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EXPERIMENTAL AND CORRELATED STUDIES ON THERMAL CONDUCTIVITY OF BERYLLIUM OXIDE PARTICLES FILLED POLYMER AS THERMAL INTERFACE MATERIAL FOR DYNAMIC HEAT FLOW IN LEDs PACKAGE

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UNIVERSITI SAINS MALAYSIA

2019

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by

NABIHAH BINTI MOHD FAUZI

Thesis submitted in fulfillment of the requirements for the degree of Master of Science

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LIST OF ABBREVIATIONS

Al	Aluminium
AM	Acrylamide
BeO	Beryllium Oxide
CCT	Color correlated temperature
CRI	Color rendering index
CTE	Coefficient of thermal expansion
DMA	Dynamic mechanical analysis
DUT	Device under test
FWHM	Full width half maximum
ICDD	International Centre for Diffraction Data
IR	Infrared
LED	Light Emitting Diode
LUX	Illuminance
MBAM	N, N'- Methylene-bis-acrylamide
PAN	Polyacrylonitrile
RPM	Revolutions per minute
SEM	Scanning electron microscope
TIMs	Thermal interface materials
TMA	Thermo-mechanical analysis
XRD	X-ray diffraction

LIST OF SYMBOLS

ΔR_{th}	Difference in thermal resistance
ΔT_J	Rise in junction temperature
°C	Degree Celsius
a, c	Lattice constant
C_p	Specific heat capacity
D	Crystallite size
E'	Storage modulus
E''	Loss modulus
k	Thermal conductivity
K	Sensitivity value (K-factor)
R_{th}	Thermal resistance
s	Seconds
Tan δ	Damping parameter
T_g	Glass temperature
T_J	Temperature junction
T_s	Surface temperature
v	Average velocity
V	Cell volume
wt %	Weight percentage
β	Full width half maximum
ε	Lattice strain
θ	Bragg diffraction angle
λ	Wavelength

KAJIAN EKSPERIMENTASI DAN BERKOLERASI KE ATAS KEKONDUKSIAN HABA POLIMER MENGANDUNGI ZARAH BERYLLIUM OKSIDA SEBAGAI BAHAN ANTARA MUKA HABA BAGI PENGALIRAN HABA DINAMIK DI DALAM PAKEJ LED

ABSTRAK

Zarah nano BeO disintesis menggunakan kaedah gel polyacrylamide dengan pelbagai suhu kalsinasi; 800, 900 dan 1000 ^{0}C. Pencirian struktur dan permukaan morfologi zarah nano BeO yang disintesis dan zarah mikro BeO yang komersil telah dianalisa menggunakan Pembelauan Sinar-X (XRD) dan Imbasan Mikroskop Elektron (SEM) untuk mengkaji struktur kristal dan permukaan morfologi zarah BeO yang akan memberi manfaat untuk dijadikan pengisi. Pengukuran kekonduksian haba telah dijalankan pada kedua-dua zarah (sintesis (800 ^{0}C) dan komersil) BeO dalam matrik epoksi untuk mengkaji kesan saiz zarah pada kekonduksian haba komposit epoksi. Berdasarkan pemerhatian, struktur dan permukaan morfologi kristal yang menghasilkan kualiti kristal terbaik untuk zarah BeO serta saiz optimum zarah yang meningkatkan lagi kekonduksian terma komposit epoksi telah diperhatikan pada zarah mikro BeO yang komersil. Campuran zarah BeO yang diisi komposit epoksi telah disediakan dengan pelbagai peratus pengisi (10, 30 dan 50 wt%) menggunakan pengaduk mekanikal. Kemudian, analisis Pembelauan Sinar-X (XRD), mekanik dinamik (DMA) dan haba mekanik (TMA) telah dijalankan untuk mengkaji struktur kristal, sifat haba dan mekanikal komposit. Pengukuran kekonduksian haba telah dijalankan untuk mengkaji kesan kepelbagaian peratus pengisi BeO pada nilai kekonduksian haba komposit epoksi. Nilai kekonduksian haba maksimum adalah 1.369

W/mK pada 50 % berat BeO dengan 14.36 % peningkatan daripada epoksi matrik. Peningkatan komposit epoksi yang diisi zarah BeO sebagai TIM untuk pakej LED kemudian dibincangkan dalam bentuk bacaan pengukuran rintangan haba fana (R_{th}) dan suhu pada persimpangan (T_J) serta parameter optik seperti Suhu Warna Berkolerasi (CCT) dan kecerahan cahaya. Pengimejan IR menunjukkan pengurangan suhu permukaan cip (T_s) pada setiap arus elektrik. Kajian ini menyimpulkan bahawa komposit yang diisi 50 % berat zarah BeO meningkatkan kadar pelepasan haba dan prestasi optik yang paling baik untuk pakej LED yang berkuasa tinggi dan dicadangkan sebagai bahan permukaan haba yang cekap dalam pengurusan haba di dalam aplikasi elektronik berkuasa tinggi.

