate to further increase the graphene filling ratio to see whether it can further increase thermal conductivity or not.
2. The present TIM can be compare with the available TIM in the current market to ensure the marketability of our TIM is relevant. Further study need to be done and new parameter can be made like using high temperature and higher pressure to increase its performance.

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