EXPERIMENTAL AND CORRELATED STUDIES ON THERMAL CONDUCTIVITY OF BERYLLIUM OXIDE PARTICLES FILLED POLYMER AS THERMAL INTERFACE MATERIAL FOR DYNAMIC HEAT FLOW IN LEDs PACKAGE

ABSTRACT

BeO nanoparticles were synthesized by polyacrylamide gel route with varied calcination temperature; 800, 900 and 1000 ^{0}C. Structural characterization and surface morphology between synthesized BeO nanoparticles and commercial BeO microparticles were analyzed by X-ray diffraction analysis (XRD) and Scanning electron microscope analysis (SEM) in order to study the crystallinity and structural properties as well as surface morphology of BeO particles that would be beneficial for filler application. Thermal conductivity measurement analysis was carried out on both synthesized (800 ^{0}C) and commercial BeO particles filled epoxy matrix in order to figure out the effect of particle size on the thermal conductivity of epoxy composites. The structural properties and surface morphology that yielded the best crystallinity quality for BeO particles as well as the optimum size which enhanced the thermal conductivity of filled epoxy composites were observed at commercial BeO microparticles. BeO particles filled epoxy composite has been prepared with varied filler loading (10, 30 and 50 wt%) by mixing using the mechanical stirrer. Then, X-ray diffraction analysis (XRD), dynamic mechanical analysis (DMA) and Thermo-mechanical analysis (TMA) were carried out in order to study the crystallinity, thermal and mechanical properties of the composites. The maximum thermal conductivity value of 1.369 W/mK was obtained at 50 wt% BeO filled composite and

showed 14.36 % of improvement from neat epoxy. The improvement of BeO particles filled epoxy composites as TIMs for LED package is then discussed in the forms of measurement readings for thermal transient resistance (R_{th}) and temperature junction (T_J) as well as optical parameters such as Correlated Color Temperature (CCT) and luminous brightness. The IR imaging analysis indicated a lower chip surface temperature (T_s) at all driving currents. This study concludes that 50 wt% of BeO particles filled epoxy composites yields the optimum thermal and optical improvements of given high power LED package and would be suggested as thermally efficient interface materials in thermal management for high power electronic applications.

CHAPTER 1

INTRODUCTION

1.1 Overview

This chapter generally focuses on the brief introduction of light-emitting diodes (LEDs) and the issues that need to be handled well in the heat management of this device and how thermal interface material (TIMs) worked as a crucial component in order to obtain good heat dissipation in the LEDs package. Besides that, problem statement and research contributions are also discussed in this chapter as well as objective and the outline of the overall chapters.

1.2 Introduction

LEDs have attracted a great deal of attention from the lighting industry as it holds a promising characteristic of being versatile and environmental friendly (Chen, Lin et al. 2013). During the last decade, LEDs have been changing the world of lighting due to their efficiencies, size, and durability. LED has become one of the standard methods of producing lights. The energy efficiency of LED also has been improved a lot and make the technology growing faster in sectors of enabling new lighting systems (Anithambigai, Mutharasu et al. 2014). The LED technology has made a tremendous change in the worldwide lighting industry as the energy conversion efficiency in LED keeps increasing.

However, despite the rapid progress of this technology, there are some obstacles that need to be handled well such as heat management of this device as it absolutely affects the

LED lifetime and efficiency. For instance, the cooling solutions of an LED do not usually cater for the rapid increase of heat generated at the active region as it was only 20-30% of LED's electric power was converted into visible light and the rest will be wasted as heat (Park, Shin et al. 2006). Thus, it is essential for the heat generated at the device to be dissipated as effectively as possible. However, the interface problem always exists with the LED package. Thus, there will be an excessive rise in the junction temperature and followed by thermal runaway and catastrophic failures when this technology has shown poor performances in the thermal managements (Arik, Petroski et al. 2002).

Therefore, thermal interface materials (TIMs) as crucial components of advanced high-density electronic packaging are needed in the system in order to obtain a good heat dissipation. Thus, it will prevent the failure of electronic components due to overheating (Schelling, Shi et al. 2005). TIMs are used to filling the air gaps between the LED and the substrate. It would also be able to conform to the rough surfaces and possess a high thermal conductivity in order to provide an effective heat conduction path between the two solid surfaces (Xu, Munari et al. 2009). Hence, choosing the best material for the application of TIM is a very essential thing to do so that we could meet the stringent requirement for better thermal managements (Schelling, Shi et al. 2005).

1.3 Problem statement

Nowadays, despite the rapid progress of LED technology, there are so many major drawbacks happened especially in the thermal management of electronic packaging. The interface problem always exists with the LEDs package, the excessive rise in the thermal resistance and junction temperature would always cause thermal runaway and catastrophic failures. In-efficient thermal dissipation by TIM has always become a challenging problem with the existing polymer matrix. The enhancement of the thermal conducting properties of TIM applied in the system-level studies of LED would be vital.

- The enhancement of thermal conducting and material properties in term of size for filler application have not been addressed properly.

- The enhancement of mechanical and thermal properties of BeO filled polymer matrix as TIM at various filler loading have not been addressed properly.

- The challenging problem faces in heat dissipation in electronic packaging application as enhancement of thermal and material properties of TIM have not been addressed properly.

1.4 Objectives

- To study and compare the structural properties between synthesized BeO nanoparticles and commercial microparticles for filler applications.
- To fabricate the BeO particles filled polymer composites at various filler loading and analyses the material properties of the composites.
- To investigate the thermal and optical performances of LED package with BeO particles filled polymer composites as a TIM for effective heat dissipation in an electronic packaging application.

1.5 Novelties

- BeO filled polymer composite as thermal interfaces material is proposed, prepared, and optimized.
- Structural, mechanical and thermal properties of BeO filled polymer composites have been exploited in order to analyze the performance of proposed BeO filled TIM.
- Thermal and optical properties of LED attached to BeO filled epoxy composites as TIM in the package have been examined and the outcome data of the analysis could be used to develop BeO particles filled polymer matrix with higher heat transfer efficiency.

1.6 Thesis outline

i. **Chapter 1: Introduction**

This chapter generally focuses on the brief introduction of LEDs and current issues faced by the industries. Besides that, the objective and the outline of the overall chapter are discussed.

ii. **Chapter 2: Literature review**

This chapter generally focuses on the basic knowledge of thermal management, thermal interface material and thermal conductivity of particles filled polymer composites. In order to identify the optimum filler properties for polymer composites, understanding of the basic thermal management is needed and will be discussed here.

iii. **Chapter 3: Methodology**

In this chapter, the general methodology that is carried out for this experiment is presented here. The details of the methodology comprised of the synthesize of BeO particles by polyacrylamide gel route, preparation of BeO filled epoxy resin composite, structural characterization of BeO powder for the first part and the second one is for BeO filled epoxy resin in the form of a solid cube have been presented here. In addition, the procedure of thermal conductivity analysis of BeO filled epoxy resin by Transient Plane Source analysis, Infrared Imaging analysis, thermal and optical measurement of LED packages also have been explained here.

iv. **Chapter 4: Result and discussion**

This chapter extensively report the experimental outcome data collected from each analysis and has been presented in the form of tables and graphs. Discussions of the results have been stated in this chapter.

v. **Chapter 5: Conclusion and Recommendation**

Chapter 5 concludes the final conclusion obtained from the present work and proposed future recommendations for further development in the thermal management industries.

CHAPTER 2

LITERATURE REVIEW

2.1 Overview

This chapter generally focuses on the basic knowledge of the thermal management, thermal interface material and thermal conductivity of particles filled polymer composites. In order to identify the optimum filler for polymer composites, an understanding of the basic thermal management is needed and will be discussed here.

2.2 Brief Introduction on Light Emitting Diode

Light-emitting diodes (LEDs) are solid-state lighting sources which are also a kind of semiconductor device that is commonly used in indicator light, display board, communications, medical services, signage, general illumination and others (Poppe, Farkas et al. 2010). During the last decade, LEDs are changing the world of lighting due to their efficiencies, size, weight, durability, robustness, good temporal stability, fast switching times, lifetime cost and better impact resistance. It also holds a promising characteristic of being versatile and environmentally friendly with low ultra-violet (UV) radiation (Chen, Lin et al. 2013). The LED lighting is becoming more popular as incandescent lamps are being phased out globally. This is due to the capabilities of LEDs in transforming the electric energy into light energy directly in high efficiency and its life-span can reach as long as ten thousands hours to one hundred thousand hours. Besides that, only 20-30% of

LED’s electric power were converted into visible light, the rest will convert into a heat (Poppe, Farkas et al. 2010).

LEDs range from a narrow spectral band are available in a wide range of colors such as red, yellow, green, or blue to white, to the distribution of luminous intensity and variation types and shapes, depending on the mixing of the color and the design of the package. The colors of red/green/blue/orange/yellow phosphors with blue LED chips can be used to produce white light for White LED packages (Arik, Petroski et al. 2002). These days, due to the market share power, manufacturers are required to take each level of LED core technology process (LED die, LED packages, Lighting Module System) into consideration so that the development of the best final LED packages product can be produced. This is because of the high-quality product demands from the market share power which required product based on the optimal thermal dissipation, high external quantum efficiency, and high electrical power conversion efficiency. (Lasance 2014).

There are two types of LED that have been divided based on the input electrical power of the LED itself. The input power for low power LED packaging is generally lower than 0.1 Watt meanwhile larger than 1 W for high power LED packaging. High power LEDs have been promoted into the market with the development of blue light that can stimulate YAG phosphor to produce a white LED which makes the size and the input current of the LED (> 100 mA) to be bigger than usual (Poppe, Farkas et al. 2010). However, despite the rapid progress of this technology, the operating parameters of high power LEDs also keep increasing, which will cause thermal runaway and device failure if the heat management in the LED cannot be handled properly.

2.3 Thermal Management Backgrounds of LED

In the LED system, the thermal design of the device becoming the most essential things to be considered in LED production as LEDs become more powerful and applicable to many applications. The technological advancement of this industry becoming more rapid as it is highly dependent on efficient thermal management. As the development of this technology increases from time to time, there are still lots of challenges to overcome and improvements to be made, especially in the heat dissipation (Yue, Lu et al. 2011). Thus, it is important for us to ensure that the heat generated in the device to be dissipated as effectively and quickly as possible so that the true benefit of this technology is achieved. Moreover, with better thermal heat management, their performances and reliability can always be optimized.

The heat generated within the LED devices would affect the efficiency of the light generation process. Thus, the decrease in the brightness of the LEDs would make the device dimmer. Correspondingly, if the heat dissipated from junction to ambient ineffectively, the overheating device due to the accumulation of heat inside the dies will affect their chips and the whole performances (thermal, electrical and optical characteristics) as well as reliability of the devices (Huang and Chen 2011). Thus, it is important for us to manage the temperature of the LED properly so that it could be operated at an appropriate temperature range and control the temperature of all components in the devices. This will result in an efficient heat transfer within the semiconductor component package (LED) to the heat sink and finally released to the atmosphere as illustrated in Figure 2.1.

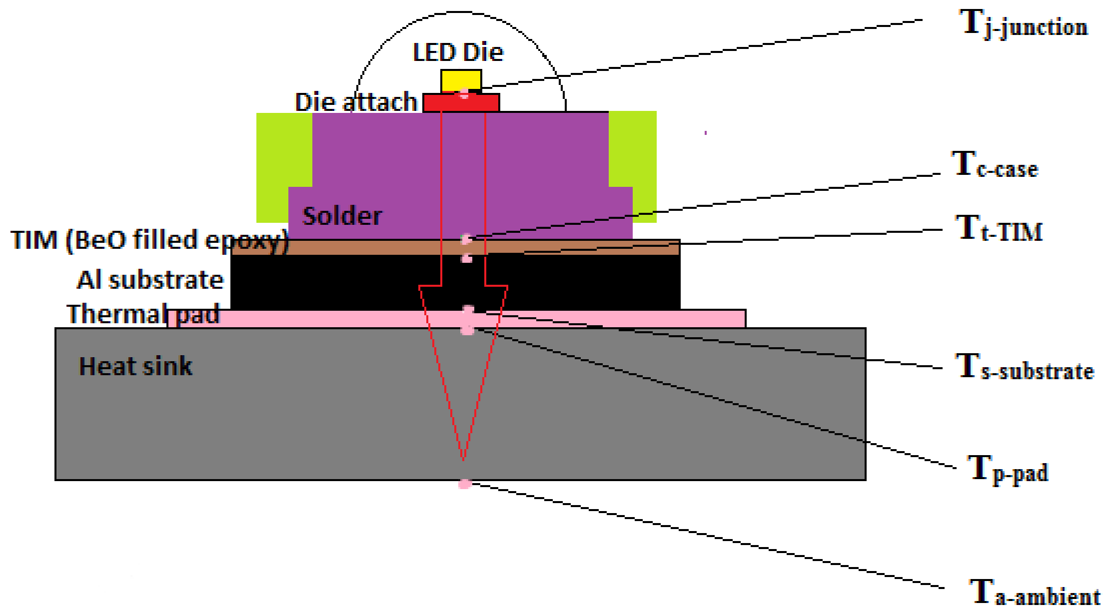

Figure 2.1 Schematic diagram of heat dissipation from junction to ambient

There are many factors that will affect the rise of the device temperature as heat flow from the junction to ambient (T_j- T_a) (see Figure 2.1). Some of the factors that influence the temperature rise in the device are due to the die itself, the thermal resistance of each contact point (R_{th}), choices of TIMs and the air gap between two surface contacts (Poppe, Farkas et al. 2010). All of this factor must be taken care of so that the heat generated within the LEDs can be efficiently removed to the atmosphere. Since the primary path for heat dissipation from the device to the atmosphere is through the conductive ways, the presence of the heat sink is one of the important elements to create an effective thermal conductive path (Park, Shin et al. 2006).

The two surfaces will be brought to contact as we attached the heat sink to the LEDs package. However, the interface problem exists when the two such surfaces are joined. The heat dissipation may occur only at the high points (metal-metal) of the contact surfaces

but air-filled gaps will be eventually created by the lower points (metal-air) as presented in Figure 2.2. There are more than 90% of air-filled in between the gaps which can be considered as a very poor conductor (Singh, Pal et al. 2010). It is already can be pointed out as the significant resistance to the heat flow even only with this problem. Therefore, TIMs are required to fill up the voids and maximize the contact surfaces between the heat source and the heat sink as well as minimize the thermal contact resistances between it at the same time (Mirjalili, Chuah et al. 2012). Thus, this will result in an increase in the thermal transfer efficiency of the LEDs package.

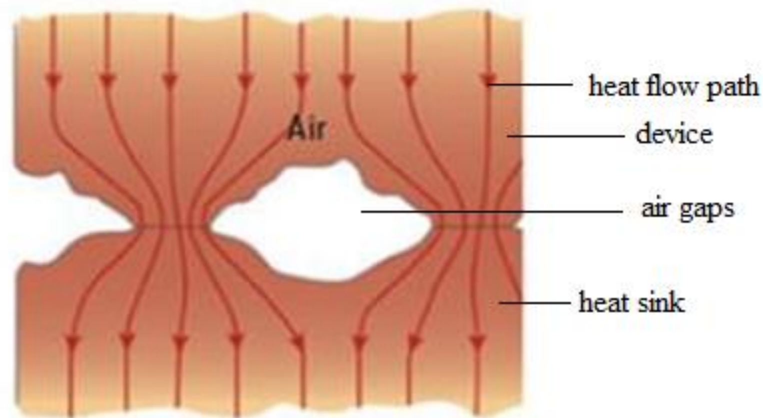

Figure 2.2 The air gaps in between surfaces in mechanical contact (Sarvar, Whalley et al. 2006)

2.4 Thermal Interfaces Material

An effective heat conducted from high power LEDs to the heat sink and then to the atmosphere will require an efficient thermal connection managed by proper thermal interface material (TIMs). However, the interface problem exists when the substrate and the heat sink attached to the LED's package. The void created when the two contact surfaces joined will be filled with air which is a poor conductor. Therefore, TIM as a crucial component of advanced high-density electronic packaging is required between the base-plate of LED and the cooling system. TIMs are needed in the system to eliminate those air gaps by conforming to the rough and uneven mating surfaces as well as minimize the thermal contact resistances (R_{th}) (Gwinn and Webb 2003) as shown in Figure 2.3.

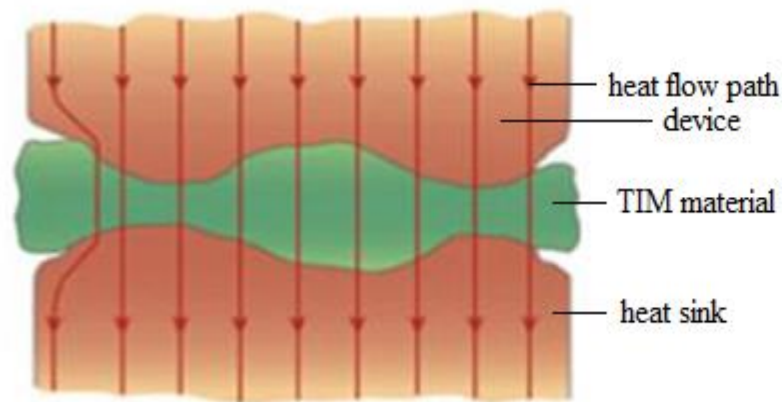

Figure 2.3 TIMs filled the air gaps between the two surfaces (Sarvar, Whalley et al. 2006)

The main bottlenecks in reducing the junction-to-ambient thermal resistances are to make sure an efficient heat dissipation from the TIMs to the heat sink. A preferable TIM should provide the highest possible thermal conductivity during the chip operation, good adhesion, and conformability to fill the gaps between the two rough surfaces (Tong 2011,

Chen, Ginzburg et al. 2016). As TIM has a higher thermal conductivity than air, the resistances across the joint will be decreased. Hence, it will enhance the efficiency of heat dissipation in the LEDs package. There are many different types of materials that have been created, developed and enhanced in response to the changing needs for the LEDs system. Some types of TIMs that have been applied in most of the high-power semiconductor devices especially in LEDs package are thermal conductive paste and greases, thermal gap filler pads, phase change materials, thermal epoxies, thermal tapes, gels and solders (Tong 2011).

One of the preferable TIM used in LEDs system is a thermal paste. It is always one of the thermally conductive compounds but usually electrically insulating (see Figure 2.4). Even though it is notoriously user-unfriendly as excess paste usually flows out past the edges but low thermal resistance (R_{th}) can be achieved and the thinnest possible thermal joint is formed between the two mating flat surfaces at their high points. Furthermore, it will also be able to conform to rough and uneven surfaces. Besides that, any thermal paste would be possible to be applied to any size and shape of the LEDs package and substrates. For this type of thermal paste, once it is cured, it will stay at the place and not be able to be removed anymore (Tong 2011).

Figure 2.4 Example of thermal paste that has been used in electronic devices (Kohli, Sobczak et al. 2001)

Therefore, this type of TIM usually possesses a very low thermal conductivity as most of the pastes used would be made from the polymer-based material. It is due to the typical thermal conductivity of amorphous polymer which has a lower thermal conductivity than those for metal and ceramic materials (Subramani and Devarajan 2013). Most of the polymers would have low thermal conductivity (0.1–0.5 W/mK), which is not sufficient for many applications that require high heat conduction (Nazempour 2012). However, polymers would also have many other advantages such as good process ability, light-weight material, low water absorption, high electrical resistivity, high voltage breakdown strength, corrosion resistance, and most importantly, it can be purchased in a low cost. Polymer-based thermally conductive materials are therefore often desired in many applications (Gwinn and Webb 2003).

Furthermore, one of the well-known polymer that has always been used in this application as a thermal paste/TIM is an epoxy resin. Epoxy resins are widely used as matrices for high-performance composite materials, surface coatings, and adhesive joints. It is due to its low density, ease of manufacturing capability and a large range of

applications as well as good mechanical properties, high impact resistance, good adhesion properties, high resistance to environmental degradation, and possesses good dielectric properties (Xu, Munari et al. 2009). However, it could not dissipates a huge amount of heat generated in devices as it possesses a low thermal conductivity at 0.19 W/mK (Jiao, Rak et al. 2000). To increase the thermal conductivity of the polymers, addition of high conductive material into the polymer matrix without affecting the physical nature of TIM is recommended. Their crystallinity, mechanical, structural and thermal properties could be also improved by the addition of high conductive fillers. Besides that, uncured epoxy resins have only poor mechanical, chemical and heat resistance properties. Good properties could be obtained by the curing process and curing may be achieved by forming a copolymer with hardeners. However, hardeners should be used in a sufficient amount to cure the epoxy composition (Al-Naser, Zhou et al. 2016).

Composite materials are made up of reinforcing material and matrix, and during the composite mixing process, the matrix material undergoes a series of the complex processes of physical, chemical and compounds changes with the reinforced material to form a whole body (Hong, Kim et al. 2017). Therefore, the matrix properties would directly affect the properties of composite materials and the choices of process parameters. The mechanical properties of the composites, undoubtedly depend mainly on the reinforcing material, but the role of the matrix cannot be ignored. Polymer matrix resin is one of the famous matrices that has been widely used for thermal application in a semiconductor device. Polymer Matrix Composite is a material consisting of a polymer (resin) matrix combined with a fibrous reinforcing dispersed phase. It was designed to transfer loads between the fibers through the matrix so that the load is uniformly distributed (Hussain, Hojjati et al. 2006).

Polymer Matrix Composites are very popular due to their essential advantages such as a sufficient dielectric property, a low cost material, facile process ability, high flexibility and possess light-weight material (Borah and Kim 2016). Recently, polymers reinforced with organic or inorganic particle fillers are becoming more common in applications that required highly conductive polymer. Hence, the thermal conductivity of polymers will be enhanced by the addition of particulate particles such as carbon-based fillers, metallic fillers, and ceramic fillers as their thermal conductivity are way much higher than the epoxy one (Ngo and Byon 2017).

2.5 Particulate Particles Filled Polymer Composites

Particulates fillers are another type of reinforcement in composite materials. Fillers are used in matrix resins for variety of reasons: cost reduction, improved processing, good optical effects, high thermal conductivity, good control of thermal expansion, good electrical properties and improved mechanical properties (Park and Seo 2011). Each filler type has different properties and these, in turn, are influenced by the particle size, shape, and surface chemistry. Filler loading is also one of the critical factor that should be consider in order to decide the good properties of composite materials. Up to date, most of the particulate filler that has been widely used in the thermal application is carbon-based fillers, metallic fillers and ceramic fillers (Wong and Bollampally 1999).

Graphite, carbon fiber and carbon black are well-known as traditional carbon-based fillers. Meanwhile, powders of aluminum, silver, copper, and nickel are commonly used as metallic particles (Rahaman, Khastgir et al. 2018). However, the filling of a polymer

with metallic particles may result in both the increase of thermal conductivity and electrical conductivity in the composites (Nazempour 2012). Lastly, ceramic powder reinforced polymer materials has become a trend and been used extensively as electronic materials as it offers good thermal conduction with low electrical conductivity. Being aware of the high electrical conductivity of metallic particles, several ceramic materials such as aluminum nitride (AlN), boron nitride (BN), silicon carbide (SiC), magnesium oxide (MgO) and beryllium oxide (BeO) gained more attention as thermally conductive fillers due to their high thermal conductivity and notable electrical insulator (Wilson, Paul et al. 2005). Table 2.1 present the thermal conductivity of all types of fillers.

Table 2.1 Thermal conductivities of some thermally conductive fillers (Donnet, Bansal et al. 1993, Pierson 1993, Nazempour 2012)

Material	**Thermal Conductivity at 25 °C, (W/m.K)**
Graphite	100 ~ 400 (on plane)
Carbon black	6 ~ 174
Carbon Nanotubes	2000 ~ 6000
Diamond	2000
PAN-based Carbon Fibre	8 ~ 70 (along the axis)
Pitch-based Carbon Fibre	530 ~1100 (along the axis)
Copper	483
Silver	450
Gold	345
Aluminum	204
Nickel	158
Boron Nitride	250 ~ 300
Aluminum nitride	200
Beryllium oxide	260
Aluminum oxide	20 ~ 29
Magnesium oxide	30
Zinc oxide	21

Among various materials (metals, carbon materials, and ceramics) that can be used as high thermal conductive materials, ceramics have been considered as ideal candidates for semiconductor devices owing to their electrical insulating nature, high thermal conductivity, and low thermal expansion coefficient (Hong, Kim et al. 2017). Besides that, metal-based and carbon-based materials also have superior electrical properties but due to their potential short circuits, which allow unintended electrical paths caused by the lack or

very low impedance of carbon or other electrically conductive material (Mamunya 2002), it limits the use of both metal and carbon composites for applications in highly semiconductor devices. It is because of the signal propagation or attenuation that could hardly be avoided from the both of material. Distinguished from others, polymer-based composites with ceramic fillers have drawn attention because of their low dielectric permittivity, high electrical resistivity, high thermal conductivity, low thermal expansion and notable electrical insulator (Yu 2002).

Among all the ceramic particles that have been stated above, beryllium oxide (BeO) is one of the best ceramic filler that could be embedded in polymer matrix due to its properties such as high thermal conductivity (260 W/m.K) (Xu, Munari et al. 2009), high melting point, high intensity, high insulation nature, high chemical and thermal stability, low dielectric constant, low dielectric loss and notable electrical insulator (Nazempour 2012). Thus, BeO particles have been chosen as the filler particles as its properties can be exploited more as there are not too many works and researches have been published especially in term of composites.

In addition, there are some reasons why we are using the bare polymer (epoxy resin) filled with ceramic particles (BeO) as a paste rather than commercially used paste. Firstly, most of the commercial paste (eg. silicon paste) is a metal-based composite as metallic filler such as silver (Ag) has been used in order to increase the thermal conductivity. Thus, the thermal conductivity of the paste is usually high but at the same time, it is also electrically conducting which is also in high potential of short circuits and signal propagation or attenuation. Thus, it does not suitable to be used as TIMs for dynamic heat flow in high-power semiconductor. Meanwhile, aside from the high quality of epoxy resin

that has been mentioned in pg.14 and 15, polymer-based composites could also be able to offer us an option in mixing process with the preferable filler particles such as ceramic filler particles like BeO. Thus, the paste will be possessed with high thermal conductivity but a notable electrical insulator. Besides that, there are novelties that could be found as the polymer-based material with ceramic fillers such as BeO particles filled composites would be further exploited for its material properties in terms of thermal, mechanical and structural as it is rarely used by other researchers in their works.

2.6 Properties of Beryllium Oxide Particles

Beryllium oxide is also known as beryllia, it is an inorganic compound with the formula BeO. This colorless solid is a notable electrical insulator with a higher thermal conductivity than any other non-metal except diamond and exceeds that of most metals (Greenwood and Earnshaw 2012). As an amorphous solid, beryllium oxide is white in color. Its high melting point leads to its uses as a refractory material. Among the oxides, beryllium oxide (BeO) is unique for oxides as it combines excellent electrical insulating properties (>1013 Ω.cm) with high thermal conductivity which is 260 W/(m.K) compared to thermal conductivity of MgO (30 W/m.K), AIN (220 W/m.K), and BN (30 W/m.K) (Nazempour 2012). It also possesses high chemical and thermal stability, good mechanical strength, high melting point, and high insulation nature, low dielectric constant and high corrosion-resistant. These properties may be exploited for a diverse range of applications for thermal management systems in integrated electronics. The properties of BeO particles have been stated in Table 2.2.

Table 2.2 Properties of BeO particles (Ropp 2012, Kim S 2019)

Properties	**Unit**
Chemical formula	BeO
Molar mass	25.011 $gmol^{-1}$
Density	3.01 g/cm^3
Melting point	2,507 °C (4,545 °F; 2,780 K)
Boiling point	3,900 °C (7,050 °F; 4,170 K)
Solubility in water	0.00002 g/100 mL
Thermal conductivity	330 W/m.K
Heat capacity (C)	25.5 J/mol.K
Bandgap	10.6 eV
Refractive index (n_D)	1.719
Appearance	Colourless
Odor	Odorless

No other oxide ceramic material collectively exhibits these highly desirable properties and characteristics. BeO ceramic conducts heat more quickly than virtually all metals with the exception of copper and silver. BeO also exhibits extremely low dielectric loss characteristics possess high electrical resistivity and offers excellent strength with high specific stiffness properties. Besides that, BeO provides all the desirable physical and dielectric characteristics exhibited by aluminum oxide or aluminum nitride. In addition, it offers thermal conductivity, *k* ten times higher than aluminum oxide (Al_2O_3) and 50 % higher than aluminum nitride (AlN). It also possesses a dielectric constant that is lower than both Al_2O_3 and AlN and weight per given volume that is fully a quarter less than Al_2O_3 and nearly 10 % less than AlN. As a thermally conductive and electrically insulating material, its performance is exceeded only by that of the diamond (C) (Donnet, Bansal et

al. 1993, Singh 2012). Furthermore, BeO has an outstanding combination of physical and chemical properties as stated in Table 2.3.

Table 2.3 Physical and chemical properties of BeO particles (Basu and Kalin 2011)

Physical and chemical properties of BeO particles
• reactivity with water vapor at high temperature (1000 °C).
• chemically stable oxides, resisting both carbon reduction and molten metal attack at high temperatures.
• thermal conductivity of BeO is extremely high (260 W/m.K) in comparison with other ceramics, particularly below 300 °C.
• widely applied in nuclear technology, vacuum electronics technology, microelectronics, and photoelectron technology especially in the thermal application.
• most often used as an electronic substrate because it gives an effective heat sink when its high thermal conductivity and good electrical resistivity were exploited.
• an electrical insulator with a bulk resistivity on the order of 1016 Ωcm.
• has relatively very good thermal shock resistance due to its high thermal conductivity.
• has a density fully 25 % lower than Alumina (3.01g/cm^3).
• Coefficient of Thermal Expansion (CTE) for BeO is comparable to many other oxide ceramics and many metals, as well, at approximately 8.9 x 10^{-6} K^{-1}.

Hence, beryllium oxide powder with high performances are necessaries to obtain advanced BeO particle ceramics. For the development of new products, synthesis and characterization of BeO particles are also one of the important process that should be taken seriously. One of the methods to synthesize BeO powder is the polyacrylamide gel route method. Before that, it should be informed that Beryllium Oxide particles are harmful to

our health as it is labeled as hazardous particles in dust form. Hence, there are cautions that should be followed properly in handling BeO particles throughout our experiment and the minimization of exposure of BeO particles throughout the entire experiment should be taken into account.

2.7 Hazard of Beryllium Oxide

BeO is carcinogenic in powdered form and may cause chronic beryllium disease. Once fired into solid form and as contained in finished products, it is safe to handle and presents no special health risks if not subjected to machining that generates dust. Most manufacturing operations, conducted properly on well-maintained equipment, are capable of safely processing BeO-containing materials. However, like many industrial materials, BeO may present a health risk if handled improperly. The inhalation of dust, mist or fume containing beryllium can cause a serious lung condition in some individuals. The degree of hazard varies, depending on the form of the product, how it is processed and handled, as well as the amount of BeO in the product as stated in commission regulation (EU 2008). One should read the product-specific Safety Data Sheet (SDS) for additional environmental, health and safety information before working with BeO.

Beryllium oxide can enter the body in three ways; eye or skin contact, ingestion or swallowing, and of most concern, by inhalation or breathing. However, there is no special health risk is associated with eye contact with beryllium oxide. As with any material processing operation, injury can result from particulate irritation or mechanical injury to the eyes from contact with dust or particles. Use proper protection, such as safety glasses

with side shields, goggles or face shields, to prevent eye injury. Besides that, through skin contact, particulate that becomes lodged under the skin has the potential to induce sensitization and skin lesions. A cut or laceration received from a sharp edge of beryllium oxide ceramic is no different from cuts received by ceramic materials and routine first aid treatment is appropriate. Cuts or lacerations must be thoroughly washed to remove all particulate debris from the wound (EU 2008).

Then, ingestion can occur when dust, mist or fume contacts hands, clothing, food and drinks which is followed by eating, drinking, smoking, and nail-biting. Thus, always practice good personal hygiene by not eating, drinking or smoking in manufacturing areas, and wash hands before doing so in designated areas. The last one is the most concern one which is through inhalation. People who are sensitive to inhaled beryllium particles can develop a serious and sometimes fatal lung disease called chronic beryllium disease ("CBD"). Chronic (Long Term) health effects may take months or years to develop. CBD is a condition in which the tissues of the lungs become inflamed, restricting the exchange of oxygen between the lungs and the bloodstream (Luz, Wu et al. 2018).

CBD does not occur in most people. However, it is not currently possible to tell who is potentially allergic and who is not. Therefore, all workers need to be protected and airborne beryllium particles must be controlled by Potential Health Effects from Exposure to Beryllium Oxide (Beryllia) SF300. Although there is no credible evidence that beryllium causes cancer in humans, cancer research is still continuing and Materion Ceramics Inc. recommends that caution should be maintained since beryllium oxide, like many other commonly used metal substances, has been listed by OSHA as a potential cancer hazard. Thus, processing beryllium oxide shall be conducted in accordance with the Beryllium